Policy Assessment for the Review of the Secondary National Ambient Air Quality Standards for Oxides of Nitrogen, Oxides of Sulfur and Particulate Matter

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Policy Assessment for the Review of the Secondary National Ambient Air Quality Standards for Oxides of Nitrogen, Oxides of Sulfur and Particulate Matter

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1 INTRODUCTION

This document, *Policy Assessment for the Review of the Secondary National Ambient Air Quality Standards for Oxides of Nitrogen, Oxides of Sulfur and Particulate Matter* (hereafter referred to as PA), presents the policy assessment for the U.S. Environmental Protection Agency's (EPA's) current review of the secondary national ambient air quality standards (NAAQS) for oxides of nitrogen (N oxides), oxides of sulfur (SO_X), and particulate matter (PM).[1] This review differs from the review of the secondary standards for oxides of nitrogen and sulfur completed in 2012 in that the current review includes consideration of the secondary PM standards, in addition to the secondary standards for oxides of nitrogen and sulfur. Given the contribution of nitrogen compounds to PM, including but not limited to those related to N oxides, the current review provides for an expanded and more integrated consideration of N deposition and the current related air quality information. Regarding PM, welfare effects associated with visibility impairment, climate effects, and materials effects (i.e., damage and soiling) are being addressed in the separate review of the NAAQS for PM. In the context of the secondary standards for oxides of nitrogen,[2] oxides of sulfur and PM, the scope pertains to the protection of the public welfare from adverse effects related to ecological effects.[3]

This PA, prepared by staff of the EPA's Office of Air Quality Planning and Standards,[4] considers key policy-relevant issues, drawing on those identified in the *Integrated Review Plan for the Secondary National Ambient Air Quality Standards for Ecological Effects of Oxides of Nitrogen, Oxides of Sulfur and Particulate Matter* (IRP; U.S. EPA, 2017) and the *Integrated Science Assessment for Oxides of Nitrogen, Oxides of Sulfur and Particulate Matter – Ecological*

[1] This review focuses on the presence in ambient air of oxides of nitrogen, oxides of sulfur, and particulate matter. The standards that are the focus of this review are the secondary standards for NO_2, set in 1971 (36 FR 8186, April 30, 1971), for SO_2, set in 1971 (36 FR 8186, April 30, 1971), for PM_{10}, set in 2012 (78 FR 3085, January 15, 2013), and for $PM_{2.5}$, set in 2012 (78 FR 3085, January 15, 2013). These standards are referred to in this document as the "current" or "existing" standards.

[2] In this document, the term, oxides of nitrogen, refers to all forms of oxidized nitrogen (N) compounds, including NO, NO_2, and all other oxidized N-containing compounds formed from NO and NO_2. This follows usages in the Clean Air Act section 108(c): "Such criteria [for oxides of nitrogen] shall include a discussion of nitric and nitrous acids, nitrites, nitrates, nitrosamines, and other carcinogenic and potentially carcinogenic derivatives of oxides of nitrogen." By contrast, within much of the air pollution research and control communities, the terms "oxides of nitrogen" and "nitrogen oxides" are restricted to refer only to the sum of NO and NO_2, and this sum is commonly abbreviated NO_X. Where used in this document (e.g., Chapter 2), the definition used is provided.

[3] Welfare effects of PM other than ecological effects, such as visibility effects and materials damage, were addressed in the separate PM NAAQS review completed in 2020 and are part of the reconsideration of that 2020 decision, a proposed decision for which was published early in 2023 (88 FR 5558, January 27, 2023).

[4] The terms "staff," "we," and "our" throughout this document refer to the staff in the EPA's Office of Air Quality Planning and Standards (OAQPS).

Criteria (ISA or 2020 ISA; U.S. EPA, 2020).This document is organized into seven chapters, encompassing information on air quality, the nature of effects and exposure conditions associated with effects, relationships between deposition and air quality metrics, and a review of the standards. A detailed description of chapters within this document (and associated appendices) is provided in section 1.5 below. In this introductory chapter, we present information on the purpose of the PA (section 1.1), legislative requirements for reviews of the NAAQS (section 1.2), and an overview of the history of the N oxides, SO_X, and PM NAAQS reviews (section 1.3). Section 1.4 describes progress and next steps in the current review.

1.1 PURPOSE

The PA, when final, presents an evaluation, for consideration by the EPA Administrator, of the policy implications of the currently available scientific information, assessed in the ISA, any quantitative air quality, exposure or risk analyses based on the ISA findings, and related limitations and uncertainties. Ultimately, final decisions on the secondary N oxides, SO_X, and PM NAAQS will reflect the judgments of the Administrator. The role of the PA is to help "bridge the gap" between the Agency's scientific assessment and quantitative technical analyses, and the judgments required of the Administrator in determining whether it is appropriate to retain or revise the NAAQS.

In evaluating the question of adequacy of the current standards and whether it may be appropriate to consider alternative standards, the PA focuses on information that is most pertinent to evaluating the standards and their basic elements: indicator, averaging time, form, and level.[5] These elements, which together serve to define each standard, must be considered collectively in evaluating the public health and public welfare protection the standards afford.

The development of the PA is also intended to facilitate advice to the Agency and recommendations to the Administrator from an independent scientific review committee, the Clean Air Scientific Advisory Committee (CASAC), as provided for in the Clean Air Act (CAA). As discussed below in section 1.2, the CASAC is to advise on subjects including the Agency's assessment of the relevant scientific information and on the adequacy of the current standards, and to make recommendations as to any revisions of the standards that may be

[5] The indicator defines the chemical species or mixture to be measured in the ambient air for the purpose of determining whether an area attains the standard. The averaging time defines the period over which air quality measurements are to be averaged or otherwise analyzed. The form of a standard defines the air quality statistic that is to be compared to the level of the standard in determining whether an area attains the standard. For example, the form of the annual NAAQS for fine particulate matter ($PM_{2.5}$) is the average of annual mean concentrations for three consecutive years, while the form of the 3-hour secondary NAAQS for SO_2 is the second-highest 3-hour average in a year. The level of the standard defines the air quality concentration used for that purpose.

appropriate. The EPA generally makes available to the CASAC and the public one or more drafts of the PA for CASAC review and public comment.

In this PA, we consider the available scientific information, as assessed in the *Integrated Science Assessment for Oxides of Nitrogen, Oxides of Sulfur and Particulate Matter – Ecological Criteria*, (ISA [U.S. EPA, 2020]) which included literature through May 2017, and additional policy-relevant quantitative air quality, exposure and risk analyses. Advice and comments from the CASAC and the public on the PA has informed the evaluation and conclusions in this final PA.

The PA is designed to assist the Administrator in considering the currently available scientific evidence and quantitative air quality, exposure and risk information, and in formulating judgments regarding the standards. The final PA will inform the Administrator's decision in this review. Beyond informing the Administrator and facilitating the advice and recommendations of the CASAC, the PA is also intended to be a useful reference to all interested parties. In these roles, it is intended to serve as a source of policy-relevant information that supports the Agency's review of the secondary NAAQS for N oxides, SO_X, and PM, and it is written to be understandable to a broad audience.

1.2 LEGISLATIVE REQUIREMENTS

Two sections of the CAA govern the establishment and revision of the NAAQS. Section 108 (42 U.S.C. 7408) directs the Administrator to identify and list certain air pollutants and then to issue air quality criteria for those pollutants. The Administrator is to list those pollutants "emissions of which, in his judgment, cause or contribute to air pollution which may reasonably be anticipated to endanger public health or welfare"; "the presence of which in the ambient air results from numerous or diverse mobile or stationary sources"; and for which he "plans to issue air quality criteria…." (42 U.S.C. § 7408(a)(1)). Air quality criteria are intended to "accurately reflect the latest scientific knowledge useful in indicating the kind and extent of all identifiable effects on public health or welfare which may be expected from the presence of [a] pollutant in the ambient air…." 42 U.S.C. § 7408(a)(2).

Section 109 (42 U.S.C. 7409) directs the Administrator to propose and promulgate "primary" and "secondary" NAAQS for pollutants for which air quality criteria are issued (42 U.S.C. § 7409(a)). Under section 109(b)(2), a secondary standard must "specify a level of air quality the attainment and maintenance of which, in the judgment of the Administrator, based on

such criteria, is requisite to protect the public welfare from any known or anticipated adverse effects associated with the presence of [the] pollutant in the ambient air."[6]

In setting primary and secondary standards that are "requisite" to protect public health and welfare, respectively, as provided in section 109(b), the EPA's task is to establish standards that are neither more nor less stringent than necessary. In so doing, the EPA may not consider the costs of implementing the standards. See generally, *Whitman v. American Trucking Ass'ns*, 531 U.S. 457, 465-472, 475-76 (2001). Likewise, "[a]ttainability and technological feasibility are not relevant considerations in the promulgation of national ambient air quality standards" (*American Petroleum Institute v. Costle*, 665 F.2d 1176, 1185 (D.C. Cir. 1981)). However, courts have clarified that in deciding how to revise the NAAQS in the context of considering standard levels within the range of reasonable values supported by the air quality criteria and judgments of the Administrator, EPA may consider "relative proximity to peak background … concentrations" as a factor (*American Trucking Ass'ns, v. EPA*, 283 F.3d 355, 379 (D.C. Cir. 2002)).

Section 109(d)(1) of the Act requires periodic review and, if appropriate, revision of existing air quality criteria to reflect advances in scientific knowledge on the effects of the pollutant on public health and welfare. Under the same provision, the EPA is also to periodically review and, if appropriate, revise the NAAQS, based on the revised air quality criteria.[7]

Section 109(d)(2) addresses the appointment and advisory functions of an independent scientific review committee. Section 109(d)(2)(A) requires the Administrator to appoint this committee, which is to be composed of "seven members including at least one member of the National Academy of Sciences, one physician, and one person representing State air pollution control agencies." Section 109(d)(2)(B) provides that the independent scientific review committee "shall complete a review of the criteria…and the national primary and secondary ambient air quality standards…and shall recommend to the Administrator any new…standards and revisions of existing criteria and standards as may be appropriate…." Since the early 1980s, this independent review function has been performed by the CASAC of the EPA's Science Advisory Board.

Section 109(b)(2) specifies that "[a]ny national secondary ambient air quality standard prescribed under subsection (a) shall specify a level of air quality the attainment and maintenance of which in the judgment of the Administrator, based on such criteria, is requisite to

[6] Under CAA section 302(h) (42 U.S.C. § 7602(h)), effects on welfare include, but are not limited to, "effects on soils, water, crops, vegetation, manmade materials, animals, wildlife, weather, visibility, and climate, damage to and deterioration of property, and hazards to transportation, as well as effects on economic values and on personal comfort and well-being."

[7] This section of the Act requires the Administrator to complete these reviews and make any revisions that may be appropriate "at five-year intervals."

protect the public welfare from any known or anticipated adverse effects associated with the presence of such air pollutant in the ambient air." Consistent with this statutory direction, EPA has always understood the goal of the NAAQS is to identify a requisite level of air quality, and the means of achieving a specific level of air quality is to set a standard expressed as a concentration of a pollutant in the air, such as in terms of parts per million (ppm), parts per billion (ppb), or micrograms per cubic meter ($\mu g/m^3$). Thus, while deposition-related effects are included within the "adverse effects associated with the presence of such air pollutant in the ambient air," EPA has never found a standard that quantifies atmospheric deposition onto surfaces to constitute a national secondary ambient air quality standard.

1.3 BACKGROUND ON CRITERIA AND SECONDARY STANDARDS FOR NITROGEN OXIDES AND SULFUR OXIDES AND PARTICULATE MATTER

Secondary NAAQS were first established for oxides of nitrogen, oxides of sulfur and particulate matter in 1971 (36 FR 8186, April 30, 1971). Since that time, the EPA has periodically reviewed the air quality criteria and secondary standards for these pollutants, with the most recent reviews that considered the evidence for ecological effects of these pollutants being completed in 2012 and 2013 (77 FR 20218, April 3, 2012; 78 FR 3086, January 15, 2013). The subsections below summarize key proceedings from the initial standard setting in 1971 to the last reviews in 2012-2013. Key aspects of the scientific evidence supporting the standards is summarized in sections 3.1 and 3.2 below.

1.3.1 Nitrogen Oxides

The EPA first promulgated NAAQS for oxides of N in April 1971 after reviewing the relevant science on the public health and welfare effects in the 1971 *Air Quality Criteria for Nitrogen Oxides* (air quality criteria document or AQCD).[8] With regard to welfare effects, the 1971 AQCD described effects of NO_2 on vegetation and corrosion of electrical components linked to particulate nitrate (U.S. EPA, 1971). The primary and secondary standards were both set at 0.053 ppm NO_2 as an annual average (36 FR 8186, April 30, 1971). In 1982, the EPA published an updated AQCD (U.S. EPA, 1982a). Based on the 1982 AQCD, the EPA proposed to retain the existing standards in February 1984 (49 FR 6866, February 23, 1984). After considering public comments, the EPA published the final decision to retain these standards in June 1985 (50 FR 25532, June 19, 1985).

[8] In reviews initiated prior to 2007, the AQCD provided the scientific foundation (i.e., the air quality criteria) for the NAAQS. Since that time, the Integrated Science Assessment (ISA) has replaced the AQCD.

The EPA began a second review of the primary and secondary standards for oxides of nitrogen in 1987 (52 FR 27580, July 22, 1987). In November 1991, the EPA released an updated draft AQCD for CASAC and public review and comment (56 FR 59285, November 25, 1991). The CASAC reviewed the draft document at a meeting held on July 1, 1993, and concluded in a closure letter to the Administrator that the document provided "an adequate basis" for EPA's decision-making in the review (Wolff, 1993). The final AQCD was released later in 1993 (U.S. EPA, 1993). Based on the 1993 AQCD, the EPA's Office of Air Quality Planning and Standards (OAQPS) prepared a Staff Paper,[9] drafts of which were reviewed by the CASAC (Wolff, 1995; U.S. EPA, 1995a). In October 1995, the EPA proposed not to revise the secondary NO_2 NAAQS (60 FR 52874; October 11, 1995). After consideration of the comments received on the proposal, the Administrator decided not to revise the NO_2 NAAQS (61 FR 52852; October 8, 1996). The subsequent (and most recent) review of the N oxides secondary standard was a joint review with the secondary standard for SO_X, which was completed in 2012 (see section 1.3.4 below).

1.3.2 Sulfur Oxides

The EPA first promulgated secondary NAAQS for sulfur oxides in April 1971 based on the scientific evidence evaluated in the 1969 AQCD (U.S. DHEW, 1969a [1969 AQCD]; 36 FR 8186, April 30, 1971). These standards, which were established on the basis of evidence of adverse effects on vegetation, included an annual arithmetic mean standard, set at 0.02 ppm SO_2,[10] and a 3- hour average standard set at 0.5 ppm SO_2, not to be exceeded more than once per year. In 1973, based on information indicating there to be insufficient data to support the finding of a study in the 1969 AQCD concerning vegetation injury associated from SO_2 exposure over the growing season, rather than from short-term peak concentrations, the EPA proposed to revoke the annual mean secondary standard (38 FR 11355, May 7, 1973). Based on consideration of public comments and external scientific review, the EPA released a revised chapter of the AQCD and published its final decision to revoke the annual mean secondary standard (U.S. EPA, 1973; 38 FR 25678, September 14, 1973). At that time, the EPA additionally noted that injury to vegetation was the only type of SO_2 welfare effect for which the evidence base supported a quantitative relationship, stating that although data were not available

[9] Prior to reviews initiated in 2007, the Staff Paper summarized and integrated key studies and the scientific evidence, and from the 1990s onward also assessed potential exposures and associated risk. The Staff paper also presented the EPA staff's considerations and conclusions regarding the adequacy of existing NAAQS and, when appropriate, the potential alternative standards that could be supported by the evidence and information. More recent reviews present this information in the Policy Assessment.

[10] Established with the annual standard as a guide to be used in assessing implementation plans to achieve the annual standard was a maximum 24-hour average concentration not to be exceeded more than once per year (36 FR 8187, April 30, 1971).

at that time to establish a quantitative relationship between SO_2 concentrations and other public welfare effects, including effects on materials, visibility, soils, and water, the SO_2 primary standards and the 3-hour secondary standard may to some extent mitigate such effects. The EPA also stated it was not clear that any such effects, if occurring below the current standards, are adverse to the public welfare (38 FR 25679, September 14, 1973).

In 1979, the EPA announced initiation of a concurrent review of the air quality criteria for oxides of sulfur and PM and plans for development of a combined AQCD for these pollutants (44 FR 56730, October 2, 1979). The EPA subsequently released three drafts of a combined AQCD for CASAC review and public comment. In these reviews, and guidance provided at the CASAC August 20-22, 1980 public meeting on the first draft AQCD, the CASAC concluded that acidic deposition was a topic of extreme scientific complexity because of the difficulty in establishing firm quantitative relationships among emissions of relevant pollutants, formation of acidic wet and dry deposition products, and effects on terrestrial and aquatic ecosystems (53 FR 14935, April 26, 1988). The CASAC also noted that a fundamental problem of addressing acid deposition in a criteria document is that acid deposition is produced by several different criteria pollutants: oxides of sulfur, oxides of nitrogen, and the fine particulate fraction of suspended particles (U.S. EPA, 1982b, pp. 125-126). The CASAC also felt that any document on this subject should address both wet and dry deposition, since dry deposition was believed to account for a substantial portion of the total acid deposition problem (53 FR 14936, April 26, 1988; Lippman, 1987). For these reasons, CASAC recommended that, in addition to including a summary discussion of acid deposition in the final AQCD, a separate, comprehensive document on acid deposition be prepared prior to any consideration of using the NAAQS as a regulatory mechanism for the control of acid deposition.

Following CASAC closure on the AQCD for oxides of sulfur in December 1981, the EPA released a final AQCD (U.S. EPA, 1982b), and the EPA's OAQPS prepared a Staff Paper that was released in November 1982 (U.S. EPA, 1982c). The issue of acidic deposition was not, however, assessed directly in the OAQPS staff paper because the EPA followed the guidance given by the CASAC, subsequently preparing the following documents to address acid deposition: *The Acidic Deposition Phenomenon and Its Effects: Critical Assessment Review Papers, Volumes I and II* (U.S. EPA, 1984a, b) and *The Acidic Deposition Phenomenon and Its Effects: Critical Assessment Document* (U.S. EPA, 1985) (53 FR 14935 -14936, April 26, 1988). Although these documents were not considered criteria documents and had not undergone CASAC review, they represented the most comprehensive summary of scientific information relevant to acid deposition completed by the EPA at that point.

In April 1988, the EPA proposed not to revise the existing secondary standards for SO_2 (53 FR 14926, April 26, 1988). This proposed decision with regard to the secondary SO_2

NAAQS was due to the Administrator's conclusions that (1) based upon the then-current scientific understanding of the acid deposition problem, it would be premature and unwise to prescribe any regulatory control program at that time and (2) when the fundamental scientific uncertainties had been decreased through ongoing research efforts, the EPA would draft and support an appropriate set of control measures (53 FR 14926, April 26, 1988). This review of the secondary standard for SO_X was concluded in 1993, subsequent to the Clean Air Act Amendments of 1990 (see section 1.3.3 below). The EPA decided not to revise the secondary standard, concluding that revisions to the standard to address acidic deposition and related SO_2 welfare effects was not appropriate at that time (58 FR 21351, April 21, 1993). In describing the decision, the EPA recognized the significant reductions in SO_2 emissions, ambient air SO_2 concentrations and ultimately deposition expected to result from implementation of the Title IV program, which was expected to significantly decrease the acidification of water bodies and damage to forest ecosystems and to permit much of the existing damage to be reversed with time (58 FR 21357, April 21, 1993). While recognizing that further action might be needed to address acidic deposition in the longer term, the EPA judged it prudent to await the results of the studies and research programs then underway, including those assessing the comparative merits of secondary standards, acidic deposition standards and other approaches to controlling acidic deposition and related effects, and then to determine whether additional control measures should be adopted or recommended to Congress (58 FR 21358, April 21, 1993).

1.3.3 Related Actions Addressing Acid Deposition

In 1980, Congress created the National Acid Precipitation Assessment Program (NAPAP). During the 10-year course of this program, a series of reports were issued and a final report was issued in 1990 (NAPAP, 1991). On November 15, 1990, Amendments to the CAA were passed by Congress and signed into law by the President. In Title IV of these Amendments, Congress included a statement of findings including the following: "1) the presence of acidic compounds and their precursors in the atmosphere and in deposition from the atmosphere represents a threat to natural resources, ecosystems, materials, visibility, and public health; … 3) the problem of acid deposition is of national and international significance; … 5) current and future generations of Americans will be adversely affected by delaying measures to remedy the problem…". The goal of Title IV was to reduce emissions of SO_2 by 10 million tons and N oxides emissions by 2 million tons from 1980 emission levels in order to achieve reductions over broad geographic regions/areas. In envisioning that further action might be necessary in the long term, Congress included section 404 of the 1990 Amendments. This section requires the EPA to conduct a study on the feasibility and effectiveness of an acid deposition standard or standards to protect "sensitive and critically sensitive aquatic and terrestrial resources" and at the conclusion

of the study, submit a report to Congress. Five years later the EPA submitted to Congress its report titled *Acid Deposition Standard Feasibility Study: Report to Congress* (U.S. EPA, 1995b) in fulfillment of this requirement. The Report to Congress concluded that establishing acid deposition standards for sulfur and nitrogen deposition might at some point in the future be technically feasible although appropriate deposition loads for these acidifying chemicals could not be defined with reasonable certainty at that time.

The 1990 Amendments also added new language to sections of the CAA pertaining to ecosystem effects of criteria pollutants, such as acid deposition. For example, a new section 108(g) was inserted, stating that "[t]he Administrator may assess the risks to ecosystems from exposure to criteria air pollutants (as identified by the Administrator in the Administrator's sole discretion)." The definition of welfare in section 302(h) was expanded to indicate that welfare effects include those listed therein, "whether caused by transformation, conversion, or combination with other air pollutants." Additionally, in response to legislative initiatives such as the 1990 Amendments, the EPA and other Federal agencies continued research on the causes and effects of acidic deposition and related welfare effects of SO_2 and implemented an enhanced monitoring program to track progress (58 FR 21357, April 21, 1993).

1.3.4 Most Recent Review of the Secondary Standards for Oxides of Nitrogen and Oxides of Sulfur

In December 2005, the EPA initiated a joint review[11] of the air quality criteria for oxides of nitrogen and sulfur and the secondary NAAQS for NO_2 and SO_2 (70 FR 73236, December 9, 2005).[12] The review focused on the evaluation of the protection provided by the secondary standards for oxides of nitrogen and oxides of sulfur for two general types of effects: (1) direct effects on vegetation of exposure to gaseous oxides of nitrogen and sulfur, which are the type of effects that the existing NO_2 and SO_2 secondary standards were developed to protect against, and (2) effects associated with the deposition of oxides of nitrogen and sulfur to sensitive aquatic and terrestrial ecosystems (77 FR 20218, April 3, 2012).

[11] Although the EPA has historically adopted separate secondary standards for oxides of nitrogen and oxides of sulfur, the EPA conducted a joint review of these standards because oxides of nitrogen and sulfur and their associated transformation products are linked from an atmospheric chemistry perspective, as well as from an environmental effects perspective. The joint review was also responsive to the National Research Council (NRC) recommendation for the EPA to consider multiple pollutants, as appropriate, in forming the scientific basis for the NAAQS (NRC, 2004).

[12] The review was conducted under a schedule specified by consent decree entered into by the EPA with the Center for Biological Diversity and four other plaintiffs. The schedule, which was revised on October 22, 2009 provided that the EPA sign notices of proposed and final rulemaking concerning its review of the oxides of nitrogen and oxides of sulfur NAAQS no later than July 12, 2011 and March 20, 2012, respectively.

The Integrated Review Plan (IRP) for the review was released in December 2007, after review of a draft IRP by the public and CASAC (72 FR 57570, October 10, 2007; Russell, 2007; U.S. EPA, 2007). The first and second drafts of the ISA were released in December 2007 and August 2008, respectively, for the CASAC and public review (72 FR 72719, December 21, 2007; 73 FR 10243, February 26, 2008; Russell and Henderson, 2008; 73 FR 46908, August 12, 2008; 73 FR 53242, September 15, 2008; Russell and Samet, 2008a). The final ISA was released in December 2008 (73 FR 75716, December 12, 2008; U.S. EPA, 2008a [2008 ISA]). Based on the scientific information in the ISA, the EPA planned and developed a quantitative Risk and Exposure Assessment (REA), two drafts of which were made available for public comment and reviewed by the CASAC (73 FR 10243, February 26, 2008; 73 FR 50965, August 29, 2008; Russell and Samet, 2008b; 73 FR 53242, September 15, 2008; 74 FR 28698, June 17, 2009; Russell and Samet, 2009). The final REA was released in September 2009 (U.S. EPA, 2009a; 74 FR 48543; September 23, 2009).

Drawing on the information in the final REA and ISA, the EPA OAQPS prepared a PA, two drafts of which were made available for public comment and review by the CASAC (75 FR 10479, March 8, 2010; 75 FR 11877, March 12, 2010; Russell and Samet, 2010b; 75 FR 57463, September 21, 2010; 75 FR 65480, October 25, 2010; Russell and Samet, 2010a). The final PA was released in January 2011 (U.S. EPA, 2011). Based on additional discussion subsequent to release of the final PA, the CASAC provided additional advice and recommendations on the multipollutant, deposition-based standard described in the PA (76 FR 4109, January 24, 2011; 76 FR 16768, March 25, 2011; Russell and Samet, 2011).

For the purpose of protection against the direct effects on vegetation of exposure to gaseous oxides of nitrogen and sulfur, the PA concluded that consideration should be given to retaining the current standards. With respect to the effects associated with the deposition of oxides of nitrogen and oxides of sulfur to sensitive aquatic and terrestrial ecosystems, the PA focused on the acidifying effects of nitrogen and sulfur deposition on sensitive aquatic ecosystems. Based on the information in the ISA, the assessments in the REA, and the CASAC advice, the PA concluded that consideration be given to a new multipollutant standard intended to address deposition-related effects, as described in section 3.2 below.

On August 1, 2011, the EPA published a proposed decision to retain the existing annual average NO_2 and 3-hour average SO_2 secondary standards, recognizing the protection they provided from direct effects on vegetation (76 FR 46084, August 1, 2011). Further, after considering the multipollutant approach to a standard developed in the PA, the Administrator proposed not to set such a new multipollutant secondary standard in light of a number of uncertainties (summarized in section 3.2 below). Additionally, the Administrator proposed to revise the secondary standards by adding secondary standards identical to the NO_2 and SO_2

primary 1-hour standards set in 2010, noting that these new standards[13] would result in reductions in oxides of nitrogen and sulfur that would likely reduce nitrogen and sulfur deposition to sensitive ecosystems (76 FR 46084, August 1, 2011). After consideration of public comments, the final decision in the review was to retain the existing standards to address the direct effects on vegetation of exposure to gaseous oxides of nitrogen and sulfur and also, to not set additional standards particular to effects associated with deposition of oxides of nitrogen and sulfur on sensitive aquatic and terrestrial ecosystems at that time (77 FR 20218, April 3, 2012). Technical aspects of the approach described in the 2011 PA and the Administrator's decision-making are summarized in section 3.2 below.

The EPA's 2012 decision was challenged by the Center for Biological Diversity and other environmental groups. The petitioners argued that having decided that the existing standards were not adequate to protect against adverse public welfare effects such as damage to sensitive ecosystems, the Administrator was required to identify the requisite level of protection for the public welfare and to issue a NAAQS to achieve and maintain that level of protection. The D.C. Circuit disagreed, finding that the EPA acted appropriately in not setting a secondary standard given the EPA's conclusions that "the available information was insufficient to permit a reasoned judgment about whether any proposed standard would be 'requisite to protect the public welfare . . . '."[14] In reaching this decision, the court noted that the EPA had "explained in great detail" the profound uncertainties associated with setting a secondary NAAQS to protect against aquatic acidification.[15]

1.3.5 Particulate Matter

The EPA first established a secondary standard for PM in 1971 (36 FR 8186, April 30, 1971), based on the original AQCD, which described the evidence as to effects of PM on visibility, materials, light absorption and vegetation (U.S. DHEW, 1969b). To provide protection generally from visibility effects and materials damage, the secondary standard was set at 150 $\mu g/m^3$, as a 24-hour average, from total suspended particles (TSP), not to be exceeded more than once per year (36 FR 8187; April 30, 1971).[16]

In October 1979, the EPA announced the first periodic review of the air quality criteria and NAAQS for PM (44 FR 56730, October 2, 1979). As summarized in section 1.3.2 above, the

[13] The 2010 primary 1-hour standards included the NO_2 standard set at a level of 100 ppb and the SO_2 standard set at a level of 75 ppb.

[14] Center for Biological Diversity, et al. v. EPA, 749 F.3d 1079, 1087 (2014).

[15] Id. at 1088.

[16] Additionally, a guide to be used in assessing implementation plans in assessing implementation plans to achieve the 24-hour standard was set at 60 µg/m3, as an annual geometric mean (36 FR 8187; April 30, 1971).

EPA developed a new AQCD for PM and SO_X, drafts of which were reviewed by the CASAC (U.S. EPA, 1982b). Subsequently, the EPA OAQPS developed a Staff Paper (U.S. EPA, 1982d), two drafts of which were reviewed by the CASAC (Friedlander, 1982). Further, the EPA OAQPS prepared an Addendum to the 1982 staff paper, which also received CASAC (Lippman, 1986; U.S. EPA, 1986). After consideration of public comments on a proposed decision, the final decision in this review revised the indicator for PM NAAQS from TSP to particulate matter with mass median diameter of 10 microns (PM_{10}) (49 FR 10408, March 20, 1984; 52 FR 24634, July 1, 1987). With an indicator of PM_{10}, two secondary standards were established to be the same as the primary standards. A 24-hour secondary standard was set at 150 $\mu g/m^3$, with the form was one expected exceedance per year, on average over three years. Additionally, an annual secondary standard was set at 50 $\mu g/m^3$, with a form of annual arithmetic mean, averaged over three years (52 FR 24634, July 1, 1987).

In April 1994, the EPA initiated the second periodic review of the air quality criteria and NAAQS for PM. In developing the AQCD, the Agency made available three external review drafts to the public and for CASAC review; the final AQCD was released in 1996 (U.S. EPA, 1996). The EPA's OAQPS prepared a Staff Paper that was released in November 1997, after CASAC and public review of two drafts (U.S. EPA, 1996; Wolff, 1996). Revisions to the PM standards were proposed in 1996, and in 1997 the EPA promulgated revisions (61 FR 65738; December 13, 1996; 62 FR 38652, July 18, 1997). With the 1997 decision, the EPA added new standards, using $PM_{2.5}$ as the indicator for fine particles (with $PM_{2.5}$ referring to particles with a nominal mean aerodynamic diameter less than or equal to 2.5 µm). The new secondary standards were set equal to the primary standards, in all respects, as follows: (1) an annual standard with a level of 15.0 $\mu g/m^3$, based on the 3-year average of annual arithmetic mean $PM_{2.5}$ concentrations from single or multiple community-oriented monitors;[17] and (2) a 24-hour standard with a level of 65 $\mu g/m^3$, based on the 3-year average of the 98th percentile of 24-hour $PM_{2.5}$ concentrations at each monitor within an area. Further, the EPA retained the annual PM_{10} standard, without revision, and revised the form of the 24-hour PM_{10} standard to be based on the 99th percentile of 24-hour PM_{10} concentrations at each monitor in an area.

Following promulgation of the 1997 PM NAAQS, petitions for review were filed by several parties, raising a broad range of issues. In May 1999, the U.S. Court of Appeals for the

[17] The 1997 annual $PM_{2.5}$ standard was compared with measurements made at the community-oriented monitoring site recording the highest concentration or, if specific constraints were met, measurements from multiple community-oriented monitoring sites could be averaged (i.e., spatial averaging"). In the last review (completed in 2012) the EPA replaced the term "community-oriented" monitor with the term "area-wide" monitor. Area-wide monitors are those sited at the neighborhood scale or larger, as well as those monitors sited at micro- or middle-scales that are representative of many such locations in the same core-based statistical area (CBSA) (78 FR 3236, January 15, 2013).

District of Columbia Circuit (D.C. Circuit) upheld the EPA's decision to establish fine particle standards, (*American Trucking Ass'ns, Inc. v. EPA*, 175 F. 3d 1027, 1055-56 [D.C. Cir. 1999]). The D.C. Circuit also found "ample support" for the EPA's decision to regulate coarse particle pollution, but vacated the 1997 PM_{10} standards, concluding that the EPA had not provided a reasonable explanation justifying use of PM_{10} as an indicator for coarse particles (*American Trucking Ass'ns v. EPA*, 175 F. 3d at 1054-55). Pursuant to the D.C. Circuit's decision, the EPA removed the vacated 1997 PM_{10} standards, and the pre-existing 1987 PM_{10} standards remained in place (65 FR 80776, December 22, 2000). The D.C. Circuit also upheld the EPA's determination not to establish more stringent secondary standards for fine particles to address effects on visibility (*American Trucking Ass'ns v. EPA*, 175 F. 3d at 1027). The D.C. Circuit also addressed more general issues related to the NAAQS, including issues related to the consideration of costs in setting NAAQS and the EPA's approach to establishing the levels of NAAQS (as summarized in section 1.2 above).

In October 1997, the EPA initiated the third periodic review of the air quality criteria and NAAQS for PM (62 FR 55201, October 23, 1997). After the CASAC and public review of several drafts of the AQCD, the EPA released the final AQCD in October 2004 (U.S. EPA, 2004a and 2004b). The EPA's OAQPS finalized the Staff Paper in December 2005 (U.S. EPA, 2005). On December 20, 2005, the EPA announced its proposed decision to revise the NAAQS for PM and solicited public comment on a broad range of options (71 FR 2620, January 17, 2006). On September 21, 2006, the EPA announced its final decisions to revise the PM NAAQS to provide increased protection of public health and welfare, respectively (71 FR 61144, October 17, 2006). Revisions to the secondary standards were identical to those for the primary standards, with the decision describing the protection provided specifically for visibility and non-visibility related welfare effects (71 FR 61203-61210, October 17, 2006). With regard to the standards for fine particles, the EPA revised the level of the 24-hour $PM_{2.5}$ standards to 35 $\mu g/m^3$, retained the level of the annual $PM_{2.5}$ standards at 15.0 $\mu g/m^3$, and revised the form of the annual $PM_{2.5}$ standards by narrowing the constraints on the optional use of spatial averaging. With regard to the standards for PM_{10}, the EPA retained the 24-hour standards, with levels at 150 $\mu g/m^3$, and revoked the annual standards.

Several parties filed petitions for review of the 2006 PM NAAQS decision. One of these petitions raised the issue of setting the secondary $PM_{2.5}$ standards identical to the primary standards. On February 24, 2009, the D.C. Circuit issued its opinion in the case *American Farm Bureau Federation v. EPA*, 559 F. 3d 512 (D.C. Cir. 2009) and remanded the standards to the EPA because the Agency failed to adequately explain why setting the secondary PM standards identical to the primary standards provided the required protection for public welfare, including

protection from visibility impairment (*Id.* at 528-32). The EPA responded to the court's remands as part of the subsequent review of the PM NAAQS, which was initiated in 2007.

In June 2007, the EPA initiated the fourth periodic review of the air quality criteria and the PM NAAQS (72 FR 35462, June 28, 2007). Based on the NAAQS review process, as revised in 2008 and again in 2009, the EPA held science/policy issue workshops on the primary and secondary PM NAAQS (72 FR 34003, June 20, 2007; 72 FR 34005, June 20, 2007), and prepared and released the planning and assessment documents that comprise the review process (i.e., IRP [U.S. EPA, 2008b], ISA [U.S. EPA, 2009b], REA planning document for welfare [U.S. EPA, 2009c], and an urban-focused visibility assessment [U.S. EPA, 2010], and PA [U.S. EPA, 2011]). In June 2012, the EPA announced its proposed decision to revise the NAAQS for PM (77 FR 38890, June 29, 2012). In December 2012, the EPA announced its final decisions to revise the primary and secondary $PM_{2.5}$ annual standards (78 FR 3086, January 15, 2013). With regard to the secondary standards, the EPA retained the 24-hour $PM_{2.5}$ and PM_{10} standards, with a revision to the form of the 24-hour $PM_{2.5}$, to eliminate the option for spatial averaging (78 FR 3086, January 15, 2013).

Petitioners challenged the EPA's final rule. Petitioners argued that the EPA acted unreasonably in revising the level and form of the annual standard and in amending the monitoring network provisions. On judicial review, the revised standards and monitoring requirements were upheld in all respects (*NAM v. EPA*, 750 F.3d 921, D.C. Cir. 2014).

The subsequent review of the PM secondary standards, completed in 2020, focused on consideration of protection provided from visibility effects, materials damage, climate effects (85 FR 82684, December 18, 2020). The evidence for ecological effects of PM is addressed in the review of the air quality criteria and standards described in this PA.[18]

1.4 CURRENT REVIEW

In August 2013, the EPA issued a call for information in the *Federal Register* for information related to the newly initiated review of the air quality criteria for oxides of sulfur and oxides of nitrogen and announced a public workshop to discuss policy-relevant scientific information to inform the review (78 FR 53452, August 29, 2013). Based in part on the information received in response to the call for information, the EPA developed a draft IRP which was made available for consultation with the CASAC and for public comment (80 FR 69220, November 9, 2015). Comments from the CASAC and the public on the draft IRP were considered in preparing the final IRP (Diez Roux and Fernandez, 2016; U.S. EPA, 2017). In

[18] Welfare effects of PM considered in the review of the PM secondary standards completed in 2020, and reconsidered more recently, include effects on visibility and climate and materials damage (88 FR 5558, January 27, 2023).

developing the final IRP, the EPA expanded the review to also include review of the criteria and standards related to ecological effects of PM in recognition of linkages between these pollutants (oxides of nitrogen, oxides of sulfur and PM) with respect to deposition and atmospheric chemistry, as well as from an ecological effects perspective (U.S. EPA, 2017). Addressing the pollutants together enables a comprehensive consideration of the nature and interactions of the pollutants, which is important for ensuring thorough evaluation of the scientific information relevant to ecological effects of N and S deposition.

In March 2017, the EPA released the first external review draft of the *Integrated Science Assessment (ISA) for Oxides of Nitrogen, Oxides of Sulfur, and Particulate Matter Ecological Criteria* (82 FR 15702, March 30, 2017), which was then reviewed by the CASAC at a public meeting on May 24-25, 2017 (82 FR 15701, March 30, 2017) and August 31, 2017 (82 FR 35200, July 28, 2017; Diez Roux and Fernandez, 2017). With consideration of comments from the CASAC and the public, the EPA released a second external review draft (83 FR 29786, June 26, 2018), which was reviewed by the CASAC at public meetings on September 5-6, 2018 (83 FR 2018; July 9, 2018) and April 27, 2020 (85 FR 16093, March 30, 2020; Cox, Kendall, and Fernandez 2020a).[19] The EPA released the final ISA in October 2020 (85 FR 66327, October 19, 2020; U.S. EPA, 2020). In planning for quantitative aquatic acidification exposure/risk analyses for consideration in the PA, the EPA solicited public comment and consulted with the CASAC (83 FR 31755, July 9, 2018; Cox, Kendall, and Fernandez, 2020b; U.S. EPA, 2018; 83 FR 42497, August 22, 2018).

The draft PA was completed in May 2023 and made available for review by the CASAC and for public comment (88 FR 34852, May 31, 2023). The CASAC review was conducted at public meetings held on June 28-29, 2023 (88 FR 17572, March 23, 2023), and September 5-6, 2023 (88 FR 45414, July 17, 2023). The CASAC conveyed advice on the standards and comments on the draft PA in its September 27, 2023 letter to the Administrator (Sheppard, 2023). The CASAC advice on the standards is summarized in section 7.3 and considered in the conclusions in section 7.4. The CASAC comments on the draft PA have informed completion of this document. Additions and changes to the PA in consideration of those comments and public comments include the following.

- Chapter 1: A new section has been added that describes the 1990 CAA Amendments (section 1.3.3), and text has been revised or added to clarify a number of aspects including the PM effects considered in this review.
- Chapter 2: A number of revisions have been made to Chapter 2 in consideration of CASAC comments. These include an expanded overview of the acid deposition process and chemical complexity of sulfur and nitrogen oxides; more specific source

[19] A change in CASAC membership contributed to an extended time period between the two public meetings.

categorization of NH_3; and the relevance of the Clean Air Status and Trends Network (CASTNET) for this review. Some information has been moved into or repeated in Chapter 6 for improved cohesion in that chapter.

- Chapter 3: Clarification has been added regarding the effects considered in prior reviews of the PM standards and regarding some aspects of the aquatic acidification index developed in the 2012 review.
- Chapter 4: The discussion of N enrichment effects has been elevated, and the discussion of the evidence for effects in estuarine and coastal waters, particularly, has been appreciably expanded in light of CASAC comments.
- Chapter 5: The discussion of quantitative information pertaining to N enrichment effects in aquatic systems has been appreciably expanded, particularly as related to the evidence in estuarine and coastal areas, for which a new section has been added (section 5.2.3). Many revisions have been made to the description of the aquatic acidification REA and its results, both in this chapter and in the accompanying detailed appendix (5A) to provide clarification on a number of aspects, including those raised by the CASAC. Among these are the inclusion of a systematic uncertainty characterization of the aquatic acidification REA in Appendix 5A, section 5A.3.
- Chapter 6: This chapter and the accompanying appendix (6A) have been substantially expanded in light of CASAC advice and comments. For example, a new systematic uncertainty characterization of the full array of air quality analyses has been included (section 6.3), with additional sensitivity analyses to address several CASAC comments on the trajectory-based analyses (e.g., stress test the selection of the sites of influence). Further, the presentation of trajectory-based analyses has been augmented to more completely describe the methodology and the basis for methodological choices in the approach employed. The analysis itself has incorporated longer trajectories to better account for long depositional lifetimes of some pollutants. A new discussion of co-occurring trends in emissions, ambient air concentrations and estimated deposition, which were noted in several aspects of CASAC comments, has been included in section 6.2.1.
- Chapter 7: In addition to appreciable revisions to accommodate consideration of the expanded and improved aspects of Chapters 4, 5 and 6, a new section has been added that summarizes the CASAC advice on the standards in this review. The conclusions section has also been revised to take into account the changes across the PA and advice from the CASAC.

The timeline for the remainder of this review is governed by a consent decree that requires the EPA to sign a notice of proposed decision by April 9, 2024, and a final decision notice by December 10, 2024 (*Center for Biological Diversity v. Regan* [N.D. Cal., No. 4:22-cv-02285-HSG]).

1.5 ORGANIZATION OF THIS DOCUMENT

This PA includes staff's evaluation of the policy implications of the scientific assessment of the evidence presented and assessed in the 2020 ISA and of results of quantitative assessments

based on that information presented and assessed in this document. This evaluation informs staff's conclusions and identification of policy options for consideration in this review of the secondary standards addressing public welfare effects associated with the presence of oxides of nitrogen, oxides of sulfur, and PM in the ambient air.

Following this introductory chapter, this document presents policy relevant information drawn from the 2020 ISA as well as assessments that translate this information into a basis for staff conclusions as to policy options that are appropriate to consider in this review. The discussions are generally framed by addressing policy-relevant questions that have been adapted from those initially presented in the 2017 IRP.

- Chapter 2 provides an overview of current information on N oxides, SO_X, and PM-related emissions, how these pollutants are transformed in the atmosphere and contribute to deposition of S and N compounds. Chapter 2 also summarizes current air concentrations and long-term trends of these pollutants and associated deposition, as well as key aspects of the ambient air monitoring requirements.
- Chapter 3 summarizes the basis for the existing standards, describes key conclusions from 2012 review, recognizes key aspects of decision-making in NAAQS reviews and provides an overview of approach taken in this PA to consider the secondary standards with regard to protection for both direct and deposition-related effects.
- Chapter 4 provides an overview of the evidence as assessed in the 2020 ISA regarding ecosystem effects of N oxides, S oxides and PM in ambient air, and potential implications for effects of public welfare significance.
- Chapter 5 summarizes the information regarding exposure conditions associated with effects. The quantitative REA for aquatic acidification performed in this review based on the available evidence and quantitative tools is described, with associated details presented in Appendix 5A. For other categories of effects, the available quantitative information regarding direct and deposition-related effects of N oxides, SO_X, and PM to deposition related effects is summarized, with associated details regarding terrestrial effects information presented in Appendix 5B.
- Chapter 6 describes analyses and associated relationships between the deposition of S and N compounds and air quality metrics related to SO_X, N oxides, and PM in ambient air. The analyses in this chapter (for which associated details are presented in Appendix 6A) are intended to inform an understanding of the relationships between ambient air concentrations and deposition, both in locations near sources and in rural areas, where there may be sensitive ecosystems of concern for this review.
- Chapter 7 discusses evidence- and air quality/exposure/risk-based considerations and summarizes conclusions regarding an array of options appropriate for consideration. Consideration is given to the adequacy of protection afforded by the current standards for both direct and deposition-related effects. This chapter also identifies key uncertainties and associated needs for additional future research.

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2 AIR QUALITY AND DEPOSITION

This chapter begins with an overview of the atmospheric processes for N oxides and oxides of sulfur (SO_X), including those present as particulate matter (PM). This includes a description of the most relevant pollutants and how they can be transformed in the atmosphere and contribute to deposition of nitrogen (N) and sulfur (S) species (section 2.1). Subsequent sections summarize the sources of N oxides, SO_X, and PM emissions (section 2.2), describe measurement of relevant species including national monitoring networks and methods (section 2.3), describe recent observed trends in N, S, and PM species concentrations (section 2.4), and describe the way deposition estimates are developed (section 2.5).

2.1 ATMOSPHERIC TRANSFORMATION OF NITROGEN, SULFUR, AND PM SPECIES

This section briefly describes the key processes associated with atmospheric deposition of nitrogen and sulfur species, including both gaseous species and those that are present as PM. The pathway from emission to eventual deposition is specific across pollutants and is influenced by a series of atmospheric processes and often non-linear chemical transformations that occur at multiple spatial and temporal scales. Figure 2-1 is a simple schematic that identifies some of the individual pollutants that are part of oxides of nitrogen, oxides of sulfur, and PM, as well as how they can be interconnected. Each of these three categories of species are discussed more fully below.

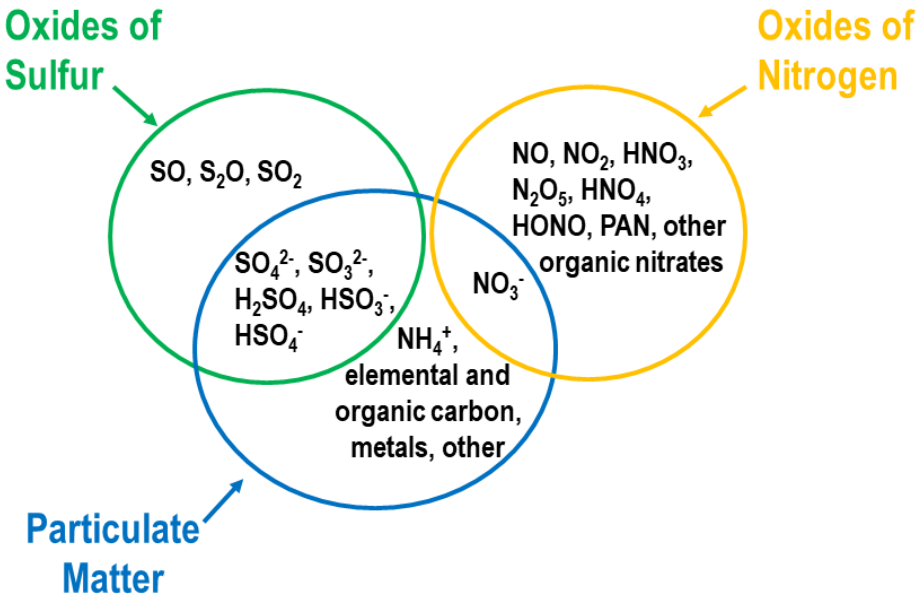

Figure 2-1. Schematic of most relevant individual pollutants that comprise oxides of nitrogen, oxides of sulfur, and particulate matter.

2.1.1 Oxides of Sulfur

Sulfur dioxide (SO_2) is one of a group of highly reactive gases collectively known as "oxides of sulfur" (SO_X). Oxides of sulfur may include sulfur monoxide (SO), SO_2, sulfur trioxide (SO_3), disulfur monoxide (S_2O), and various aerosol forms including sulfuric acid (H_2SO_4), bisulfite (HSO_3^-), sulfite (SO_3^{2-}), hydrogen bisulfate and, principally, sulfate (SO_4^{2-}). As discussed in more detail in section 2.2, SO_X is mostly emitted from combustion processes (e.g., stationary fuel combustion sources) in the form of SO_2. Aerosol SO_4^{2-} may also be emitted directly from combustion. Sulfur dioxide is generally present at higher concentrations in the ambient air than the other gaseous SO_X species (ISA, Appendix 2, section 2.1), and as a result, the indicator for the NAAQS for SO_X is SO_2.

Once emitted to the atmosphere SO_2 can react in both the gas phase and in aqueous solutions such as clouds and particles to form SO_4^{2-} (McMurry et al., 2004). There are multiple pathways for this process to occur. SO_2 is generally oxidized to sulfate following dissolution in cloud droplets, which can yield fast rates of sulfate production (up to 100% per hour). In the daytime, atmospheric oxidation may also convert gas phase SO_2 to H_2SO_4, which quickly and nearly completely condenses on existing particles or forms new sulfate particles (ISA, Appendix 2, section 2.3.2). The SO_2 to sulfate conversion typically occurs at rates of 0.1 to 10% per hour (Eatough et al., 1994), with higher rates associated with higher temperatures, sunlight, and the presence of oxidants. The conversion rates are determined by the availability of oxidants. The principal oxidizing agents for SO_2 are hydrogen peroxide, ozone and oxygen. Their relative level of influence depends on their concentration and the pH (Seinfeld and Pandis, 1998). Depending on the availability of ammonia, sulfate may also be present as ammonium bisulfate (NH_4HSO_4) or ammonium sulfate ($(NH_4)_2SO_4$).

Sulfate particles generally fall within the fine particle size range and contribute to $PM_{2.5}$ concentrations. The atmospheric lifetime of sulfate particles is relatively long, ranging from 2 to 10 days (as compared to SO_2, which is usually removed from the atmosphere within 2 days of its emission). As a result, sulfate concentrations tend to be regionally homogeneous (see section 2.4.2). Dry deposition can be an influential removal process for SO_2 on local scales, with a lifetime of approximately one day to one week. Following oxidation of SO_2 to particulate SO_4^{2-}, wet deposition is generally the primary removal process. The wet deposition lifetime for atmospheric S is about one week (2008 ISA section 2.6.3.1).

Although particulate sulfate can dry deposit, it is more efficiently removed by precipitation (wet deposition) (e.g., Mulcahy et al., 2020).

2.1.2 Oxidized Nitrogen

The oxidized nitrogen species, nitric oxide (NO) and nitrogen dioxide (NO_2), are collectively referred to as NO_X. As discussed in more detail in section 2.2, the largest sources of NO_X emissions are related to fossil fuel combustion, which includes anthropogenic sources such as power plants, industrial facilities, motor vehicles, and wood burning stoves. Non-anthropogenic sources of NO_X can include wildfires, biological soil processes, and lightning. In the atmosphere, NO and NO_2 can be converted to other forms of oxidized nitrogen, including nitric acid (HNO_3), peroxynitric acid (HNO_4), nitrous acid (HNO_2), and peroxyacetyl nitrate (PAN) or other forms of organic nitrogen. The term "oxides of nitrogen" refers to all forms of oxidized nitrogen compounds (NO_Y), including nitric oxide, nitrogen dioxide and all other oxidized nitrogen-containing compounds formed from NO and NO_2 (ISA Appendix 2, section 2.3.1). The indicator for the NAAQS for oxides of N is NO_2.

Oxidation of NO_X in the daytime or dinitrogen pentoxide (N_2O_5) hydrolysis in cold, nighttime conditions produce HNO_3. HNO_3 either settles onto surfaces directly (via dry deposition) or be scavenged by particles or cloud water to form nitrate (ISA Appendix 2, section 2.3.1). Facilitated by cold, humid conditions and the availability of excess NH_3, some of these compounds can partition from the gas phase into the solid or liquid phases as particulate nitrate (generically referred to as NO_3^-) and contribute to $PM_{2.5}$ concentrations. While almost all sulfate exists in the fine particle range, nitrate has a larger range in its size distribution and may either be fine or coarse, such that not all nitrate contributes to $PM_{2.5}$. Each form of oxidized nitrogen is removed from the atmosphere at different rates. For example, nitric acid quickly settles onto surfaces (via dry deposition) while particulate nitrate is more efficiently removed by precipitation (wet deposition).

2.1.3 Reduced Nitrogen

Reduced nitrogen, distinct from oxidized nitrogen, can also contribute to $PM_{2.5}$ formation and lead to adverse deposition-related effects. Ammonia is the most common form of atmospheric reduced nitrogen. Animal livestock operations and fertilized fields are the largest emission sources of NH_3, but there are combustion-related sources as well, such as vehicles and fires. Ammonia plays an important role as a precursor for atmospheric particulate matter and can be both deposited and emitted from plants and soils in a bidirectional exchange. NH_3 may contribute to inorganic $PM_{2.5}$ formation (as ammonium, NH_4^+) based on the availability of acid gases (HNO_3, H_2SO_4) and favorable meteorological conditions (low temperatures and high relative humidity). Ammonia reacts with gas phase HNO_3 to form ammonium nitrate or can partially or fully neutralize particle sulfate. The amount of ammonia present (along with organic compounds) is one determinant of the balance of ammonium sulfate and ammonium nitrate and

therefore influences the spatial extent of N and S deposition (ISA, Appendix 2, section 2.3.3). Ammonia tends to dry deposit near sources, but in particle form, ammonium (NH_4^+) can be transported farther distances and is most efficiently removed by precipitation. The sum of NH_3 and NH_4^+ is referred to as NH_X.

2.1.4 Atmospheric Processing

Once emitted to the atmosphere, SO_X, NO_Y, and NH_X are chemically transformed and transported until they are eventually removed from the atmosphere by deposition. The transport of emitted pollutants is a function of local and regional meteorological conditions such as wind fields and atmospheric stability that collectively govern how the pollutant species are advected and diffused. The formation of inorganic particulate matter following gas phase emission of SO_X, NO_Y and/or NH_3 is also sensitive to meteorological conditions (e.g., temperature, relative humidity), and the availability of basic (NH_3) or acidic (H_2SO_4, HNO_3) species. Along with the meteorological conditions, landscape characteristics and the chemical lifetime of a pollutant are also major factors in determining the distance at which pollutants contribute to deposition. Since the chemical form is important to determining the rate of dry and wet deposition (i.e., whether or not a pollutant deposits to the soil or vegetative surfaces), as well as the relationship between air concentrations and deposition, we use process-based models and quality-assured ambient air measurements to understand the transformation from emissions to concentrations to deposition (see sections 2.2 and 2.5).

2.2 SOURCES AND EMISSIONS OF NITROGEN, SULFUR, AND PM SPECIES

The sources and precursors to gaseous and particulate forms of SO_X, NO_Y, and NH_X vary and can include a combination of anthropogenic and natural sources. Anthropogenic sources of SO_2, NO_X, and NH_3 include power plants, industrial sources, motor vehicles, and agriculture. The National Emissions Inventory (NEI)[1] is a comprehensive and detailed estimate of air emissions of criteria pollutants, precursors to criteria pollutants, and certain hazardous air pollutants from air emissions sources. The NEI is released every three years based primarily upon data provided by State, Local, and Tribal air agencies for sources in their jurisdictions and supplemented by data developed by the EPA. For some sources, such as power plants, direct emission measurements enable the emissions estimates to be more certain than other sectors without such direct measurements. It should be recognized that emission inventories are inherently uncertain and contain assumptions that may influence the estimates of their magnitude

[1] https://www.epa.gov/air-emissions-inventories/national-emissions-inventory-nei

and trends. The 2020 NEI was released to the public on March 31, 2023. These 2020 data will be used for the summaries shown in the following sections describing emission estimates and trends. The reader is referred to the 2020 NEI[2] for further details (U.S. EPA, 2023a).

2.2.1 NO_X Emissions Estimates and Trends

Figure 2-2 shows the relative contributions of various sources to total U.S. NO_X emissions[3] in 2020, based on estimates contained in the NEI (U.S. EPA, 2023a). Anthropogenic sources account for a majority of NO_X emissions in the U.S., with highway vehicles (26%), stationary fuel combustion (25%), and non-road mobile sources (19%) identified as the largest contributors to total emissions. Highway vehicles include all on-road vehicles, including light duty as well as heavy duty vehicles, both gasoline- and diesel-powered. The stationary fuel combustion sector includes electricity generating units (EGUs), as well as commercial, institutional, industrial, and residential combustion of biomass, coal, natural gas, oil, and other fuels. Non-road mobile sources include aircraft, commercial marine vessels, locomotives, and non-road equipment. Other anthropogenic NO_X sources include agricultural field burning, prescribed fires, and various industrial processes such as cement manufacturing and oil and gas production. Natural sources of NO_X include emissions from lightning as well as from plants and soil (biogenic), which represent 12% of the total NOx emissions. In addition, fires (i.e., wild, prescribed, and agricultural) are estimated to represent 5% of the overall emissions of NO_X. Soil emission estimates come from the Biogenic Emissions Inventory System, version 4 (BEIS) model in the NEI. Biomass burning emissions (wild and prescribed fires) come from the Blue Sky Pipeline framework (developed by the U.S. Forest Service, https://github.com/pnwairfire/bluesky). More information on both these models can be found in our 2020 NEI Technical Support Document (TSD).[4]

Figure 2-3 shows the NO_X emissions density in tons/year per square mile for each U.S. County. The majority of NO_X emissions tend to be located near urban areas, which tend to have the most vehicle traffic and industrial sources. However, there are also some counties in rural areas with higher NO_X emissions due to the presence of large stationary sources such as EGUs or oil and gas extraction and generation.

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[2] https://www.epa.gov/air-emissions-inventories/2020-national-emissions-inventory-nei-data

[3] For all source categories, NO_X is compiled from emissions measurements that express NO_X mass based on the molecular weight of NO_2, which is 46 g/mole (40 CFR 51.40). Even though emissions from most sources initially consist mainly of NO, this expression of NO_X by NO_2 molecular weight is considered appropriate due to the fast rate of transformation of NO to NO_2 under ambient air conditions or when the emissions are exposed to any type of oxidant. While NO_X is made up of NO_2, NO, and, for mobile sources, HONO, the combination of these by mass is more simply done using a single molecular weight.

[4] https://www.epa.gov/system/files/documents/2023-03/NEI2020_TSD_Section8_Biogenics_0.pdf.

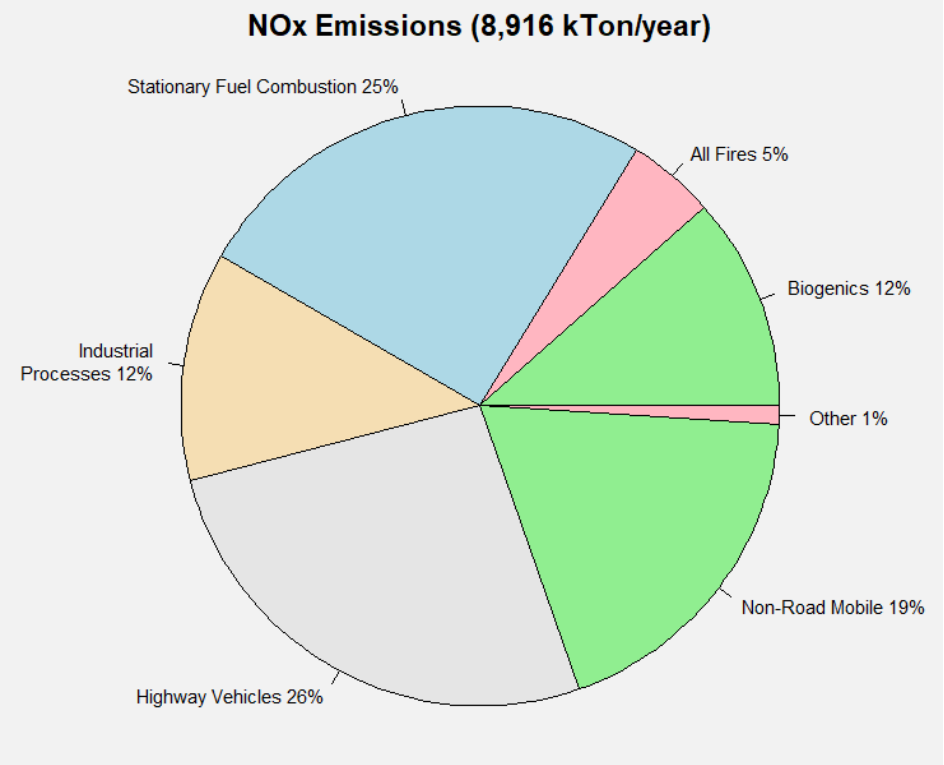

Figure 2-2. 2020 NO_X emissions estimates by source sector (U.S. EPA, 2023a). Note: The NEI, and this figure, do not include emissions from lightning.

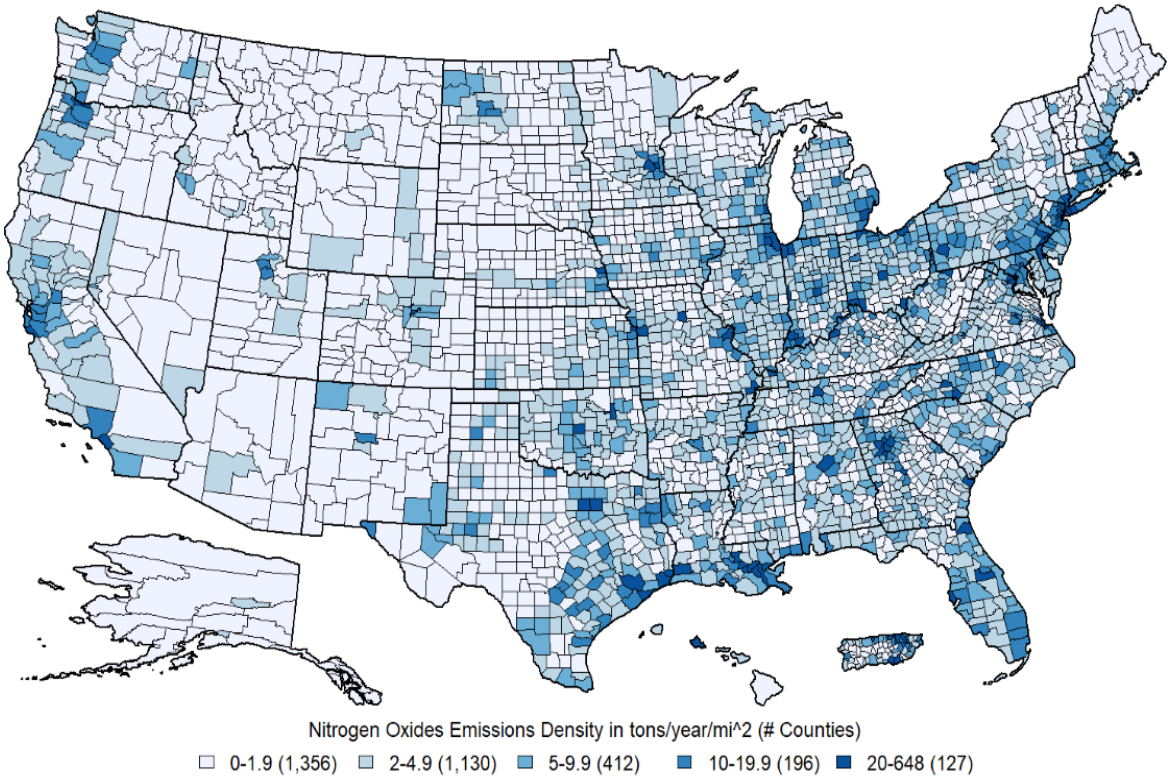

Figure 2-3. 2020 NO_X emissions density across the U.S. (U.S. EPA, 2023a).

Total anthropogenic NO_X emissions have trended strongly downward across the U.S. between 2002 and 2022 (U.S. EPA, 2023b). Nationwide estimates indicate a 70% decrease in anthropogenic NO_X emissions over this time period as a result of multiple regulatory programs (e.g., including the NO_X SIP Call, the Cross-State Air Pollution Rule (CSAPR), and the Tier 3 Light-duty Vehicle Emissions and Fuel Standards) implemented over the past two decades, as well as changes in economic conditions. As seen in Figure 2-4, the overall decrease in NO_X emissions has been driven primarily by decreases from the three largest emissions sectors. Specifically, compared to the 2002 start year, estimates for 2022 (from the 2020 NEI) indicate an 84% reduction in NO_X emissions from highway vehicles, a 68% reduction in NO_X emissions from stationary fuel combustion, and a 54% reduction in NO_X emissions from non-road mobile sources.

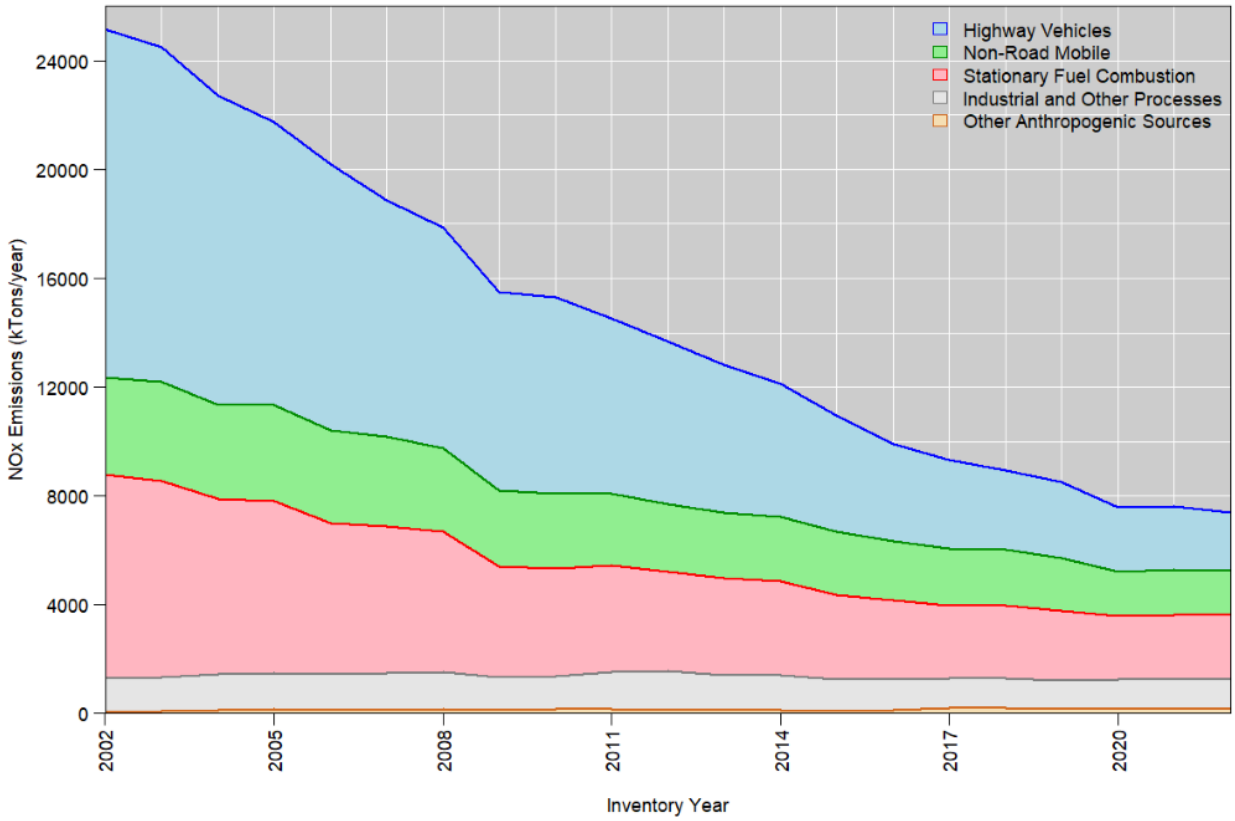

Figure 2-4. Trends in NO_X emissions by sector between 2002 and 2022 (U.S. EPA, 2023b).

2.2.2 SO_2 Emissions Estimates and Trends

Fossil fuel combustion is the main anthropogenic source of SO_2, primarily from coal-fired EGUs (48%). Sulfur is present to some degree in all fossil fuels, especially coal, and occurs as reduced organosulfur compounds. In the most common types of coal (anthracite, bituminous, subbituminous, and lignite), sulfur content varies between 0.4 and 4% by mass. Sulfur in fossil fuels is almost entirely converted to SO_2 during combustion. Other major anthropogenic sources of SO_2 emissions include industrial processes (27%) and stationary source fuel combustion (9%). Mobile sources, and agricultural and prescribed fires are smaller contributors. Figure 2-5 shows the percentage contribution of specific source categories to the total anthropogenic (plus wildfire) SO_2. Across all source categories, directly emitted sulfates are about 5% of the total emitted sulfur, although it can vary by source.

Figure 2-6 shows the SO_2 emissions density in tons/year per square mile for each U.S. county. The majority of SO_2 emissions tend to be located near large point sources such as coal-fired EGUs or large industrial facilities. Counties near urban areas also tend to have higher SO_2 emissions due to the higher concentration of industrial facilities. In some cases, counties in rural areas can also have higher emissions due to oil and gas extraction or fires.

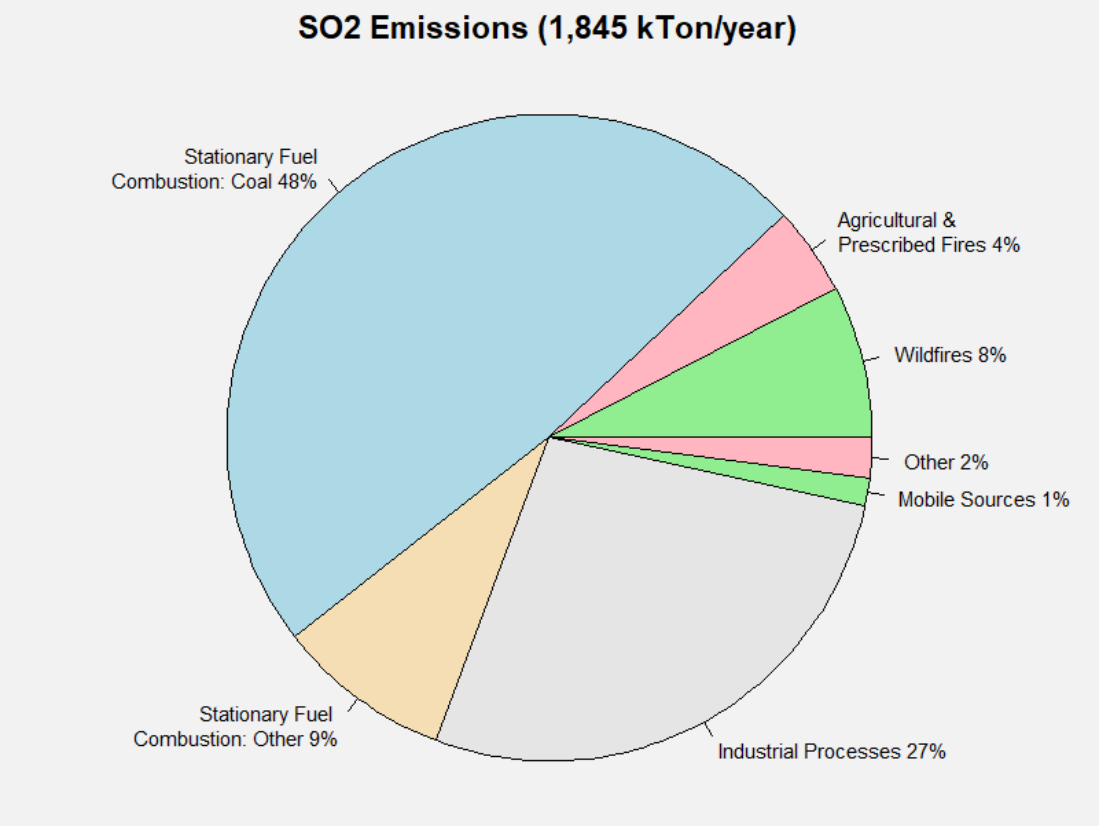

Figure 2-5. Estimates of 2020 SO_2 emissions by source sector (U.S. EPA, 2023a).

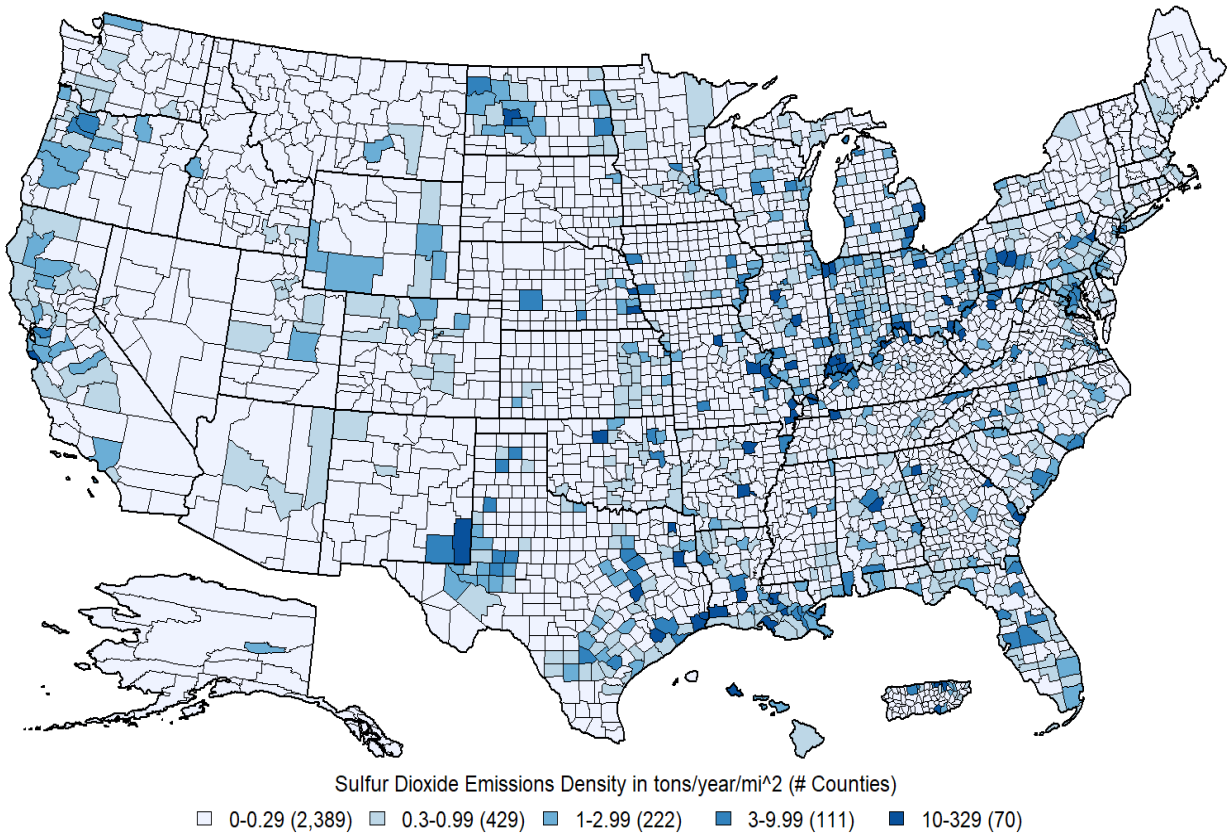

Figure 2-6. Estimates of 2020 SO_2 emissions density across the U.S. (U.S. EPA, 2023a).

Similar to NO_X, and for many of the same reasons, SO_2 emissions have declined significantly since 2002. Figure 2-7 illustrates the emissions changes over the 2002-2022 period. The data shows an 87% decrease in total SO_2 emissions over the period, including reductions of 91% in emissions from EGUs and 96% in emissions from mobile sources.

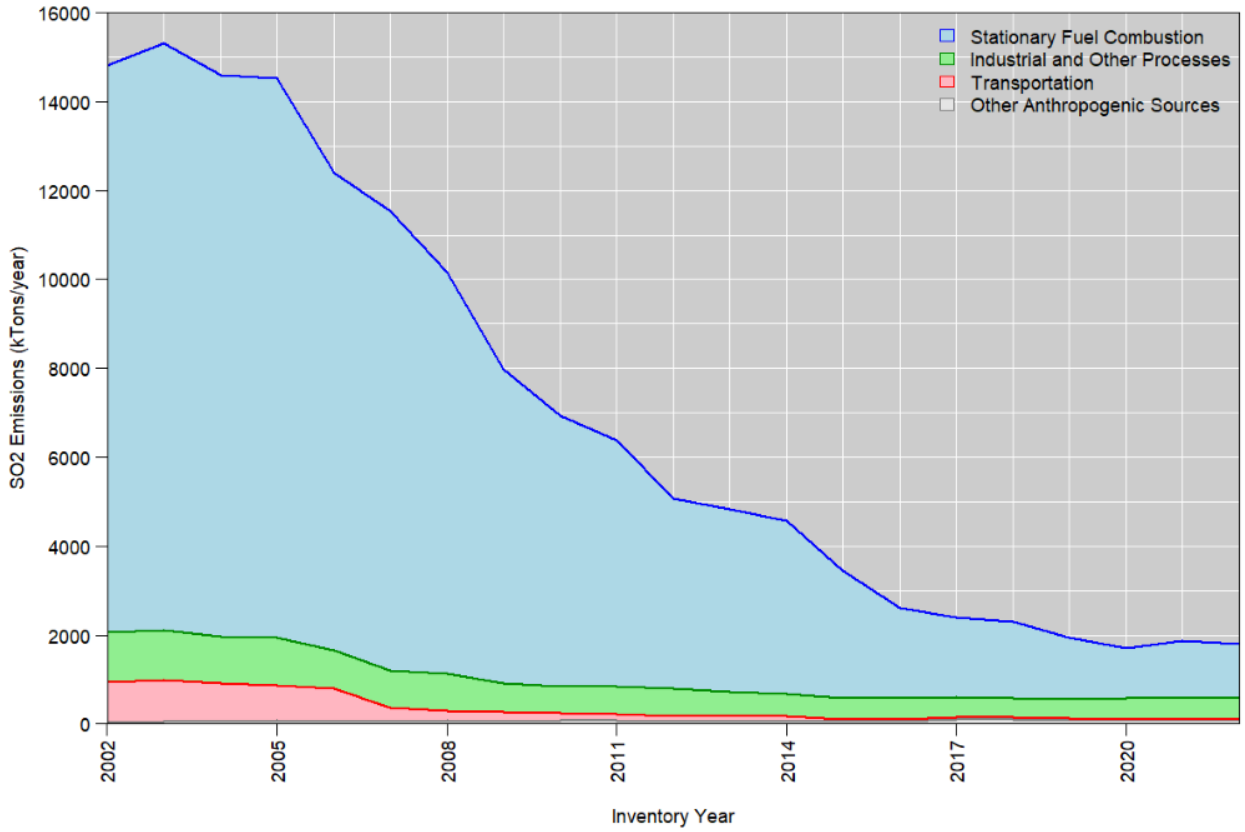

Figure 2-7. Trends in SO_2 emissions by sector between 2002 and 2022 (U.S. EPA, 2023b).

2.2.3 NH_3 Emissions Estimates and Trends

Ammonia is emitted directly into the atmosphere, unlike other atmospheric N species (e.g., organic N) that are formed through photochemical reactions. Figure 2-8 shows the percentage contribution of specific source categories to the total anthropogenic (plus wildfires) NH_3. In 2020, livestock waste (49%), fertilizer application (33%) and aggregate fires (11%) contributed most significantly to total annual emissions (5.5 million tons NH_3). Vehicles emit NH_3 due to the unintended formation of NH_3 from catalytic converters reducing NOx under fuel rich conditions for gasoline vehicles and from overdosing of urea in selective catalytic systems in modern heavy-duty vehicles (Easter and Bohac, 2016; Jeon et al., 2016; Khalek et al., 2015). While mobile source contributions to total NH_3 emissions are only about 2% at the national level, there is a growing body of evidence suggesting that vehicular sources may be underestimated in the NEI (Sun et al., 2017; Chen et al., 2022). Any underestimation in mobile source NH_3 emissions would mostly impact urban areas, where there is a lot of on-road mobile source traffic. The latest version of EPA's Motor Vehicle Emission simulator, MOVES4 (https://www.epa.gov/moves/latest-version-motor-vehicle-emission-simulator-moves), has been updated to incorporate real-world measurements of NH_3 emissions from vehicles, and it suggests

higher NH_3 emissions from onroad vehicles than previous inventories. This simulator, MOVES4, will be used in future versions of NEI. Figure 2-9 shows the NH_3 emissions density in tons per year per square mile for each U.S. county. Ammonia emissions are greatest in counties with significant agricultural output (e.g., central U.S., parts of CA, and eastern NC).

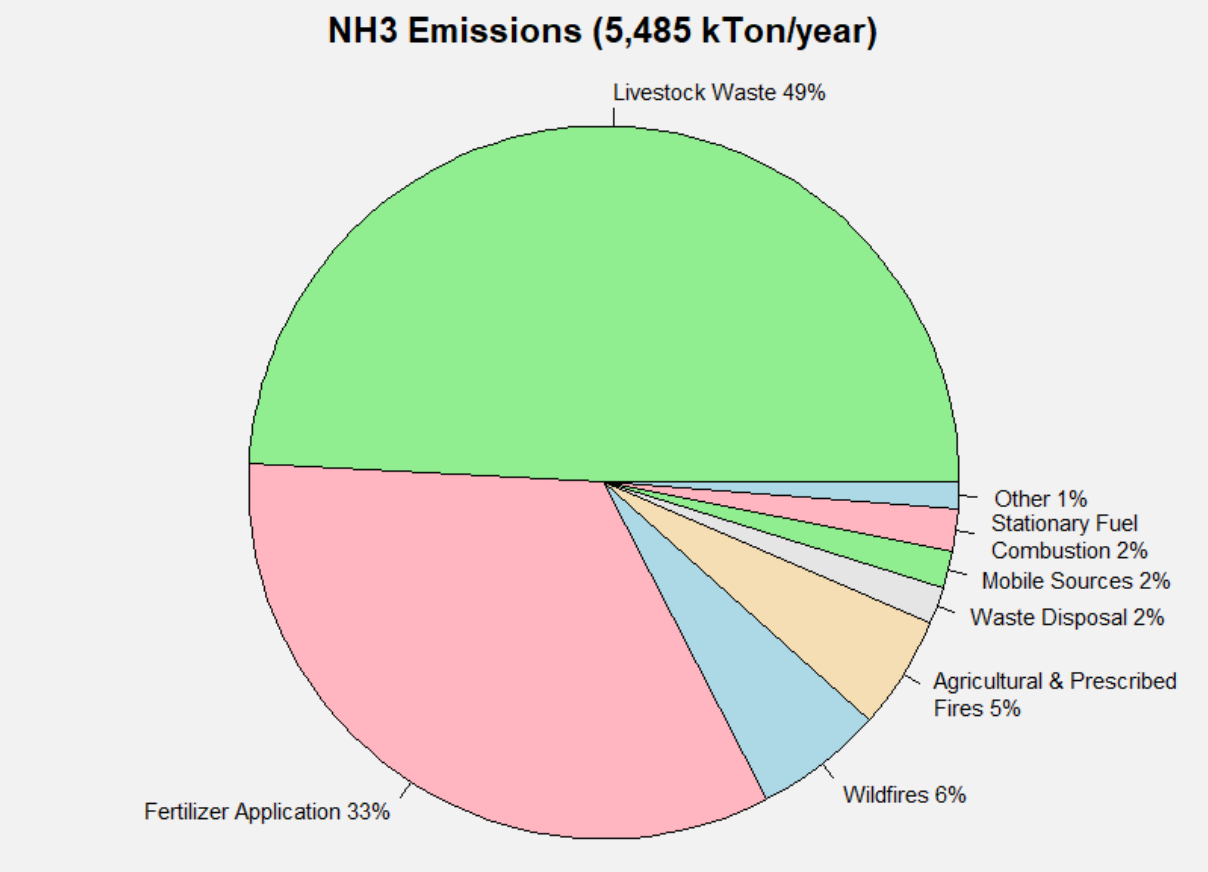

Figure 2-8. Estimates of 2020 NH_3 emissions by source sector (U.S. EPA, 2023a).

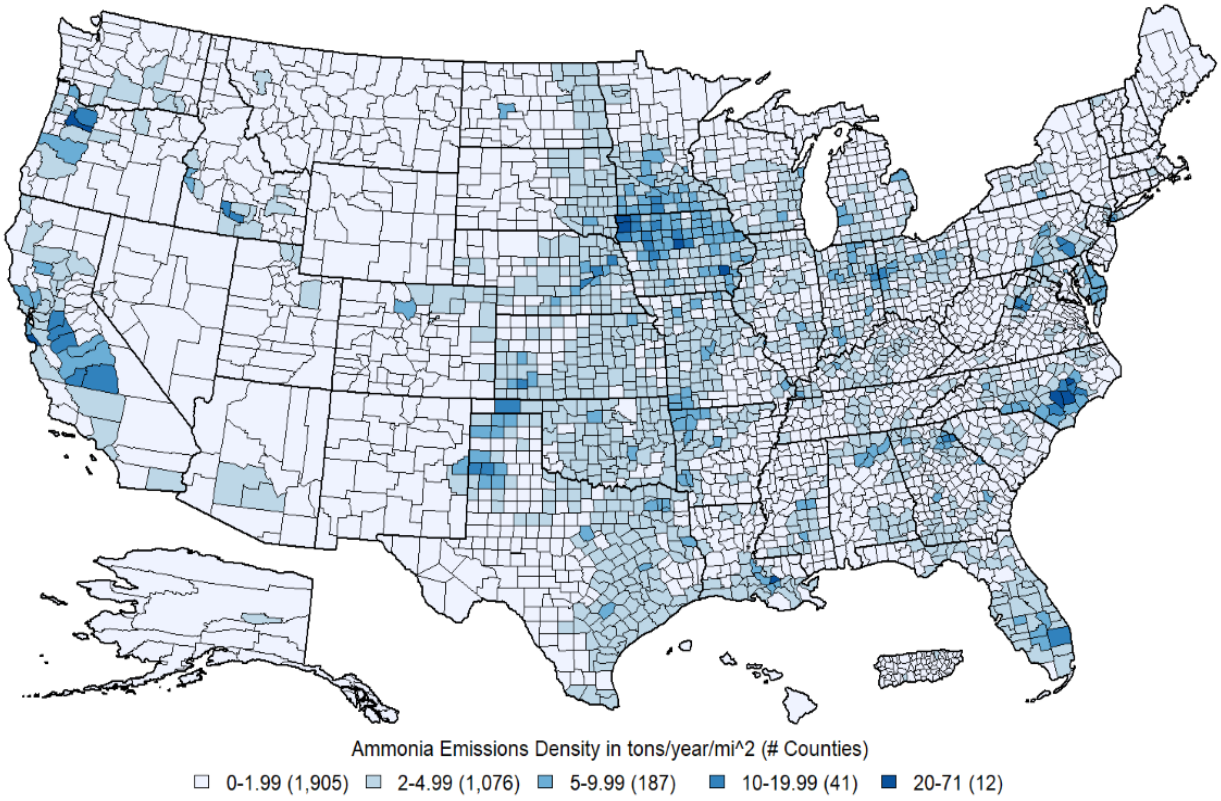

Figure 2-9. Estimates of NH_3 emissions density across the U.S. (U.S. EPA, 2023a).

Figure 2-10 shows NH_3 emission trends from 2002-2022 by sector. In comparison with NOx and SO_X emission trends, which demonstrated dramatic decreases over the past few decades, the annual rate of NH_3 emissions has increased by over 20 percent since 2002. The two largest contributors are livestock waste and fertilizer application which have increased by 11% and 44%, respectively, from 2002 to 2022. However, there is greater uncertainty in NH_3 emissions trends (ISA, Appendix 2, section 2.2.3) than with the other pollutants. This is partly due to a lack of control programs nationally for agricultural sources of NH_3. It is worth noting that variabilities associated with local management practices related to animal husbandry makes these emissions a bit more uncertain than emissions derived from, for example, direct measurements from EGU sources. The EPA has improved its models for simulating both livestock waste emissions and the fertilizer application process to inform development of the 2020 NEI which is expected to have reduced these uncertainties (U.S. EPA, 2023a).

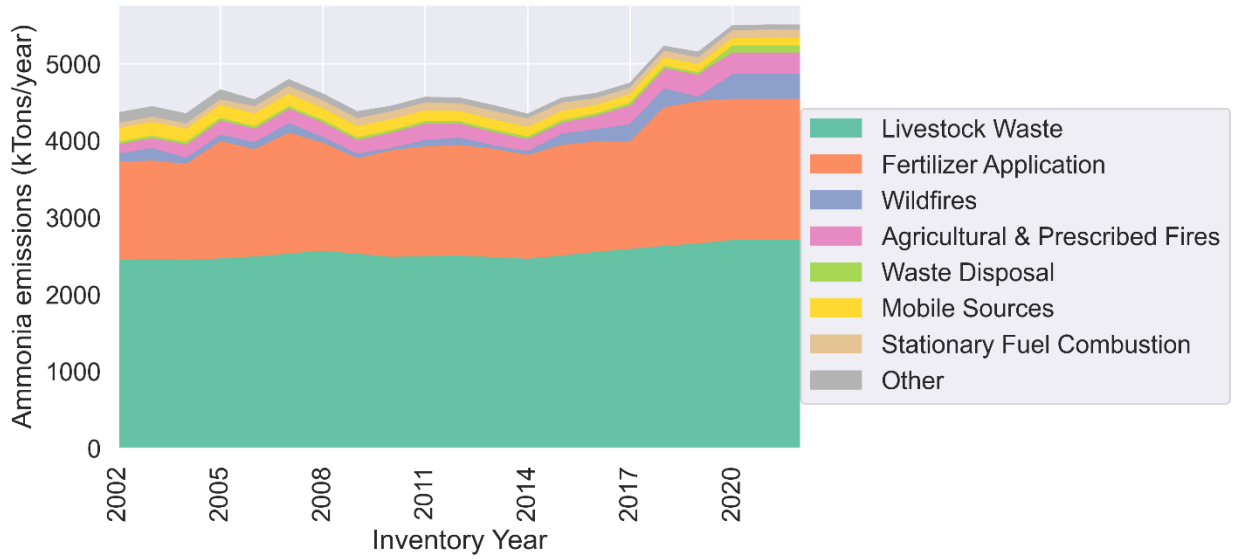

Figure 2-10. Trends in NH_3 emissions by sector between 2002-2022 (U.S. EPA, 2023b).

2.3 MONITORING AMBIENT AIR CONCENTRATIONS AND DEPOSITION

To promote uniform enforcement of the air quality standards set forth under the CAA, the EPA has established federal reference methods (FRMs) and federal equivalent methods (FEMs) for ambient air sample collection and analysis. Measurements for determinations of NAAQS compliance must be made with FRMs or FEMs. Federal reference methods and national monitoring networks have been established for NO_2 as the indicator of oxides of nitrogen, SO_2 as the indicator of sulfur oxides, and $PM_{2.5}$ and PM_{10} as indicators for PM.

As described briefly below, multiple monitoring networks measure the atmospheric concentrations of nitrogen oxides, SO_X, and PM, as well as wet deposition of N and S compounds. The largest routinely operating network that measures ambient air concentrations is the State and Local Air Monitoring Stations (SLAMS) network which includes measurement of one or more NAAQS pollutants at each site. There are three multipollutant networks involving NAAQS measurements which are largely sited at SLAMS.[5] These networks include: the National Core (NCore) multi-pollutant monitoring network, the Photochemical Assessment Monitoring Stations (PAMS) network, and the near-road network. The NCore network is notable in that it provides a core of sites, mostly located in urban areas, that provide collocated measurements of SO_2, NO, NO_Y, and PM components including ammonium, nitrate, and sulfate,

[5] A small number of multipollutant sites may have a monitor type different than SLAMS such as Tribal or Non-EPA Federal (e.g., National Park Service [NPS]).

although with sparser coverage than the FRM networks for SO_2 or NO_2. Collocated, ambient air measurements of SO_2, SO_4^{2-} and NO_Y (NO_Y is measured rather than NO_X) from NCore can be used to help estimate total deposition of oxides of nitrogen and sulfur. The primary objective of the PAMS network is to support the implementation of the ozone NAAQS; it also measures NO_Y, as well as NO_2. The near-road network is intended to capture short-term peak NO_2 concentrations for comparison to the NO_2 primary NAAQS. Many of the near-road sites are also required to have collocation with $PM_{2.5}$ and carbon monoxide (CO) monitors. One of the challenges associated with interpreting monitoring data in the context of a deposition-related secondary standard is that many, but not all, of the monitor sites are located in urban or suburban areas (where air quality concentrations are highest and human populations are greatest), while many of the areas where deposition effects are potentially of greatest concern tend to be in more rural areas.

2.3.1 NO_X Monitoring Networks

There were 491 monitoring sites, mostly in major metropolitan areas, reporting hourly NO_2 concentration data to the EPA during the 2019-2021 period; 80% of these NO_2 monitoring sites are part of the SLAMS network (U.S. EPA 2021a). This network relies on a chemiluminescent FRM and on multiple FEMs that use either chemiluminescence or direct measurement methods of NO_2. Chemiluminescent-based FRMs only detect NO in the sample stream. Therefore, a two-step process is employed to measure NO_2, based on the subtraction of NO from NO_X. Data produced by chemiluminescent analyzers include NO, NO_2, and NO_X measurements. As discussed in the ISA the traditional chemiluminescence FRM is subject to potential measurement biases resulting from interference by N oxides other than NO or NO_2 (ISA, Appendix 2, p. 2-34).[6] These potential biases are measurement uncertainties that can impact exposure analyses. However, within metropolitan areas, where a majority of the NO_2 monitoring network is located and is influenced by strong NO_X sources, the potential for bias related to other N oxides is relatively small.

Another important subset of SLAMS sites is the near-road monitoring network, which was required as part of the 2010 NO_2 primary NAAQS review and began operating in 2014. Near-road sites are required in each metropolitan statistical area (MSA) with a population of 1,000,000 or greater, and an additional near-road site is required in each MSA with a population of 2,500,000 or greater. There were 73 near-road monitors in operation during the 2019-2021 period. Finally, there are also a number of Special Purpose Monitors (SPMs), which are not required but are often operated by air agencies for short periods of time (i.e., less than 3 years) to

[6] The N oxides other than NO and NO_2 are often collectively abbreviated as NO_Z (i.e., $NO_Y = NO_X + NO_Z$).

collect data for human health and welfare studies, as well as other types of monitoring sites, including monitors operated by tribes and industrial sources. The SPMs are typically not used to assess compliance with the NAAQS. The locations of all NO_2 monitoring sites operating during the 2019-2021 period are shown in Figure 2-11.

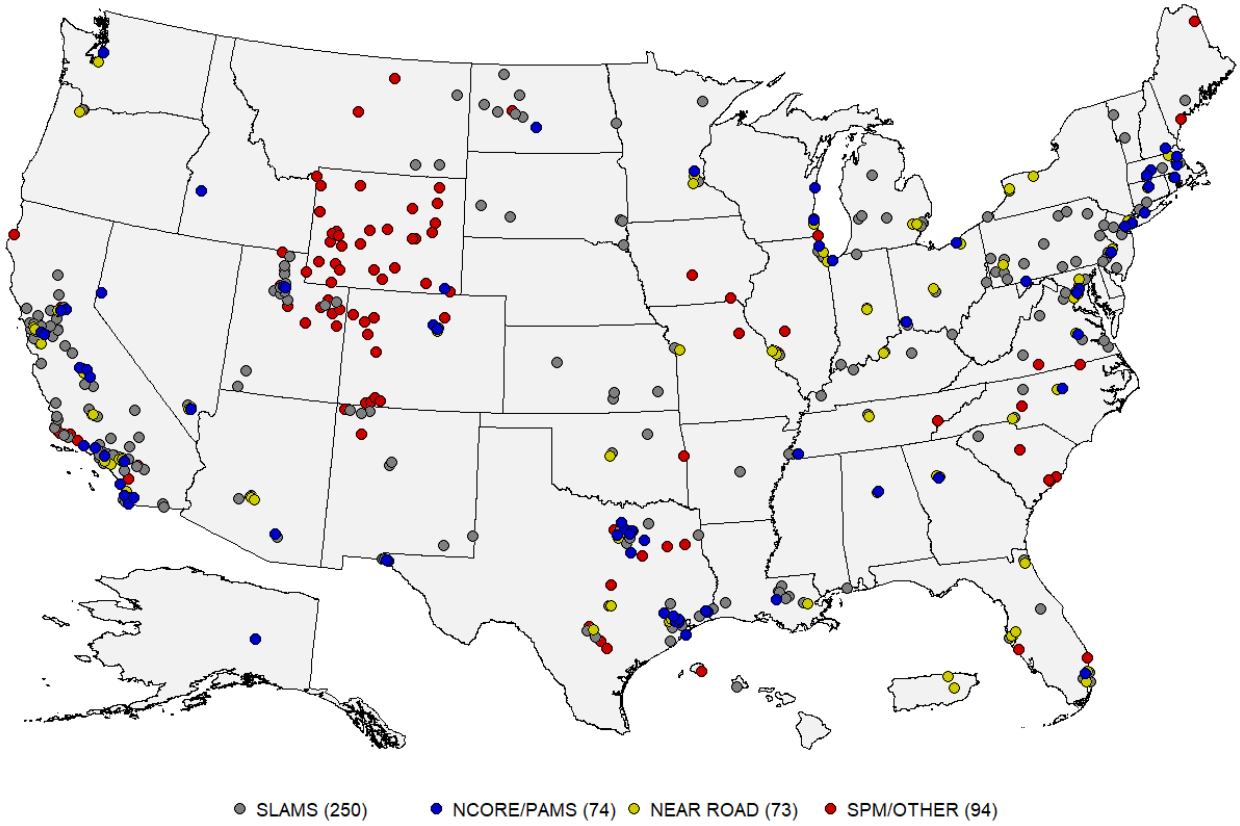

Figure 2-11. Locations of NO_2 monitors operating during the 2019-2021 period.

2.3.2 SO_2 Monitoring Networks

There were 505 monitoring sites reporting hourly SO_2 concentration data to the EPA during the 2019-2021 period (U.S. EPA 2021b). Over 75% of the SO_2 sites are part of the SLAMS network. Measurements are made using ultraviolet fluorescence instruments, which are designated as FRMs or FEMs and the data are reported as hourly concentrations with either the maximum 5-minute concentration for each hour or twelve 5-minute average concentrations for each hour. Additionally, as of 2015, States are required to monitor or model ambient air SO_2 levels in areas with stationary sources of SO_2 emissions of over 2,000 tons per year. The EPA identified over 300 sources meeting these criteria according to 2014 emissions data, and some States chose to set up ambient air monitoring sites to assess compliance with the SO_2 NAAQS. Some of these monitors are operated by the States as SLAMS monitors, while others are operated by the industrial sources. The locations of all SO_2 monitoring sites (FRM or FEM) operating during the 2019-2021 period are shown in Figure 2-12.

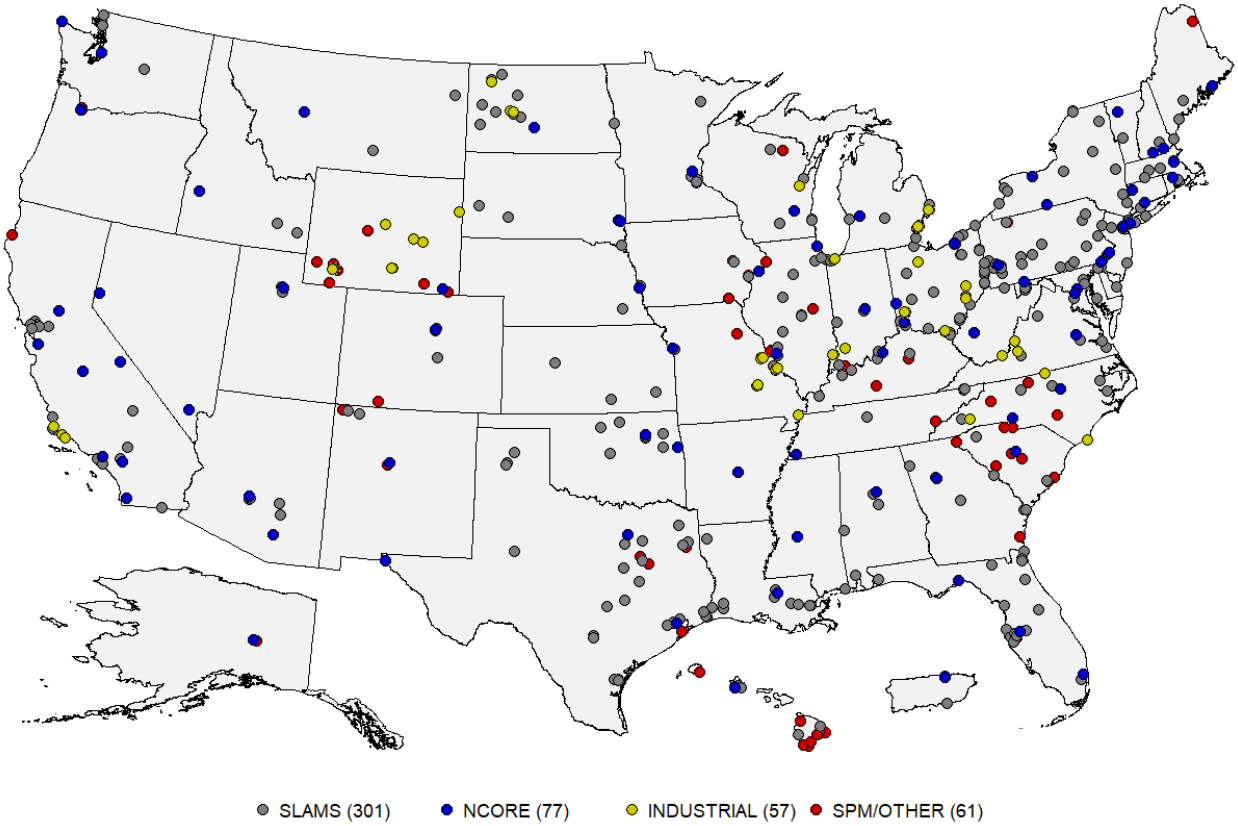

Figure 2-12. Locations of SO_2 monitors operating during the 2019-2021 period.

2.3.3 $PM_{2.5}$ and PM_{10} Monitoring Networks

As with NO_X and SO_2, the main network of monitors providing ambient air PM mass data for use in NAAQS implementation activities is the SLAMS network (including NCore). $PM_{2.5}$ monitoring was required for near-road network sites as part of the 2012 PM NAAQS review and these sites monitors were phased into the network between 2015 and 2017. Near-road sites are also required in each MSA with a population of 1,000,000 or greater. The $PM_{2.5}$ monitoring program remains one of the largest ambient air monitoring programs in the U.S. There were 1,067 monitoring sites reporting $PM_{2.5}$ data to the EPA during the 2019-2021 period (U.S. EPA 2021c). Figure 2-13 shows the locations of these monitoring sites. Approximately 50% of these monitoring sites operate automated FEMs which report continuous (hourly) $PM_{2.5}$ data while the remaining sites operate FRMs which collect 24-hour samples every day, every 3rd day, or every 6th day. There were 724 monitoring sites reporting PM_{10} data to the EPA during the 2019-2021 period. Figure 2-14 shows the locations of these monitoring sites. Approximately 61% of these monitoring sites operate FEMs that report continuous PM_{10} data while the remaining sites operate FRMs that typically collect samples every day, every 3rd day, or every 6th day.

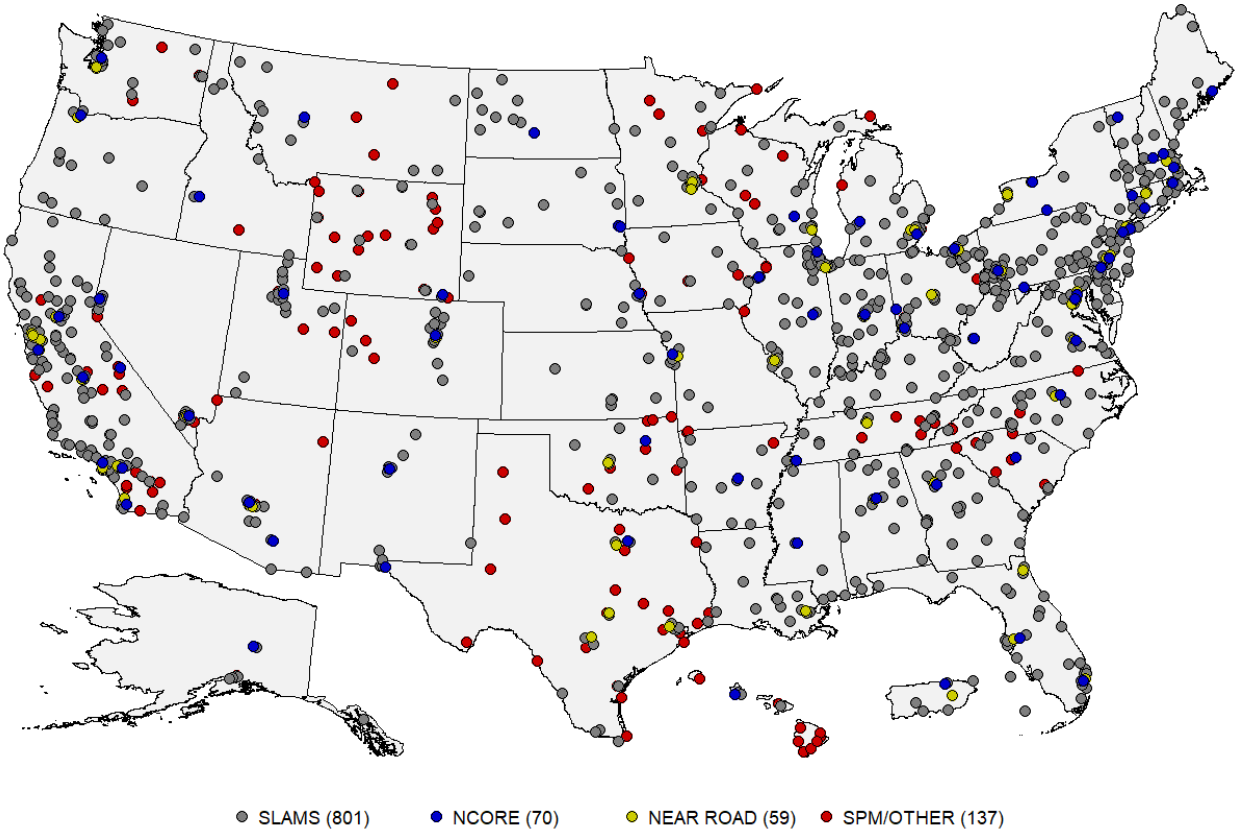

Figure 2-13. $PM_{2.5}$ mass monitors operating during the 2019-2021 period.

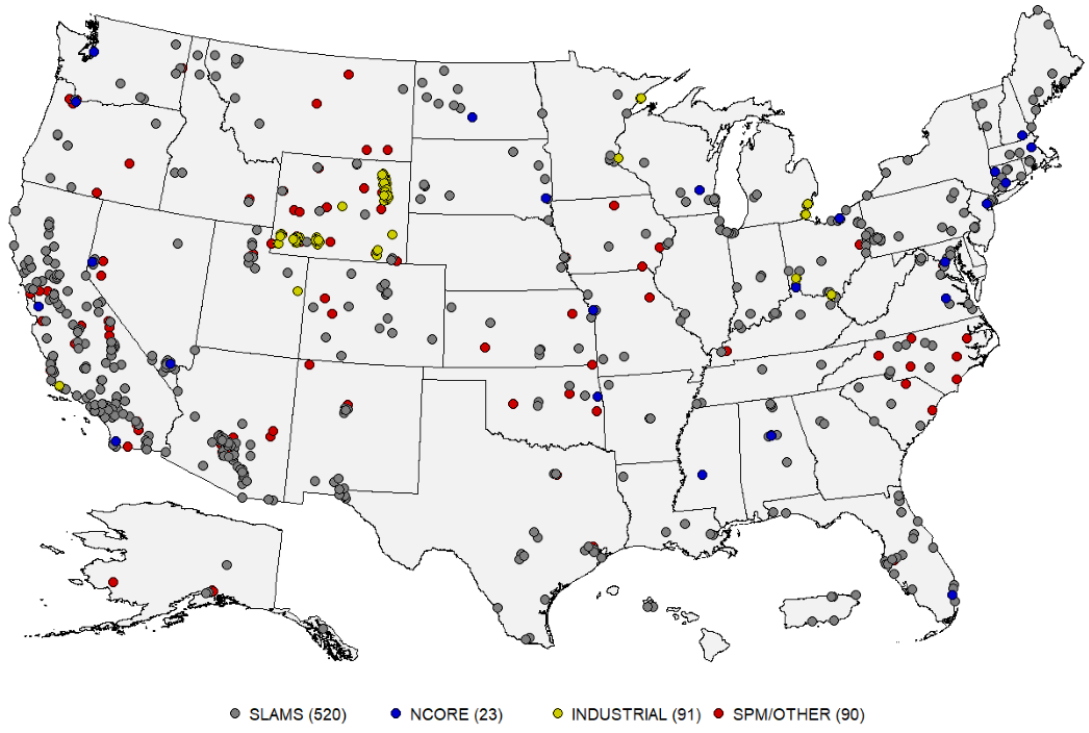

Figure 2-14. PM_{10} mass monitors operating during the 2019-2021 period.

Due to the complex nature of fine particles, the EPA and States implemented the Chemical Speciation Network (CSN) to better understand the components of fine particle mass at selected locations across the country. $PM_{2.5}$ speciation measurements are also collected at NCore stations. Additionally, specific components of fine particles are measured through the Interagency Monitoring of Protected Visual Environments (IMPROVE) monitoring program, which supports the regional haze program and tracks changes in visibility in Federal Class I areas as well as many other rural and some urban areas. The IMPROVE network consists of more than 100 monitoring sites in national parks and other remote locations and has also provided a reliable, long-term record of particulate mass and species components. The locations of the CSN (a mix of 3-day and 6-day sampling frequency) and IMPROVE (3-day sampling frequency) sites reporting speciated $PM_{2.5}$ data to the EPA during the 2019-2021 period are shown in Figure 2-15.

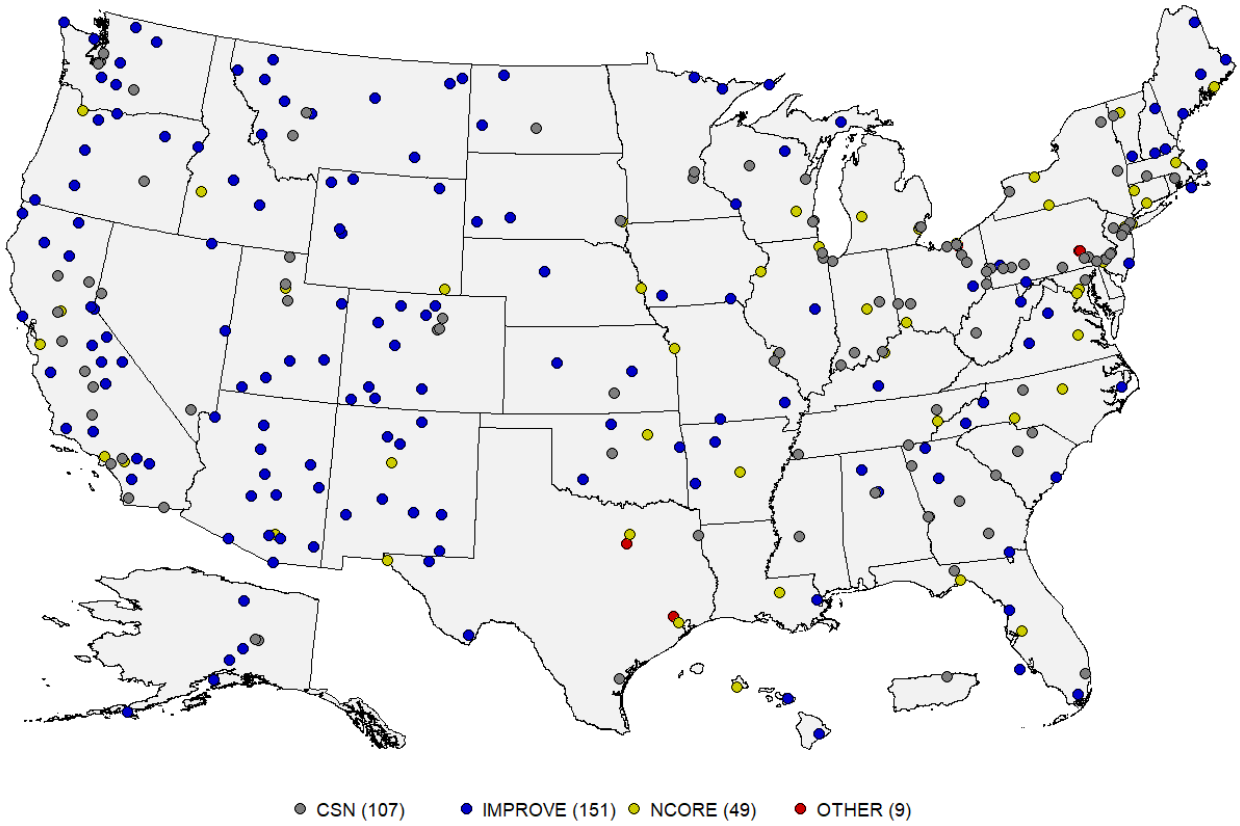

Figure 2-15. $PM_{2.5}$ speciation monitors operating during the 2019-2021 period.

2.3.4 Routine Deposition Monitoring

Wet deposition is measured as the product of pollutant concentration in precipitation and precipitation amounts (e.g., in rain or snow). Concentration in precipitation is currently measured as a weekly average by the National Atmospheric Deposition Program/National Trends Network

(NADP/NTN) across a national network of approximately 250 sites using a standard precipitation collector. The NADP precipitation network was initiated in 1978 to collect data on amounts, trends, and distributions of acids, nutrients, and cations in precipitation. The NTN is the only network (shown in Figure 2-16) that provides a long-term record of precipitation chemistry across the U.S. Sites are mainly located away from urban areas and pollution sources. An automated collector ensures that the sample is exposed only during precipitation (wet-only sampling). Nitrate, sulfate, and ammonium are all measured. Relatively high confidence has been assigned to wet deposition estimates because of established capabilities for measuring relevant chemical components in precipitation samples.

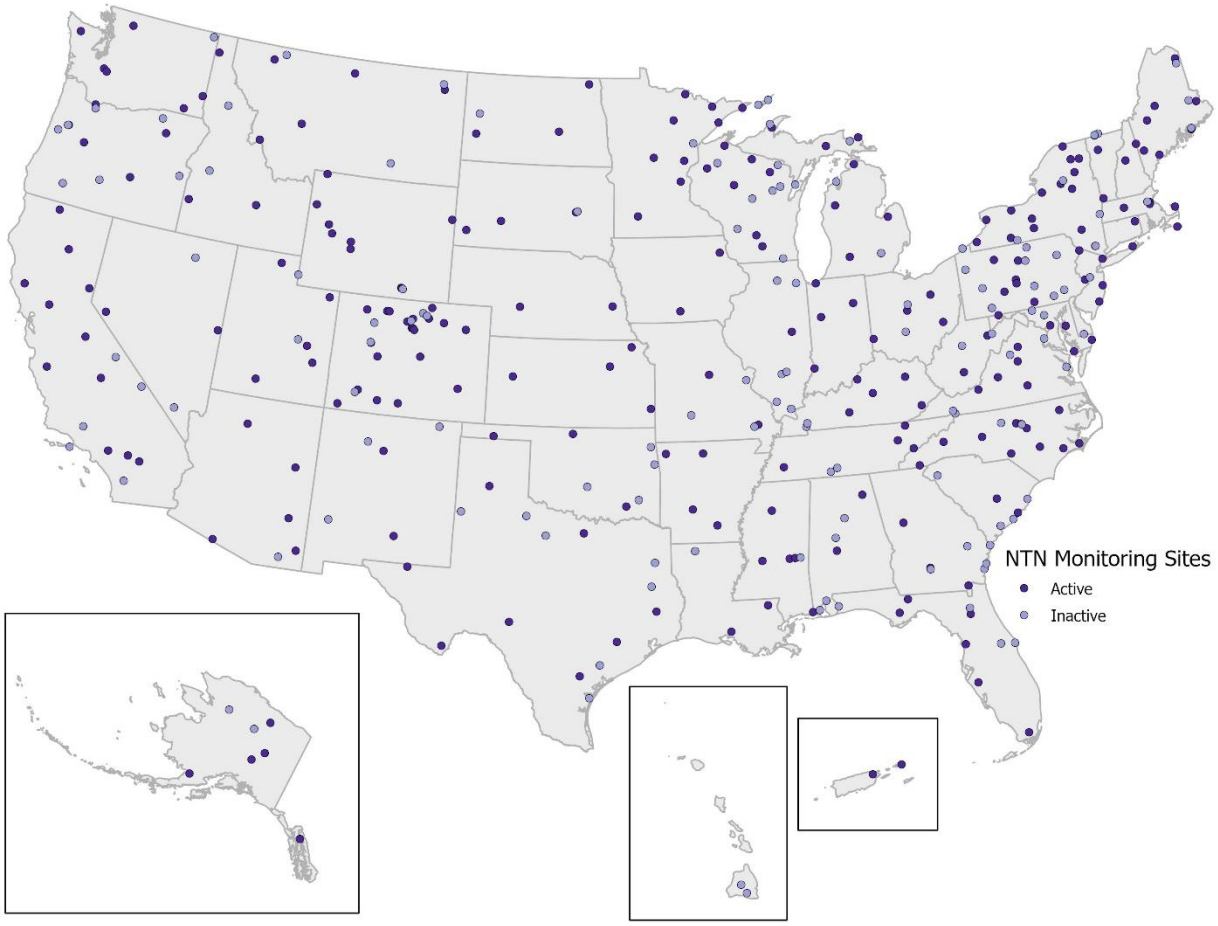

Figure 2-16. Location of NTN monitoring sites with different symbols for how many years the site has operated (through 2017). Source: NADP/NTN site information database (https://nadp.slh.wisc.edu/networks/national-trends-network/, accessed August 2023)

In contrast, direct measurements of dry deposition flux are rare and difficult, and dry deposition fluxes of gases and particles are estimated from concentration measurements by an inferential technique described in the 2008 ISA (U.S. EPA, 2008). Ambient air concentrations

are measured in the Clean Air Status and Trends Network (CASTNET), which was established under the 1991 CAA Amendments to assess trends in acidic deposition. CASTNET is a long-term environmental monitoring network with approximately 100 sites (see Figure 2-17 for a map of U.S. sites) located throughout the U.S. and Canada, managed and operated by the U.S. EPA in cooperation with other federal, state, and local partners (www.epa.gov/castnet).

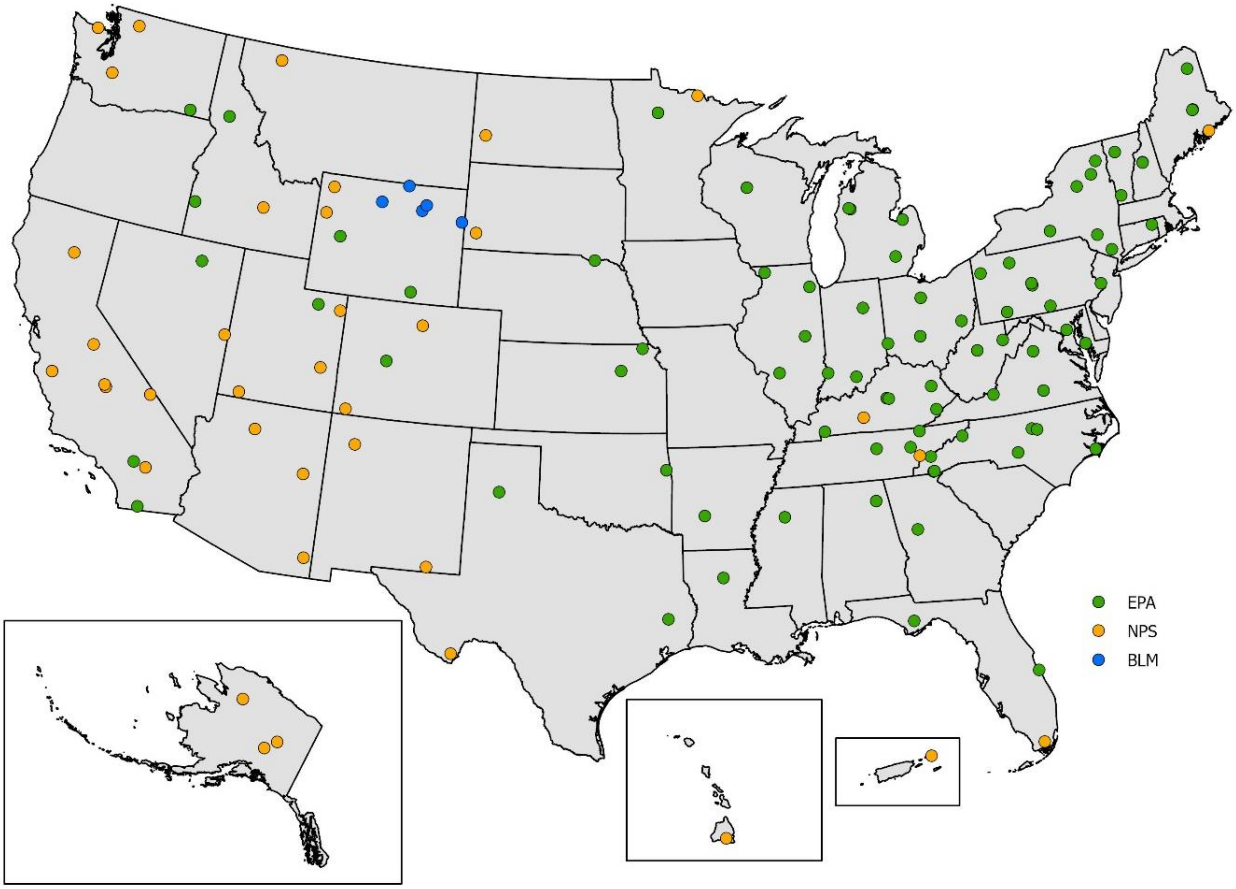

Figure 2-17. Location of CASTNET monitoring sites and the organizations responsible for collecting data. (NPS = National Park Service, BLM = Bureau of Land Management).

The CASTNET is the only network in the U.S. that provides a consistent, long-term data record of ambient air concentrations of S and N species that dry deposition fluxes can be estimated from. It complements the NTN, and nearly all CASTNET sites are collocated with or near an NTN site. Together, these two monitoring programs are designed to provide data necessary to estimate long-term temporal and spatial trends in total deposition (dry and wet). Species measured in CASTNET include: O_3, SO_2, HNO_3, nitrate, sulfate, and ammonium among others. Weekly ambient air concentrations of gases and particles are collected with an open-face 3-stage filter pack. Ozone measurements occur on an hourly basis. While CASTNET data are

more useful for estimating dry deposition than data from FRM networks, monitors are generally sparse and deposition is only determined for discrete locations. Also, not all of the species that contribute to total sulfur and nitrogen deposition are measured in CASTNET (Schwede et al., 2011). Despite these disadvantages, CASTNET data can still be very useful if used in combination with modeled estimates (Schwede et al., 2011), as discussed further in section 2.5.

The CASTNET has recently been reviewed by the EPA's Scientific Advisory Board with regard to its past functioning and current status, and to consider optimization of the network. A change in the distribution or number of sites or a shift in the instrument payload could affect our understanding of changes in deposition, potentially in response to new emission controls, as well as efforts to improve understanding of the link between air concentration and deposition. The Science Advisory Board released a draft letter of recommendations on October 11, 2023, which is available on its website (https://sab.epa.gov/ords/sab/f?p=100:18:130347838466:::18:P18_ID:2626).

There are differences in the measurement techniques that require careful consideration when used for analysis. The IMPROVE and CSN techniques are most efficient at collecting particles with a diameter smaller than 2.5 microns ($PM_{2.5}$), while the CASTNET samplers, which do not use size-selected inlets, also measure larger particles. This is relevant because larger particles are often from natural sources such as wind-blown soil, dust, or sea salt. Gas-phase nitric acid can condense onto these particles, forming particulate nitrate. Since these larger particles deposit quickly, this can be a significant portion of the total N deposition. However, as most CASTNET sites are located in rural areas, the expectation is that unless these sites are disproportionately impacted by local coarse particle sources (e.g., by sea salt in coastal areas), that most of the PM collected is $PM_{2.5}$. Furthermore, the timing of the measurements is not the same. CASTNET filter packs are deployed in the field for the entire 7-day measurement period, while IMPROVE and CSN are 24-hour measurements. Since ammonium nitrate is semi-volatile, and as temperature and humidity conditions change, these particles can evaporate off the filter as gas-phase ammonia and nitric acid. Each network deploys a different approach to minimizing these evaporative losses or capturing the volatilized nitrate and ammonia (Lavery et al., 2009). When collocated and compared to reference techniques, the correlation between these measurement techniques depends on meteorological conditions.

The NADP also maintains the Ammonia Monitoring Network (AMoN) which is collocated with CASTNET designed to capture long-term trends in ambient air NH_3 concentrations and deposition. There are currently 106 AMoN sites covering 34 states (see Figure 2-18). In part because CASTNET was developed to investigate drivers of acid rain, most AMoN sites are located in the Eastern USA. However, there are large NH_3 emission sources in the Midwest and Western USA that may not be sufficiently sampled with current AMoN

coverage. It is possible that satellite products for NH_3 concentration observations, such as the Cross-Track Infrared Sounder (Shephard et al., 2020) or Infrared Atmospheric Sounding Interferometers (Van Damme et al., 2021), may be used to infer NH_3 variability over these spatial gaps in the interim. The AMoN uses passive filter-based samplers which are deployed for two-week periods. Both gaseous ammonia and particle ammonium concentrations are measured.

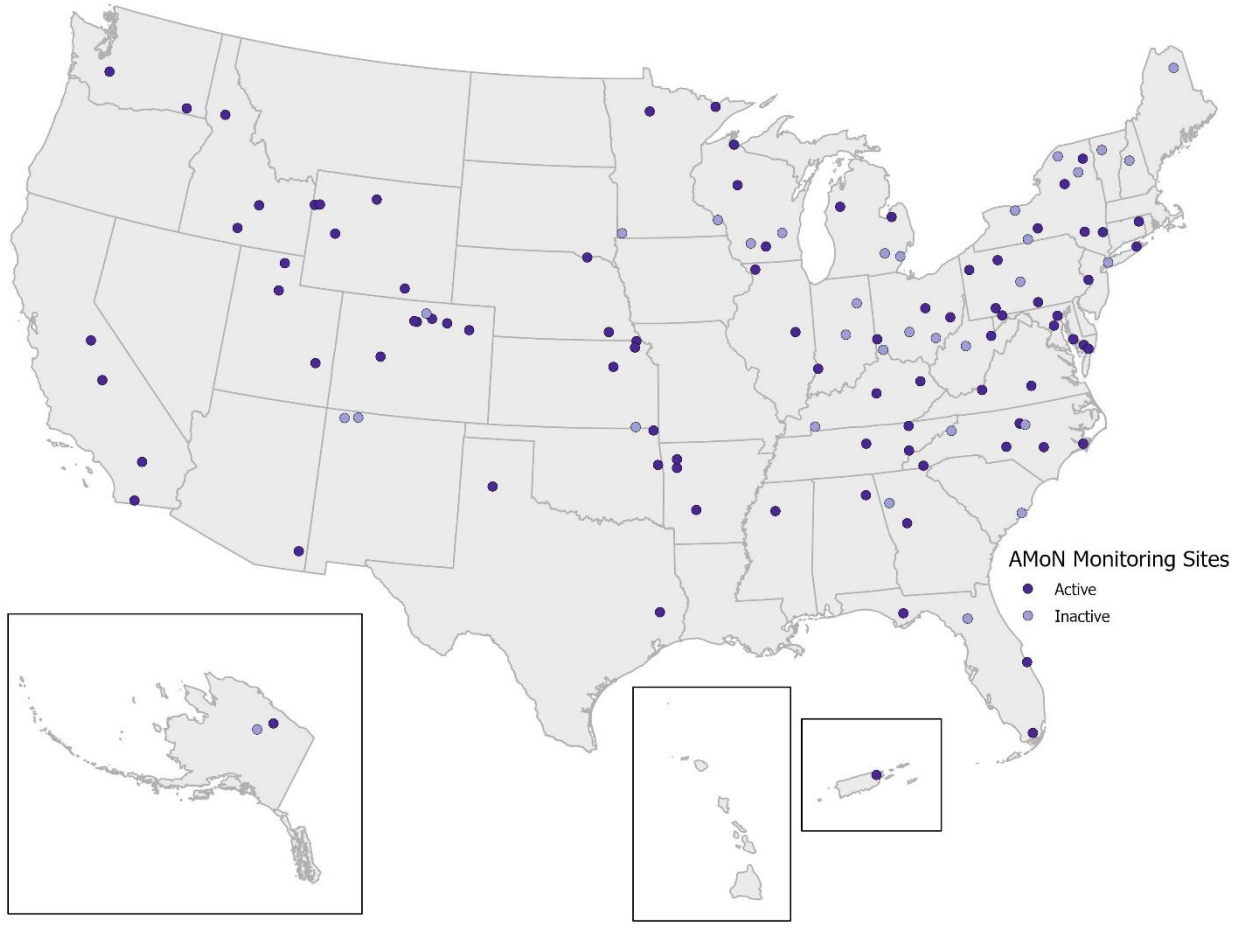

Figure 2-18. Location of AMoN monitoring sites with sites active shown in dark blue and inactive sites in light blue. (There is an additional site in AK not shown here.)

2.3.5 Satellite Retrievals

Satellite retrievals, field studies and aircraft campaigns (the latter two discussed in the next section) complement the regulatory networks for investigation into the variability, trends and drivers of N, S and PM. Satellite retrievals, in particular, provide a spatially expansive, long-term record that can bridge gaps between ground monitors and offer insight into species' trends over time.

Each of NO_2, SO_2, NH_3 and $PM_{2.5}$ are measured by existing satellites, such as MODIS and OMI for NO_2, IASI and CRIS for NH_3 and MODIS, CALIPSO, GOES-R and GOES-S, among others, for $PM_{2.5}$ via aerosol optical depth. While deposition is not measured directly,

satellite retrievals have been combined with model simulations to map deposition distributions (e.g., Kharol et al. 2018, which illustrated a shift in the dominant form of nitrogen deposition, from oxidized to reduced, over the continental U.S.). The spatial distributions of these species generally reflect our understanding based on ground measurements (e.g., Nowlan et al., 2014), lending confidence to the potential for satellite measurements to investigate variability in atmospheric composition (ISA, Appendix 2, section 2.4.2.2 and section 2.4.4.2). There has been substantial progress in improving retrieval algorithms to confidently infer a lower limit of detection, and upcoming geostationary satellite missions such as MAIA, TEMPO and TropOMI will increase the spatiotemporal resolution of concentration retrievals to improve capacity and confidence in satellite inference of species variability.

2.4 RECENT AMBIENT AIR CONCENTRATIONS AND TRENDS

2.4.1 NO_2 Concentrations and Trends

The secondary NO_2 NAAQS is the annual mean concentration, with a level of 53 ppb. There are two primary NO_2 NAAQS. One is the 98th percentile of the 1-hour daily maximum concentrations averaged over 3 years, with a level of 100 parts per billion (ppb). The other is the annual mean concentration, with a level of 53 ppb. As shown in Figures 2-19 and 2-20, there are no locations with NO_2 design values[7] in violation of these standards during the 2019-2021 period. In this period, the highest NO_2 concentrations mostly occurred in urban areas across the western U.S. (e.g., Los Angeles, Phoenix, Las Vegas, Denver). The maximum design value for the 1-hour standard during the 2019-2021 period was 80 ppb, while the annual mean design value for 2021 was 30 ppb. Both maximum design values occurred at near-road sites in the Los Angeles metropolitan area; this area has historically had some of the highest NO_2 concentrations in the U.S. For the 2019-2021 period, the mean average hourly NO_2 value, across valid monitoring sites, was 16.3 ppb.

Nitrogen dioxide concentrations have been declining across the U.S. for decades, in response to cleaner motor vehicles, emissions reductions at stationary fuel combustion sources, and economic factors. For example, in Los Angeles metropolitan area annual NO_2 design values were almost twice as high in the early 1980's (U.S. EPA, 1985). Figures 2-21 and 2-22 show the trends in the annual 98th percentile of the daily maximum 1-hour NO_2 concentrations and in the annual mean NO_2 concentrations across the U.S. going back to 1980. The trends are sharply downward for both NO_2 metrics. At the beginning of the trends record, it was not uncommon for

[7] A design value is a statistic that describes the air quality status of a given location relative to the level of the NAAQS. Design values are typically used to designate and classify nonattainment areas, as well as to assess progress towards meeting the NAAQS. Design values are computed and published annually by EPA (https://www.epa.gov/air-trends/air-quality-design-values).

locations to exceed the NO_2 NAAQS, especially the standard with the shorter averaging time. However, the last violations of the NO_2 annual standard occurred in 1991. Over the past decade, the downward trends in NO_2 levels across the U.S. have continued, but at a slower rate than what was experienced from 1980 to 2010. Given that deposition-related impacts can adversely affect ecosystems (forests/trees, streams/fish) over the course of decades (as discussed in more detail in Chapter 5 of this assessment), it is important to recognize that effects of the high NO_2 levels observed in 1980, and preceding decades when NO_2 levels were even higher, may still be impacting ecosystem health. Figure 2-23 indicates dramatic changes in HNO_3 concentrations between 1990s and 2019. Prior to 1980, the monitoring networks were somewhat sparser, but NO_2 data exist for certain cities. The EPA's very first Trends Report (U.S. EPA, 1973) reported annual average NO_2 values in five U.S. cities for the 1967-1971 period. At that time, annual average NO_2 concentrations averaged 75 ppb over the cities where data existed (i.e., off the chart of the 1980-2021 trend shown in Figure 2-22). See Table 2-1 for a summary of these older NO_2 annual means.

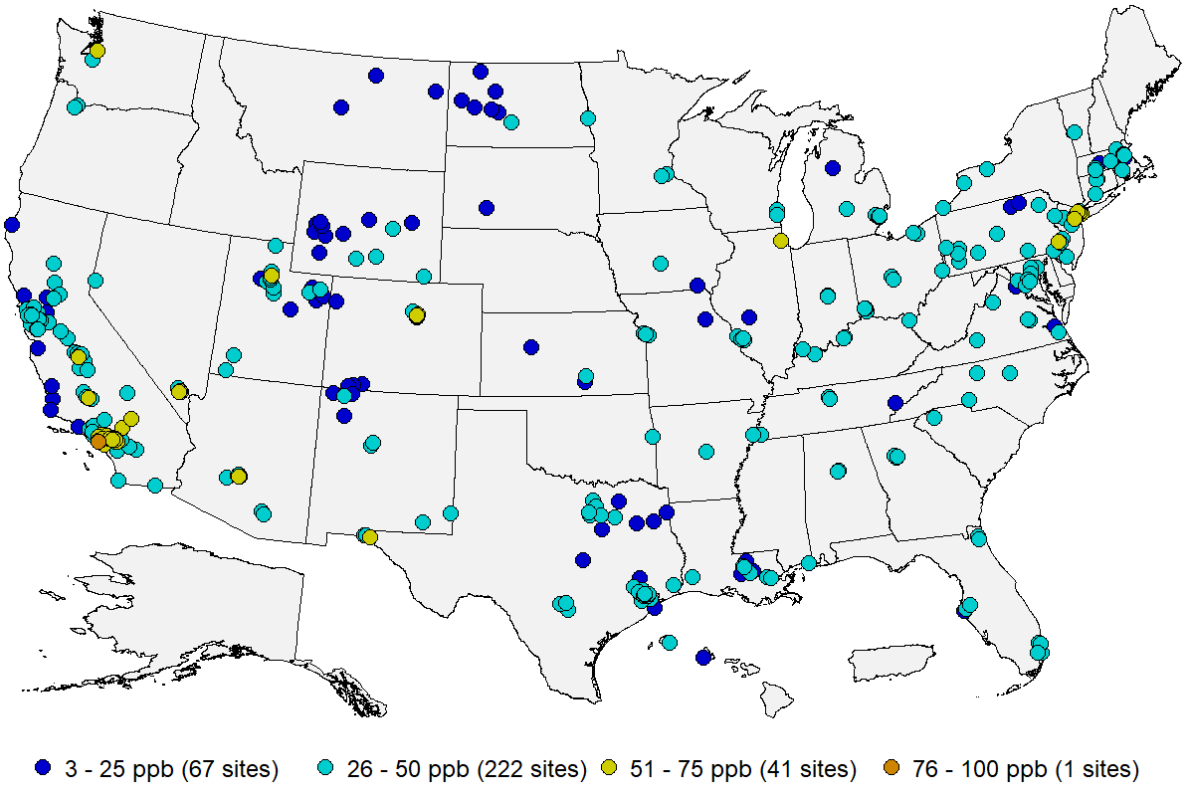

Figure 2-19. Design values for the 1-hour primary NO_2 NAAQS (98th percentile of daily maximum 1-hour concentrations, averaged over 3 years; ppb) at monitoring sites with valid design values for the 2019-2021 period.

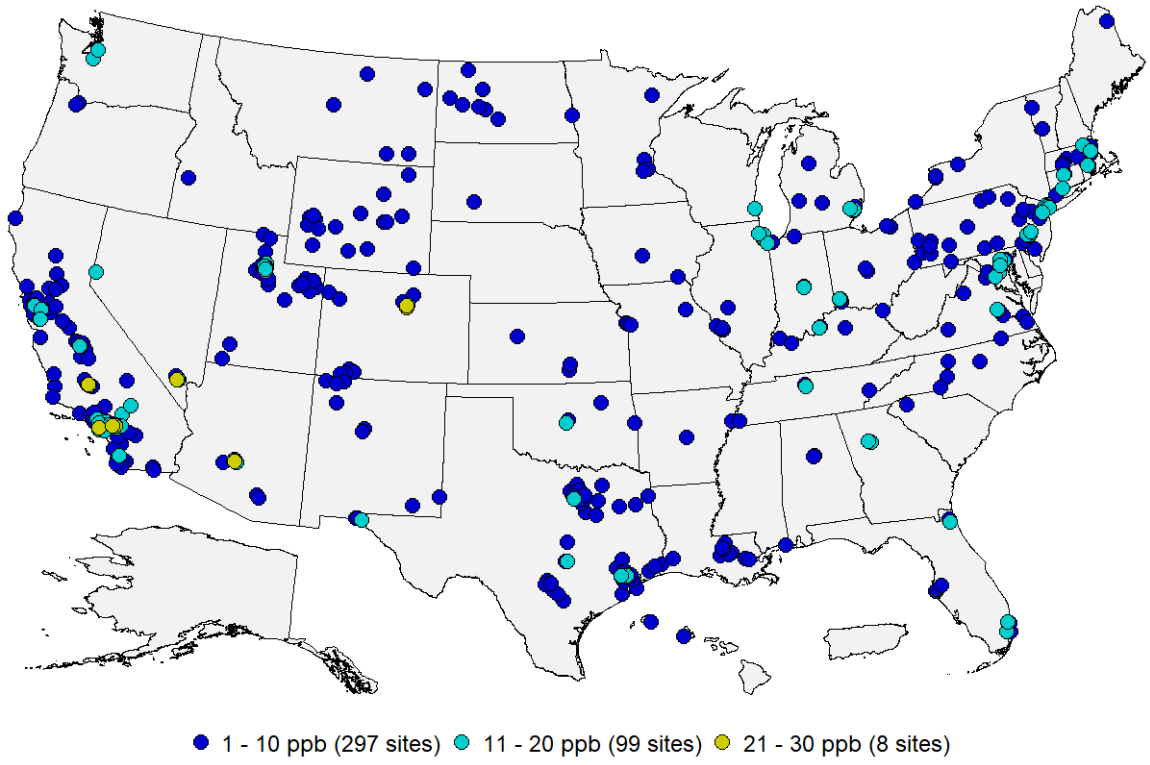

Figure 2-20. Primary and secondary NO_2 annual design values for 2021.

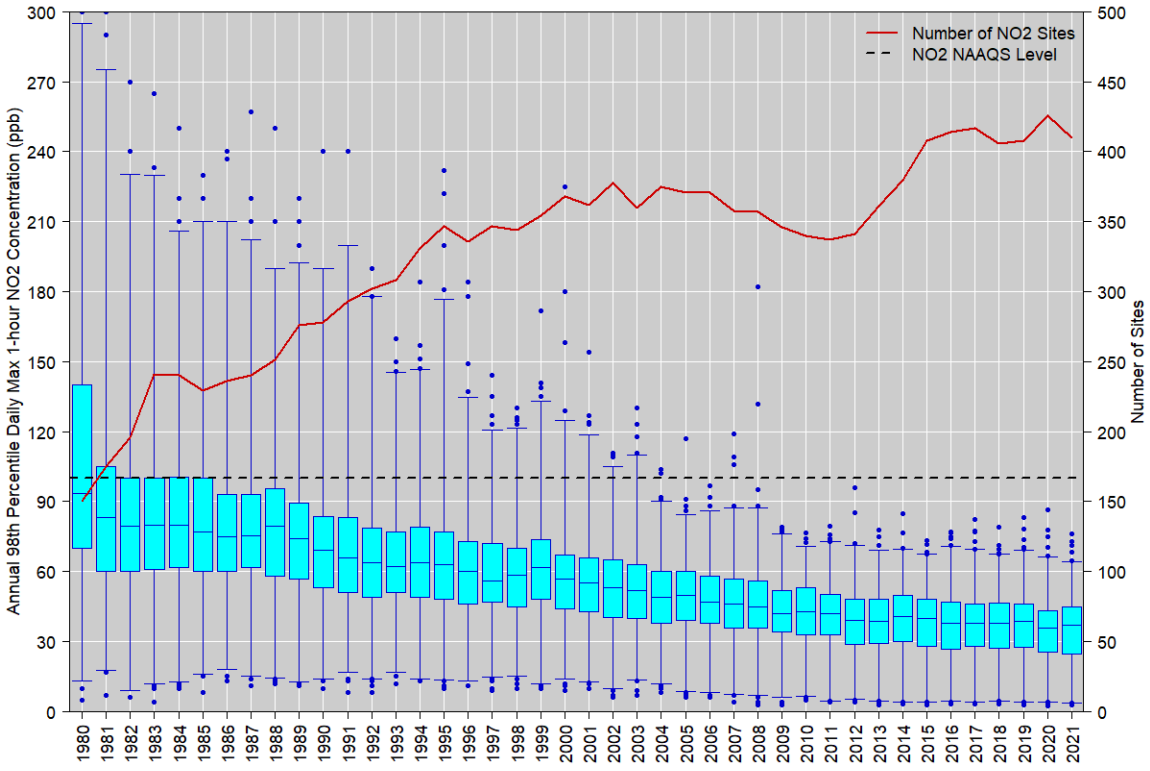

Figure 2-21. Distributions of annual 98th percentile, maximum 1-hour NO_2 values at U.S. sites. The red line shows number of sites in each boxplot per year.

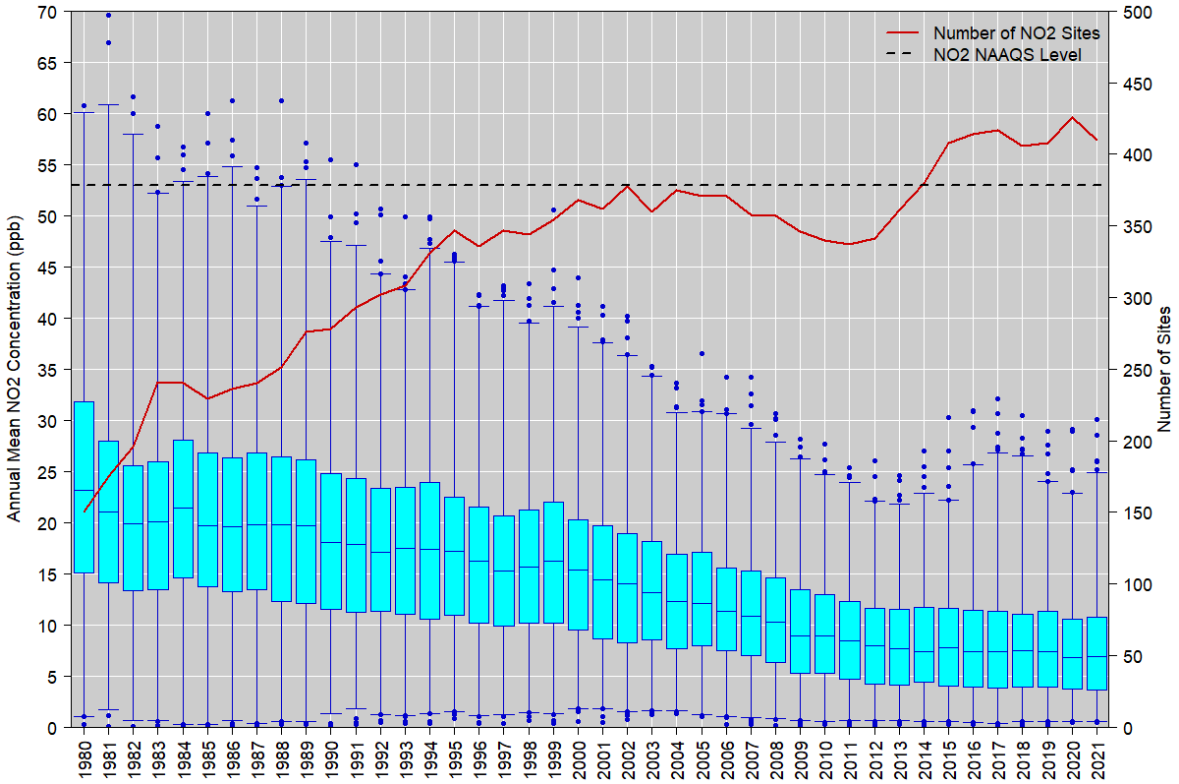

Figure 2-22. Distributions of annual mean NO_2 values at U.S. sites. The red line shows number of sites in each boxplot per year.

Table 2-1. Average annual mean NO_2 concentration in 1967-1971 in select cities.

Location	1967-1971 Annual Mean NO_2 Concentration (ppb)
Chicago	120.5
Cincinnati	60.4
Denver	65.1
Philadelphia	76.1
St. Louis	54.1
5-city average	75.3

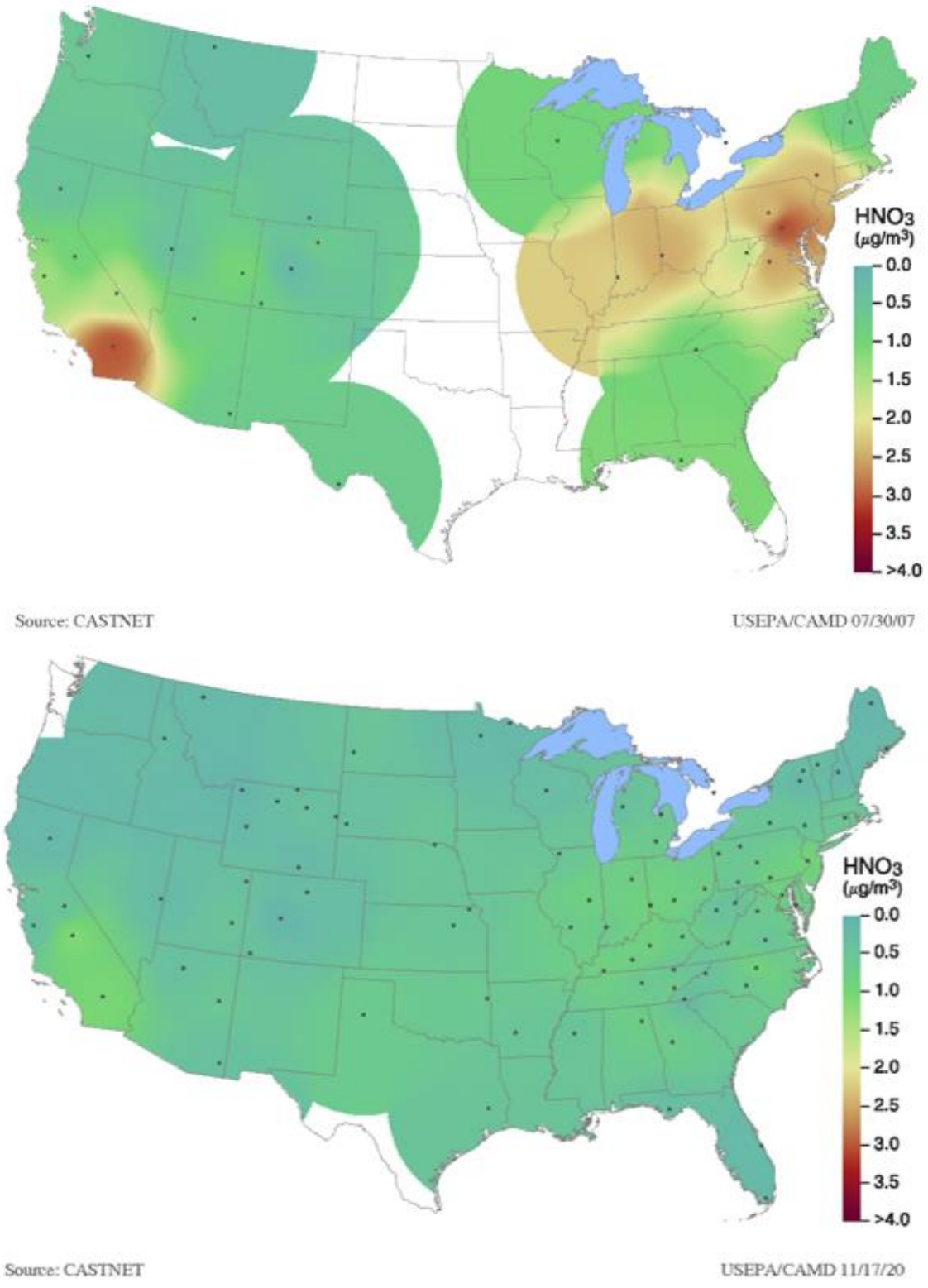

Figure 2-23. Annual average concentrations of HNO_3 in: 1996 (top) and 2019 (bottom).

2.4.2 SO_2 Concentrations and Trends

The secondary SO_2 standard is the 3-hour average concentration, with a level of 0.5 ppm (500 ppb), not to be exceeded more than once per year. The primary SO_2 standard is the 99th percentile of daily maximum 1-hour concentrations, averaged over 3 years, with a level of 75 ppb. As shown in Figure 2-24, for the 2019-2021 period, there were 15 locations with SO_2 design values in violation of the primary SO_2 standard. The maximum design value was 376 ppb at a monitoring site near an industrial park in southeast Missouri. The sites with design values exceeding the NAAQS in Hawaii are due to natural SO_2 emissions from recurring volcanic eruptions. Both peak and mean SO_2 concentrations are higher at source-oriented monitoring sites than non-source sites. Mean hourly SO_2 concentrations during 2019-2021 are 3 ppb (5.1 ppb at source-oriented sites, 1.6 ppb at urban non-source sites, and 0.9 ppb at rural non-source sites).

Figure 2-25 displays the annual second highest 3-hour average SO_2 concentrations (design values for the existing secondary standard) across the U.S. in 2021. The values at all sites with valid secondary SO_2 design values were less than the 500 ppb level and the vast majority of sites had design values that were less than 20 ppb. Like concentrations of NO_2, SO_2 concentrations have been declining across the U.S. for decades, primarily in response to emissions reductions at stationary fuel combustion sources.

Figure 2-26 shows the downward trend in design values for the primary SO_2 NAAQS over the past 40 years. The last year in which the 3-year average of the annual 99th percentile daily maximum 1-hour concentrations was greater than 75 ppb is 1994. Since then, the entire distribution of values has continued to decline such that the median value across the network of sites is now less than 10 ppb. Additional sites were added to the network in 2017 near major industrial sources of SO_2 and this likely caused the slight increase in the median concentration observed in 2017. Figure 2-27 shows the sharp downward trend in secondary SO_2 concentrations across the U.S. Again, the highest values in the distribution in recent years are from the sites near industrial sources. Figure 2-28 shows trends in annual average SO_2 concentrations, with an overall decline from 2000-2021. Additionally, Figure 2-29 presents scatterplots of annual average SO_2 concentrations (averaged over three years) and primary and secondary standard design values at SLAMS across the U.S. for the same time period.

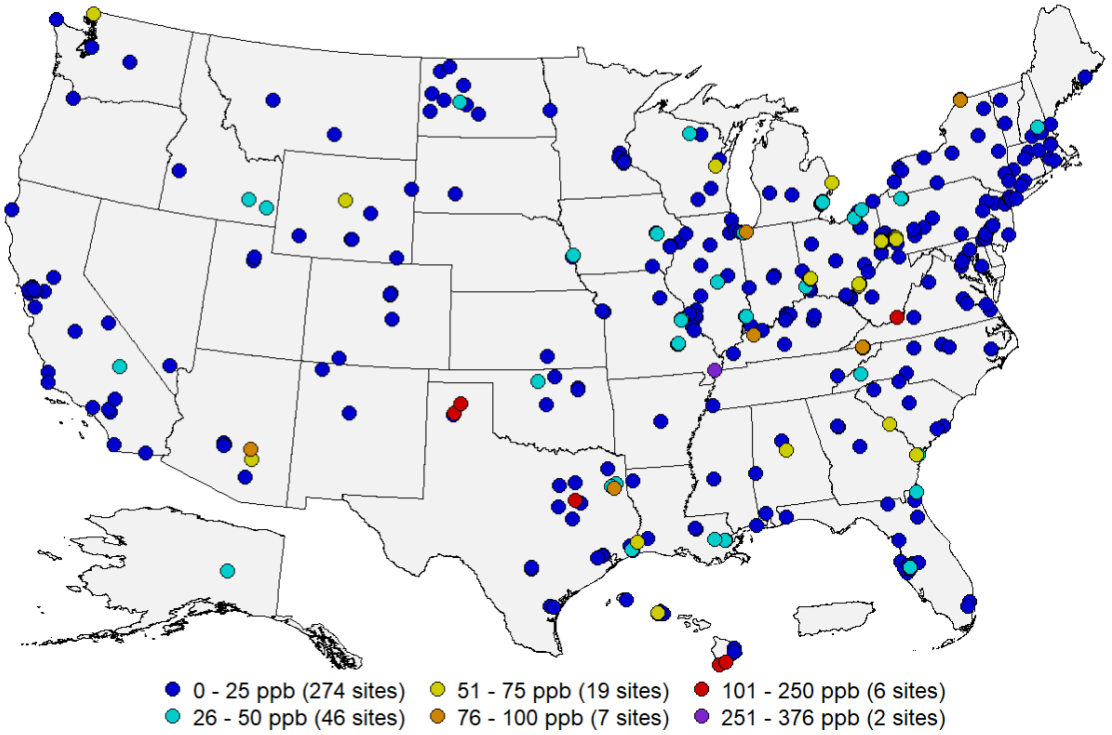

Figure 2-24. Primary SO_2 standard design values (99th percentile of 1-hour daily maximum concentrations, averaged over 3 years) for the 2019-2021 period at monitoring sites with valid design values.

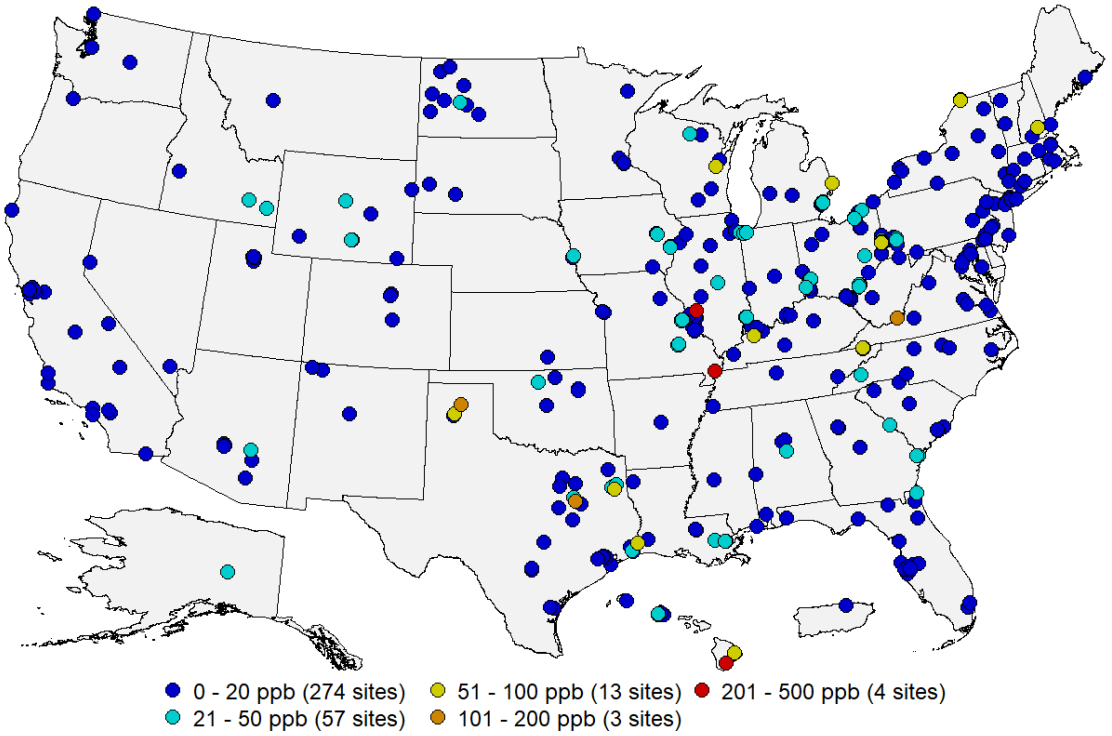

Figure 2-25. Secondary SO_2 standard design values (2nd highest 3-hourly average) for the year 2021 at monitoring sites with valid design values.

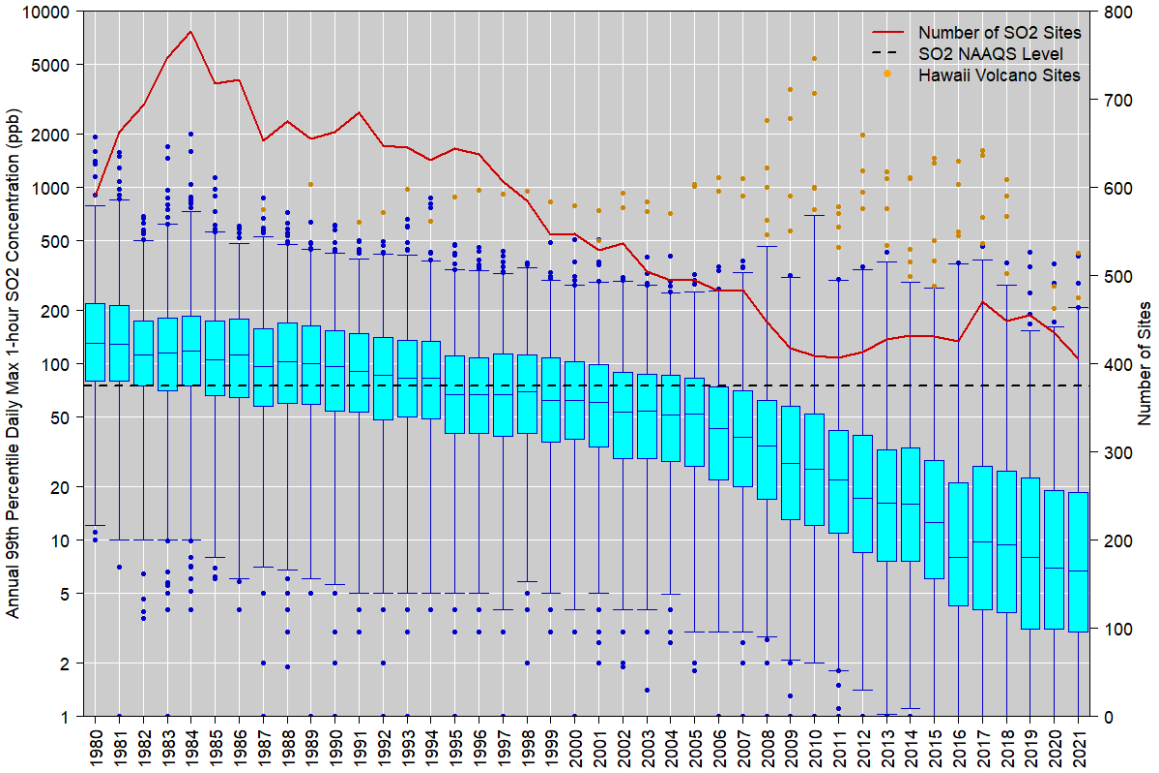

Figure 2-26. Distributions of 99th percentile of maximum daily 1-hour SO_2 design values at U.S. sites (1980-2021). The red line shows number of sites in each boxplot per year. Orange dots represent design values in Hawaii determined to have been influenced by volcanic emissions. Note: the y-axis is in logarithmic scale.

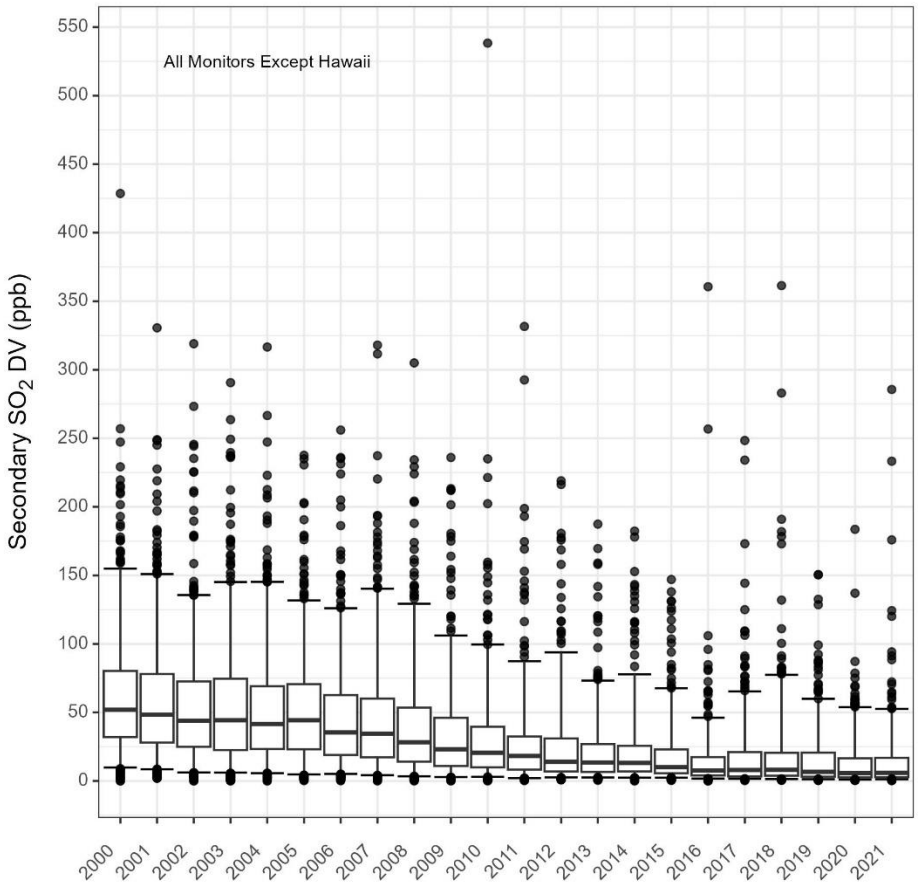

Figure 2-27. Distributions of secondary SO_2 standard design values at U.S. sites, excluding sites in Hawaii (2000-2021).

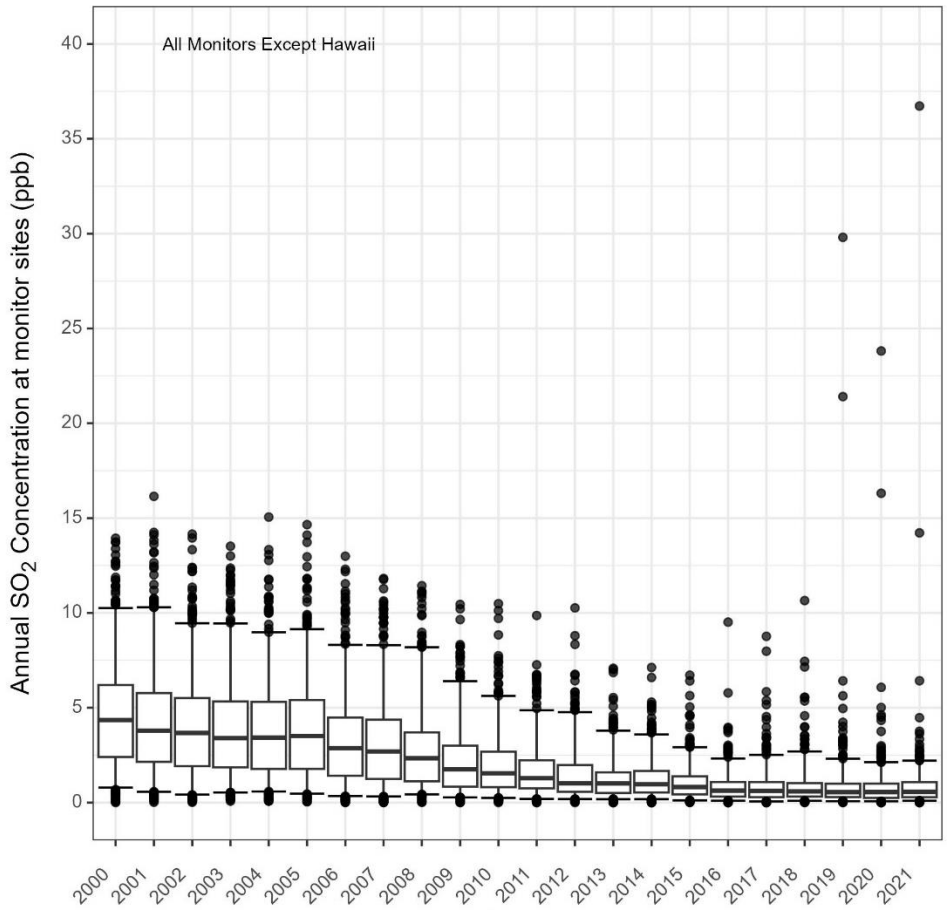

Figure 2-28. Distribution of annual average SO_2 concentrations (ppb) at SLAMS in the U.S., excluding Hawaii (2000-2021).

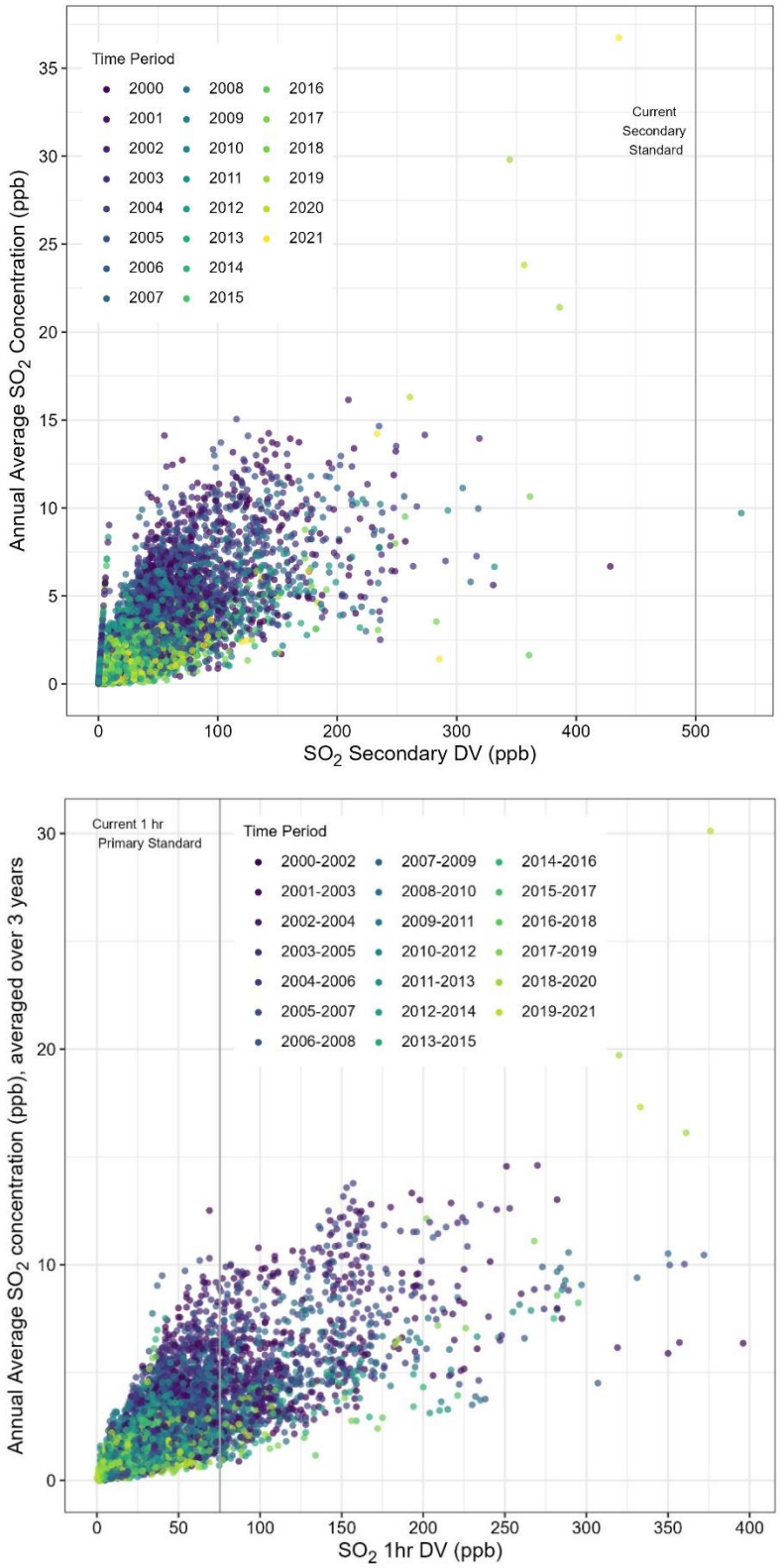

Figure 2-29. Relationship of annual SO_2 concentrations, averaged across three years, to design values for the current 3-hr secondary standard (upper) and the 1-hr primary standard (lower) at SLAMS (2000-2021). Sites in Hawaii excluded.

2.4.3 $PM_{2.5}$ Concentrations and Trends

There are two primary and two secondary standards for $PM_{2.5}$. There are standards in terms of annual means, averaged over 3 years, with levels at 12.0 µg/m^3 (primary standard) and 15.0 µg/m^3 (secondary standard). There are also 24-hour standards in terms of the 98th percentile of daily $PM_{2.5}$ values, averaged over 3 years, with a level of 35 µg/m^3(for both the primary and secondary standards). As discussed in section 2.1, $PM_{2.5}$ is a mixture of substances suspended as small liquid and/or solid particles. Figure 2-30 displays a map with pie charts showing the major $PM_{2.5}$ species as a fraction of total $PM_{2.5}$ mass as measured at selected NCore, CSN, and IMPROVE sites during the 2019 to 2021 period. The six species shown are SO_4^{2-}, NO_3^-, elemental carbon (EC), organic carbon (OC), crustal material, and sea salt. The mix of $PM_{2.5}$ components can vary across the U.S. For example, in the Appalachian region, the predominant contributor to total $PM_{2.5}$ mass is sulfate. Conversely, in the upper Midwest, the largest component term tends to be NO_3^-. This regional variability in $PM_{2.5}$ composition has implications for the spatial nature of N and S deposition.

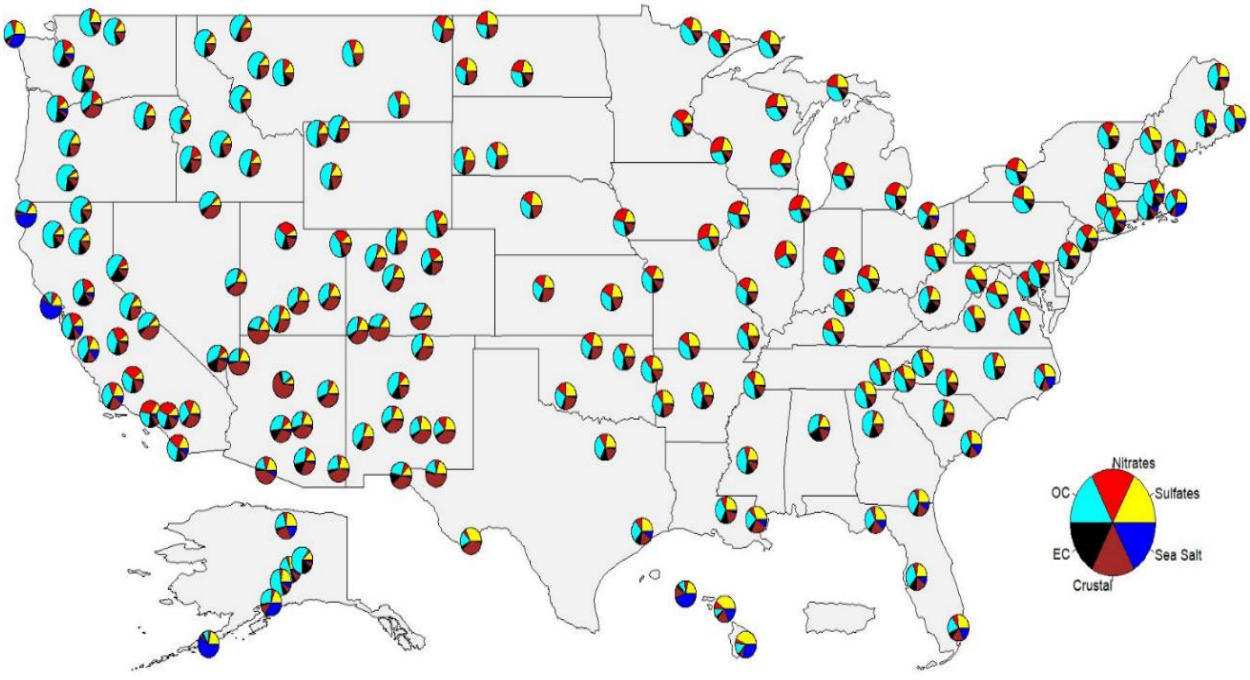

Figure 2-30. Map showing pie charts of $PM_{2.5}$ component species at selected U.S. monitoring sites based on 2019-2021 data. Note: total $PM_{2.5}$ mass may vary from site to site.

Figures 2-31 and 2-32 show maps of the annual and 24-hour $PM_{2.5}$ design values,[8] respectively, at U.S. ambient air monitoring sites based on monitoring data from the 2019-2021 period. All sites in the eastern U.S. were meeting the annual primary and secondary $PM_{2.5}$ NAAQS of 12.0 µg/m^3 and 15.0 µg/m^3, and the 24-hour primary and secondary $PM_{2.5}$ NAAQS of 35 µg/m^3 during this period. Many sites in the western U.S. were still violating the 24-hour $PM_{2.5}$ NAAQS in 2019-2021, while a smaller number of sites, mostly in California, were also violating the annual $PM_{2.5}$ NAAQS (28 sites exceed the primary NAAQS level of 12.0 µg/m^3, and 9 sites exceed the secondary annual $PM_{2.5}$ NAAQS level of 15.0 µg/m^3). It should be noted that large areas of the western U.S. were impacted by smoke from wildfires in 2020 and 2021 and these smoke-impacted concentrations are included in the 2019-2021 data shown here. The highest annual $PM_{2.5}$ design values are located in the San Joaquin Valley of California, while the highest 24-hour $PM_{2.5}$ design values are located in Mono County, California, which was heavily impacted by wildfire smoke in 2020.

Figures 2-33 and 2-34 display the average NO_3^- and SO_4^{2-} concentrations over the U.S. during the period 2019-2021. As discussed above, SO_4^{2-}concentrations are highest in the Ohio River valley and along the Gulf of Mexico, whereas NO_3^-concentrations are highest in the upper Midwest, along the northeast urban corridor, and in parts of California. Figures 2-35 and 2-36 show trends in annual average concentrations for NO_3^- and SO_4^{2-}based on sites that collected data for at least 12 out of 16 years from 2006 to 2021. Broad national reductions in NO_X emissions have resulted in substantial decreasing trends in NO_3^-concentrations in most of the U.S., especially in areas where NO_3^-concentrations were historically highest. Similarly, reductions in SO_2 emissions have resulted in significant reductions in SO_4^{2-}concentrations nationally and especially in the eastern U.S. National, annual average $PM_{2.5}$ concentrations have declined despite the relatively consistent trend in NH_3 emissions. While not shown here, trends in other $PM_{2.5}$ components like EC and OC were more variable, with some sites showing substantial decreases and the remaining sites having no clear trend. Ammonium sulfate and ammonium nitrate make up less than one-third of the $PM_{2.5}$ mass at the majority of sites and only a few sites have more than half of the $PM_{2.5}$ mass from these compounds.

There are also NAAQS for PM_{10} (24-hour primary and secondary standards, both with a level of 150 µg/m^3 that is not to be exceeded more than once per year, averaged over three years). While $PM_{2.5}$ mass is composed mainly of sulfates, nitrates, and other organic matter that can contribute to ecosystem impacts (ISA, Appendix 2, section 2.1), $PM_{10–2.5}$ is mostly composed of crustal material as well as sea salt in coastal areas. There is little discussion of

[8] The annual design value for both primary and secondary standards is an annual mean, averaged over 3 years. The 24-hour design value for both standards is the annual 98th percentile 24-hour average concentration, averaged over 3 years.

$PM_{10-2.5}$ effects in this document because these particles have faster settling velocities and the composition of this mass is expected to have less impact on deposition-related welfare impacts.

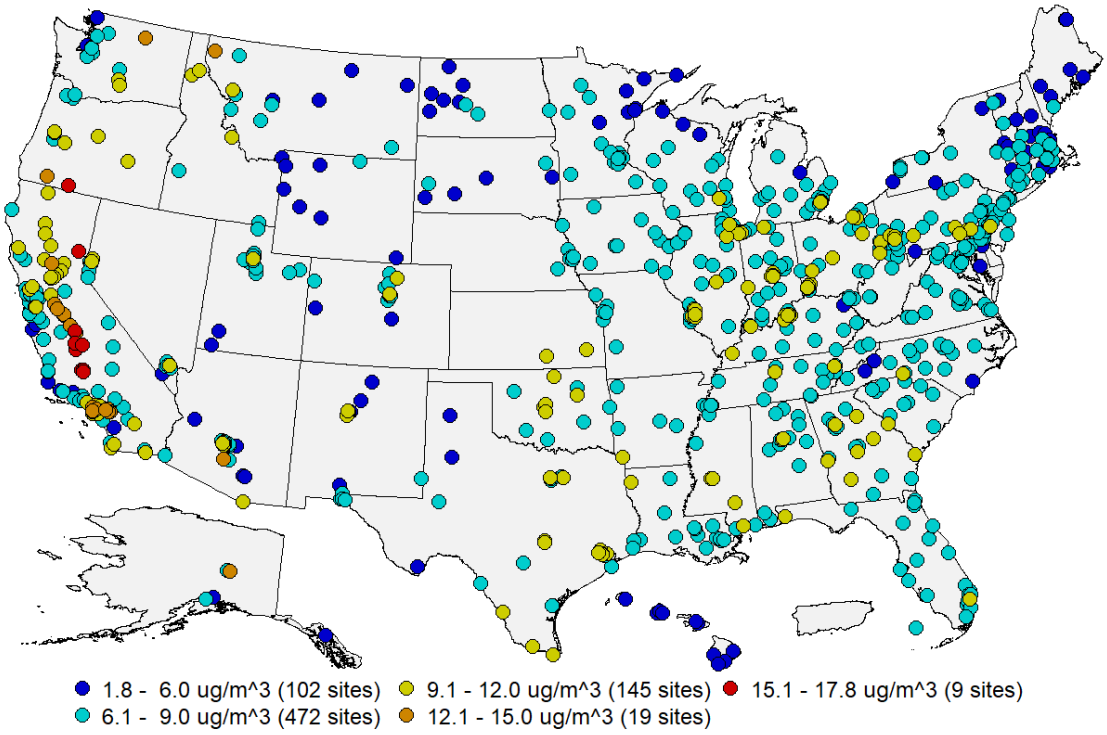

Figure 2-31. Primary and secondary annual $PM_{2.5}$ standard design values (2019-2021).

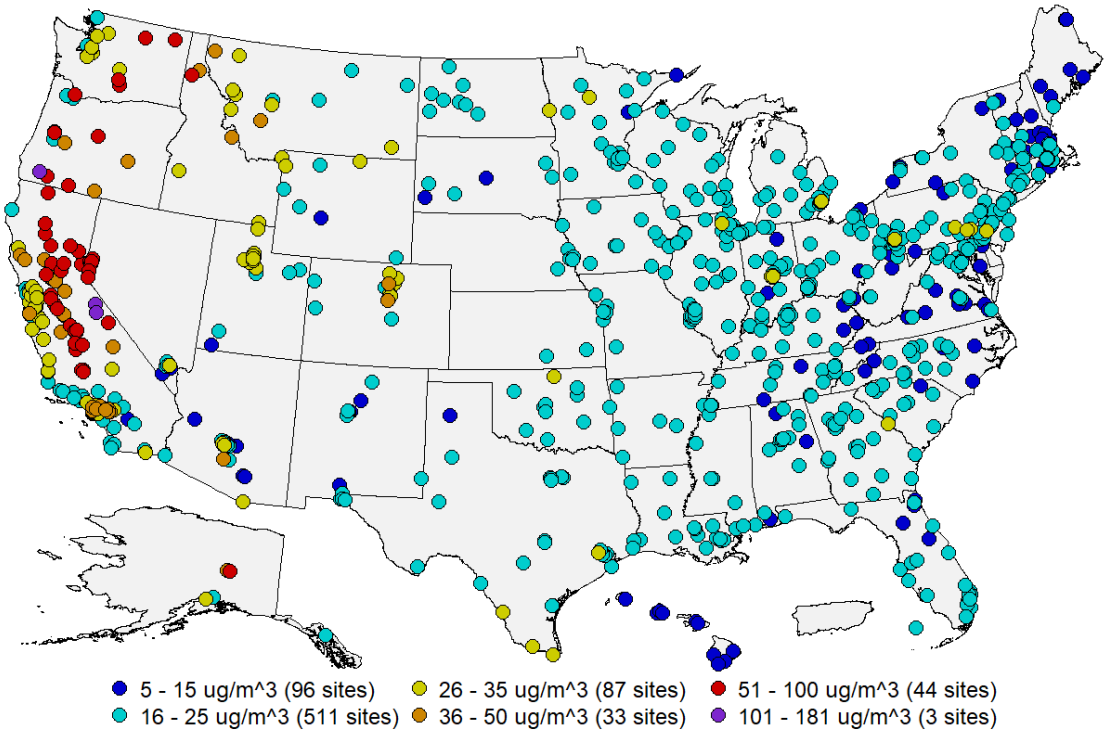

Figure 2-32. Primary and secondary 24-hour $PM_{2.5}$ design values (2019-2021 period).

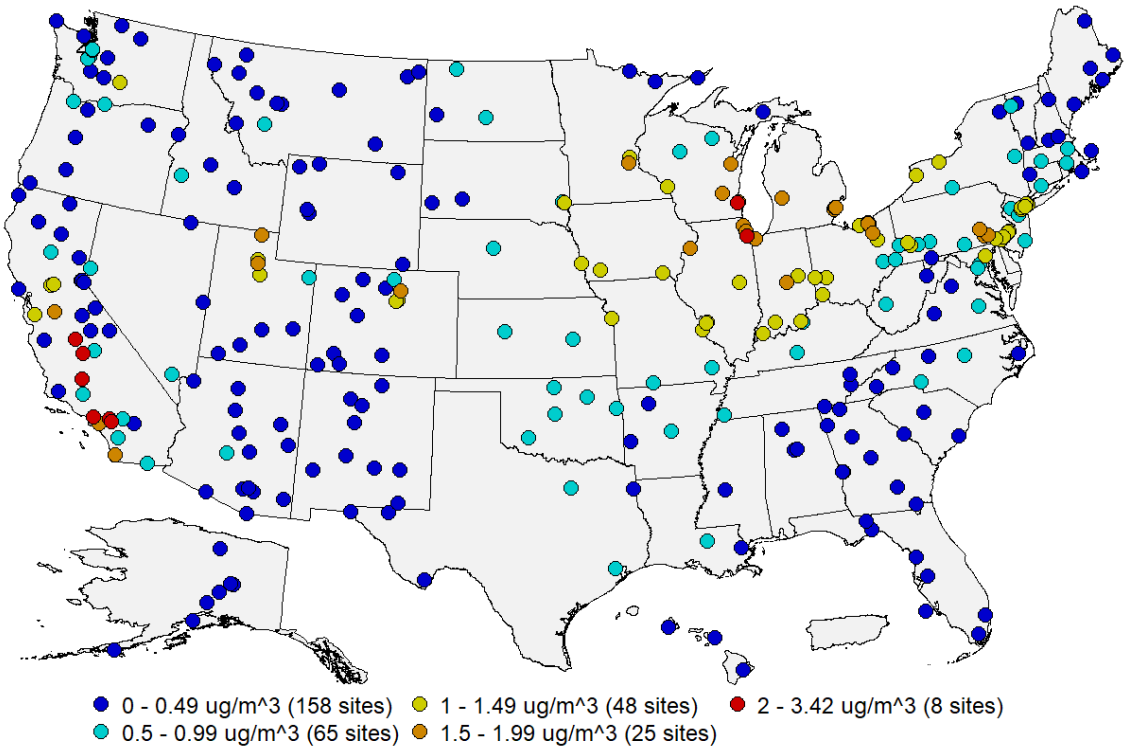

Figure 2-33. Annual average NO_3^- concentrations (µg/m^3) as measured at selected NCore, CSN, and IMPROVE sites for the 2019-2021 period.

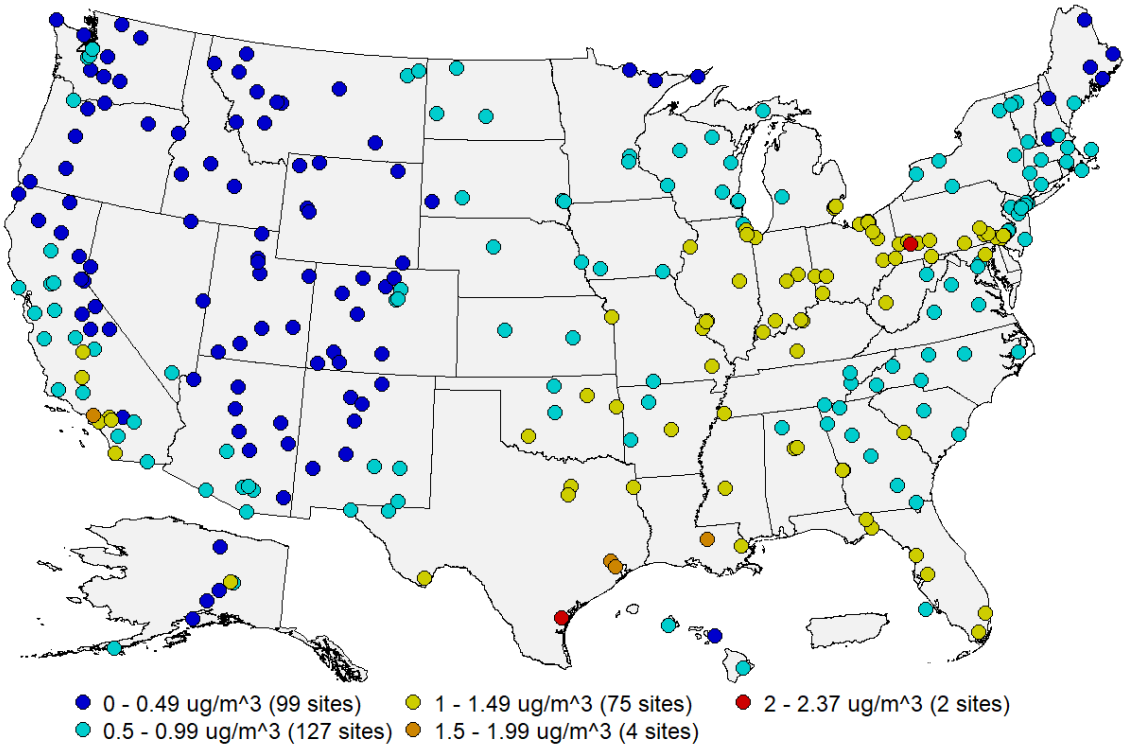

Figure 2-34. Annual average SO_4^{2-} concentrations (µg/m^3) as measured at selected NCore, CSN, and IMPROVE sites for the 2019-2021 period.

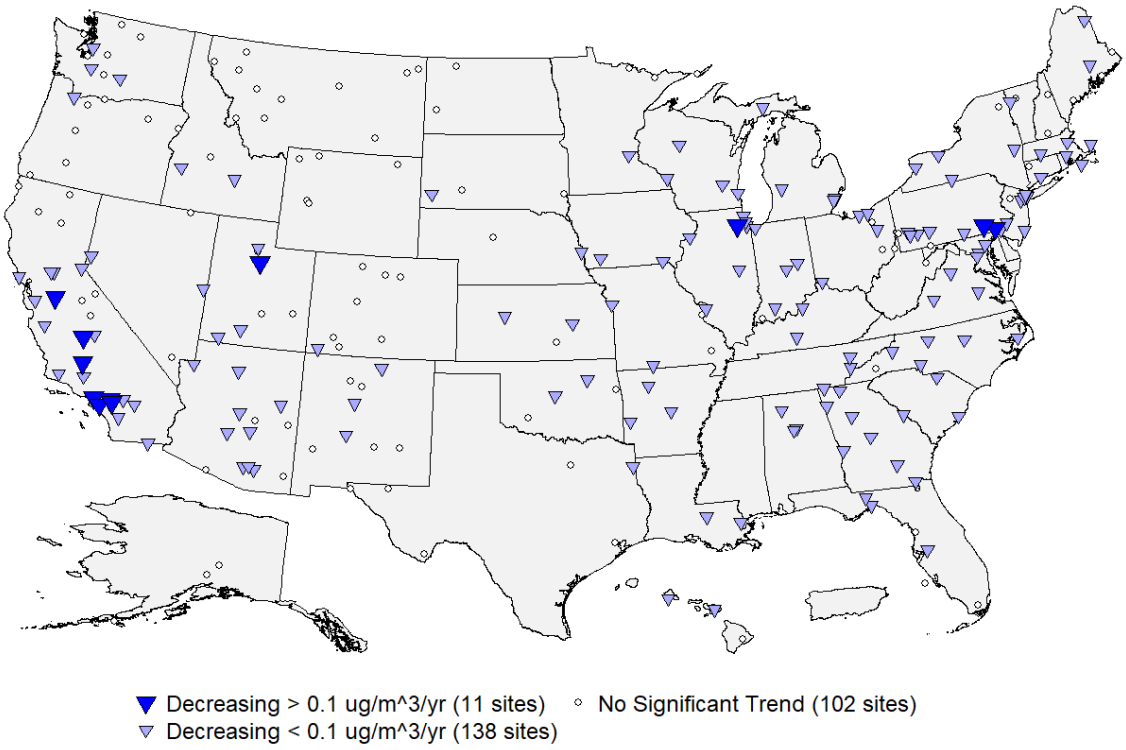

Figure 2-35. Trends in annual average concentrations for nitrate (NO_3^-) as measured at selected NCore, CSN, and IMPROVE sites from 2006 through 2021.

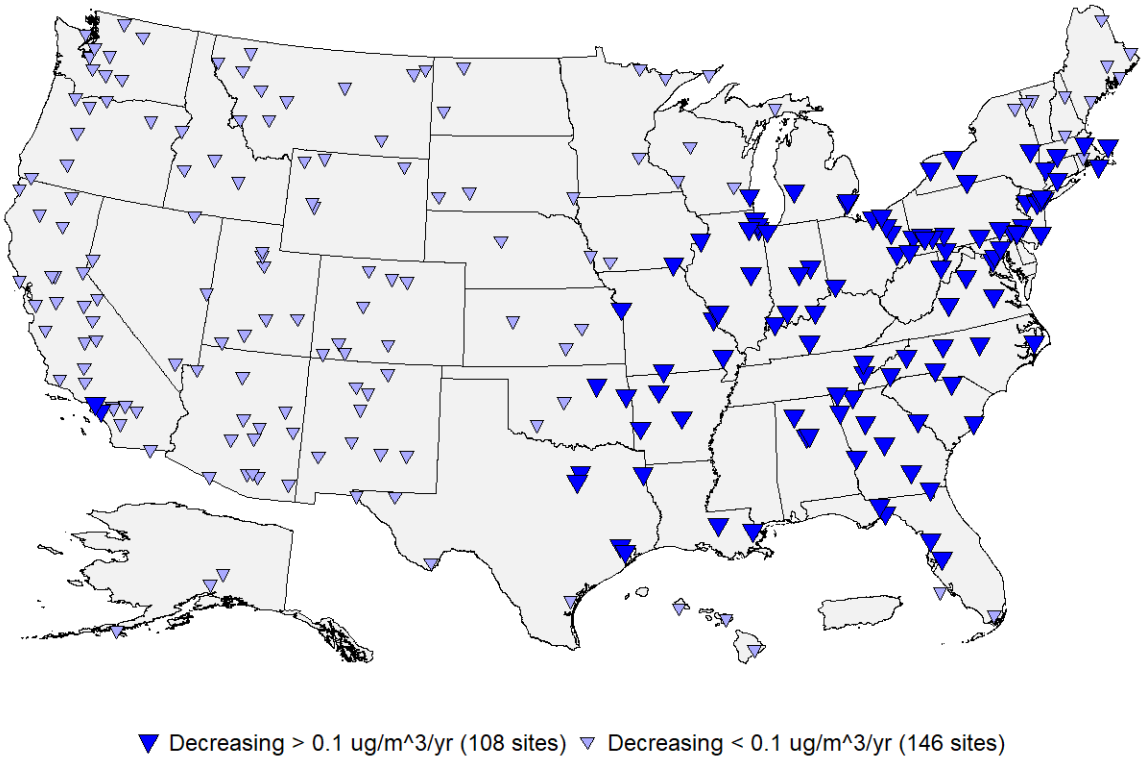

Figure 2-36. Trends in annual average concentrations for sulfate (SO_4^{2-}) as measured at selected NCore, CSN, and IMPROVE sites from 2006 through 2021.

The trends in total $PM_{2.5}$ mass between 2000 and 2021 are shown in Figures 2-37 (annual standard) and 2-38 (24-hour standard). These plots show the national distribution of $PM_{2.5}$ concentrations, along with the number of $PM_{2.5}$ monitoring sites reporting data in each year. The median of the annual average $PM_{2.5}$ concentrations decreased by 38 percent, from 12.8 µg/m^3 in 2000 to 8 µg/m^3 in 2021. Similarly, the median of the annual 98th percentile 24-hour $PM_{2.5}$ concentrations decreased by 35 percent, from 32 µg/m^3 in 2000 to 21 µg/m^3 in 2021. Both the annual average and 98th percentile 24-hour $PM_{2.5}$ concentrations decreased steadily from the early 2000s until 2016, and have fluctuated in recent years, especially in the upper tail of the distribution. These fluctuations are largely due to large-scale wildfire events that have occurred in recent years. The size of the $PM_{2.5}$ monitoring network increased rapidly following the establishment of a $PM_{2.5}$ NAAQS in 1997, and the network has been relatively stable at around 1,200 sites since 2002.

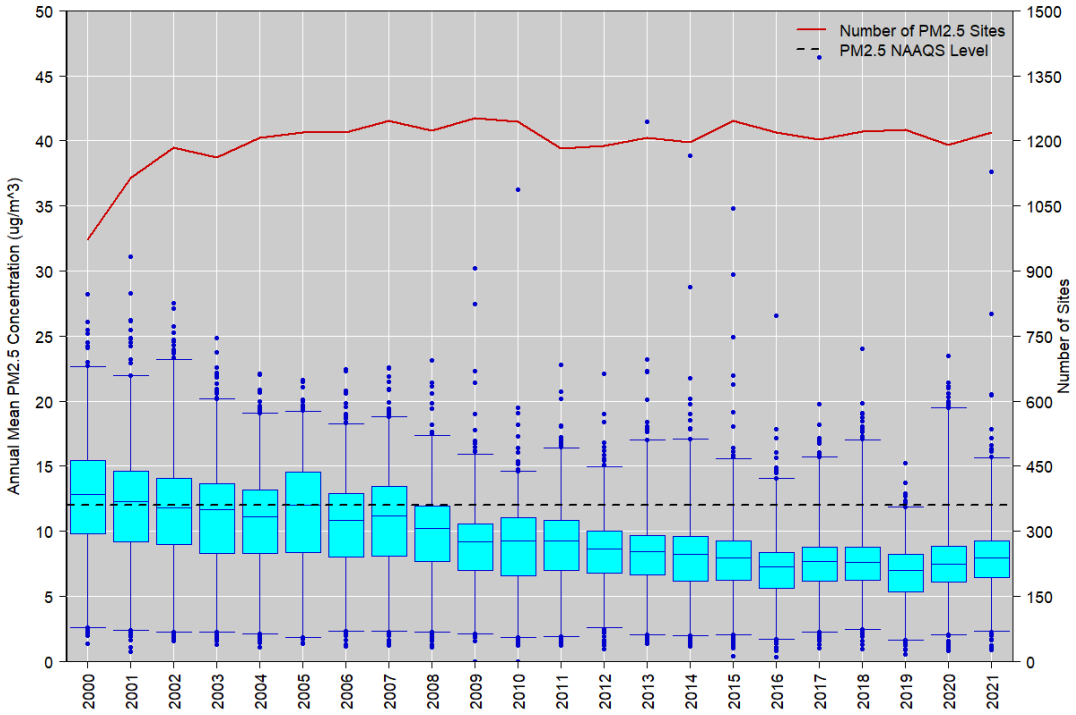

Figure 2-37. Distributions of annual mean $PM_{2.5}$ design values (µg/m^3) at U.S. sites across the 2000-2021 period. The red line shows the number of sites included in each boxplot per year.

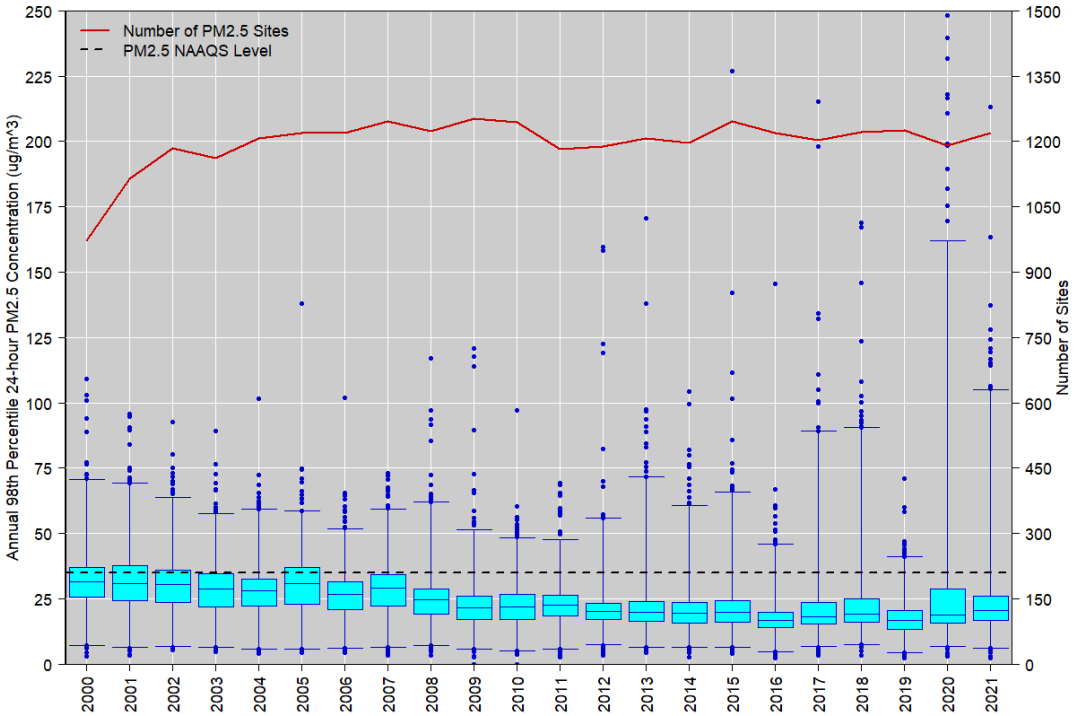

Figure 2-38. Distributions of the annual 98th percentile 24-hour $PM_{2.5}$ design values ($\mu g/m^3$) at U.S. sites across the 2000-2021 period. The red line shows the number of sites included in each boxplot per year.

2.4.4 Ammonia Concentrations and Trends

The AMoN network has collected measurements of ammonia gas since 2010 (NADP, 2012) and the number of sites within the network has increased over time. Figure 2-39 compares observed NH_3 concentrations between 2011 and 2020. The highest observed ammonia concentrations across the U.S. tend to occur in the central U.S. where values can exceed 2.4 $\mu g/m^3$. Consistent with expectations from the slightly increasing trends in ammonia emissions, we also see increases in NH_3 concentrations over this 10-year period over many parts of the country, although there can be some variability from site to site.

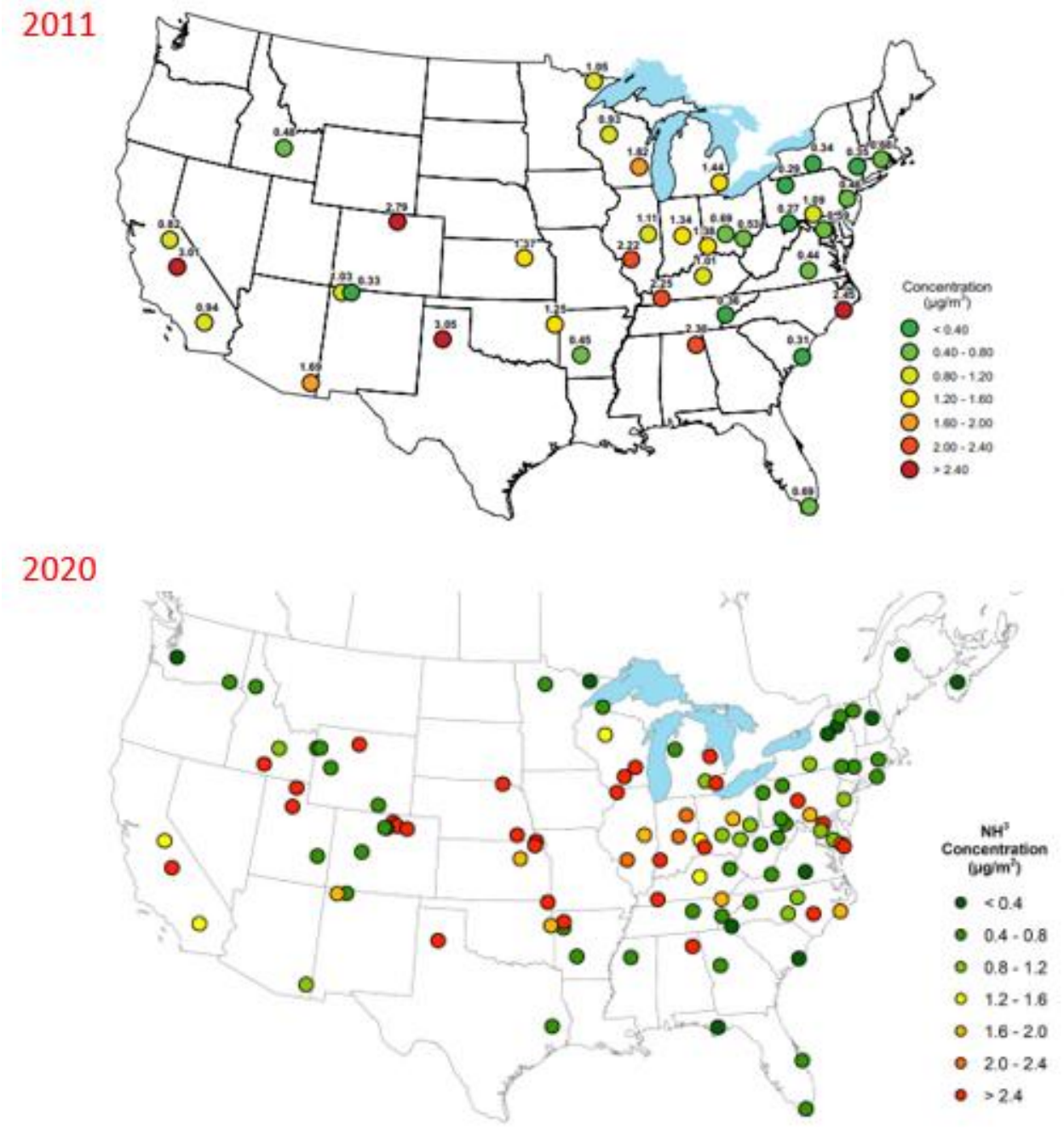

Figure 2-39. Annual average ammonia concentrations as measured by the Ammonia Monitoring Network in 2010 (top) and 2020 (bottom). Data source: NADP (2012) and NADP (2021).

2.5 NITROGEN AND SULFUR DEPOSITION

The impacts of nitrogen and sulfur emissions on public welfare endpoints via deposition are broad, complex, and variable. Contributing to the challenge of determining the impacts of these pollutants are past levels of deposition of N and S, as well as other non-air related sources of these pollutants to the surface. The focus of this review is on deposition-related impacts to ecological systems from air emissions of NO_2, SO_2, and PM. Therefore, it is important to be able to characterize deposition levels across the U.S., in order to be able to understand the relationship between pollutant concentrations, deposition, and subsequent adverse effects to public welfare. Assessing the adequacy of any standard will require the ability to relate air quality concentrations (past and present) to deposition levels (past and present). Since the previous review, the amount of N and S deposition has changed, and it is important to develop the most up-to-date datasets for the assessment of atmospheric deposition to capture these changes. This review assesses both existing measurement data and modeling capabilities.

2.5.1 Estimating Atmospheric Deposition

As introduced in section 2.3.4, measurements of deposition are incomplete and limited. While wet deposition has been routinely monitored at many locations across the U.S. for more than 30 years (NADP, 2021), dry deposition is not routinely measured. As a result, most total (wet + dry) deposition estimates are based on a combination of existing measurements and model simulations. In 2011, the NADP established the Total Deposition (TDep) Science Committee with the goal of providing estimates of total S and N deposition across the U.S. for use in estimating critical loads and other assessments. A hybrid approach has been developed to estimate total deposition based on a fusion of measured and modeled values, where measured values are given more weight at the monitoring locations and modeled data are used to fill in spatial gaps and provide information on chemical species that are not measured by routine monitoring networks (Schwede and Lear, 2014). One of the outputs of this effort are annual datasets of total deposition estimates in the U.S. which are referred to as the TDep datasets (technical updates available from NADP, 2021; ISA, Appendix 2, section 2.6).

Figure 2-40 provides a simple flowchart of the TDep measurement-model fusion. For wet deposition, the approach is to combine the concentrations of nitrate, ammonium and sulfate in precipitation as measured at NADP/NTN sites with precipitation amounts as estimated in the Parameter-elevation Relationships on Independent Slopes Model (PRISM) dataset.[9] The result is a spatially complete wet deposition dataset at 4 kilometer (km) horizontal resolution.

[9] The PRISM (Parameter-elevation Regressions on Independent Slopes Model) database is maintained by the PRISM Climate Group who compile data from multiple monitoring networks and develop spatial climate datasets to investigate short- and long-term climate patterns. https://prism.oregonstate.edu.

The dry deposition fusion is shown on the right side of Figure 2-40. The figure shows that two intermediate datasets are created as part of the TDep process: an interpolated measurement and a bias-corrected simulation. The interpolated measurement dataset relies on the CASTNET monitoring network, which measures gas-phase SO_2 and NO_Y and particulate-phase SO_4^{2-}, NO_3^-, and NH_4^+. Samples are collected for biweekly periods and chemically analyzed. The inlet allows particles of all sizes to be collected and is designed to support estimates of total oxidized nitrogen and sulfur dry deposition. The observed concentration of each chemical species is used to bias correct concentration simulations from a 12-km Community Multiscale Air Quality (CMAQ) model simulation. Because our analysis relies on the TDep representation on bias-corrected NO_2 and SO_2 concentrations, rather than directly on CMAQ simulated concentrations, we do not evaluate CMAQ concentrations in this document. The bias-corrected concentrations are then multiplied by the effective dry deposition velocity. The effective dry deposition velocity is the mean dry deposition velocity over the week-long measurement. This assessment calculates the effective dry deposition velocity, weighting the average by the hourly concentration. Meteorological processes influence both the dry deposition velocity and the concentration. The result is a set of point estimates of dry deposition. The final step is to apply inverse distance weighted interpolation based on the spatial covariance of each species (Schwede & Lear, 2014) to estimate dry deposition for the same 4-km horizontal resolution grid as the wet deposition dataset.

One shortcoming is that the measurement sites are often far apart, and the TDep interpolation does not fully capture variability between the measurement locations. The TDep method develops a bias-corrected dry deposition estimate using a CMAQ simulation. The bias correction calculates the difference between the seasonal-average CMAQ concentrations and the CASTNET concentration measurements. The bias correction at each CASTNET monitoring site is spatially interpolated to create a 4-km horizontal resolution surface. The seasonally summed CMAQ dry deposition dataset is interpolated from 12-km to the 4-km horizontal resolution then adjusted by the bias correction estimated from the modeled and measured air concentrations. This assumes that bias in concentrations can be applied to correct a bias in dry deposition, which is reasonable if the bias is due to errors in emissions or chemical production but may not be appropriate if the bias is due to inaccuracies in the dry deposition rate. The four seasonally summed datasets are summed to create an annual total dry deposition for each species. The final TDep product is a measurement-model fusion that, for dry deposition, more closely reflects measured concentrations close to CASTNet monitors while relying more heavily on modeled concentrations farther away. There is a dearth of dry deposition measurements that would be necessary to evaluate the model's representation of deposition velocity, but CMAQ modeled wet deposition and concentrations have been evaluated against ground monitors (e.g., Appel et al.,

2021, Hogrefe et al., 2023), as well as satellite data (in the case of concentrations, e.g., Pleim et al., 2019).

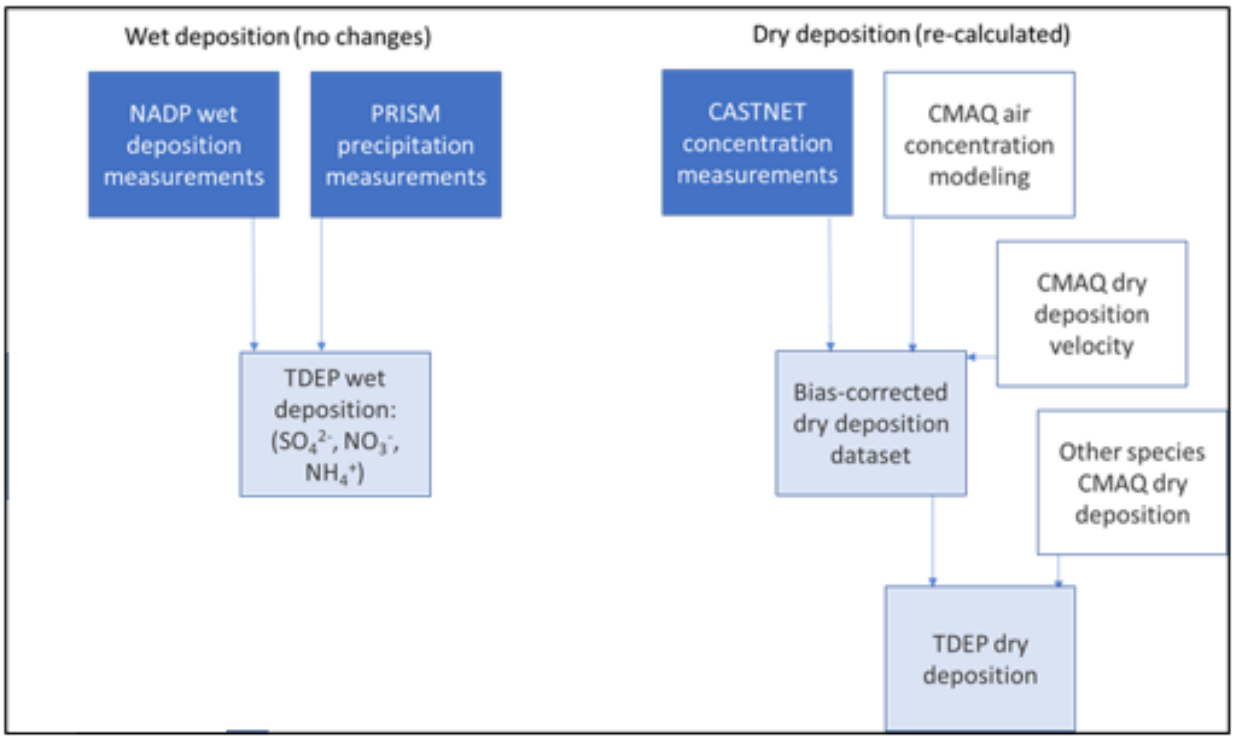

Figure 2-40. Data sources for calculating total deposition. Dark blue indicates observations, white boxes indicate chemical transport modeling results, and light blue boxes are the results of model-measurement fusion.

2.5.2 Uncertainty in Estimates of Atmospheric Deposition

Uncertainty in the resulting model-measurement fusion can be attributed to sources of deposition that are not characterized by the models or measurements, uncertainties in the CMAQ model results, and uncertainty in the spatially averaged deposition due to variability that is not accounted for in the models. While there are multiple approaches to estimating uncertainty, this review relies on what has been reported in the literature. One approach is to compare the results from multiple models with similar scientific credibility. To the extent that different models employ different scientific assumptions or parameterizations, this approach can give insight into the scientific uncertainty. Another approach is to compare the modeling results to measurements, or to withhold a subset of the data to be used as validation. This approach can provide a more quantitative assessment, but it is limited by the availability of measurements. This section summarizes the relevant studies that were used to provide a general assessment of uncertainty in TDep estimates of N and S deposition.

One source of uncertainty in the model-measurement fusion is the origin of the deposition data. Some components of deposition are directly measured, some are the result of

combining model results and measurements, and some are from modeling results only. The first step in assessing uncertainty is to assess the uncertainty from each part of the TDep calculation. Wet deposition is calculated using NADP NTN nitrogen and sulfur wet deposition measurements, which are spatially interpolated and combined with the PRISM estimates of precipitation. The PRISM dataset compares well with NADP NTN precipitation measurements (Daly et al., 2017) and the meteorological simulations from this assessment.

Dry deposition relies on a combination of measurements and models and is more challenging to assess. For oxidized nitrogen, air concentration of HNO_3 and NO_3^- particulate matter are measured at CASTNET monitoring sites. Several other compounds, such as NO_2, HONO, N_2O_5, and organic nitrogen compounds formed from photochemistry, are either not routinely measured or not routinely measured in remote areas. The CMAQ model estimates that the deposition of the latter compounds (NO_2, HONO, N_2O_5) is on average 13% of the oxidized nitrogen deposition and is largest near emission sources and urban areas (Walker et al., 2019).

For reduced nitrogen compounds, CASTNET includes measurements of NH_4^+, and AMoN includes measurements of NH_3 and often these monitors are collocated. However, because of the relative paucity of ammonia measurements, they are not used for bias correction as part of the TDep model-measurement fusion. Dry deposition of ammonia is from the CMAQ simulation. Lastly, sulfur-based compounds, SO_2 and particulate matter SO_4^{2-} are measured at CASTNET monitoring sites.

The CMAQ model is used to estimate the dry deposition velocity for all species. Like any complex system, the effect of uncertainties in one model process can be reduced by compensating processes. For example, consider uncertainties in the dry deposition velocity. If the simulated rate of dry deposition is too high, then dry deposition would be higher in the model. The enhanced dry deposition would also cause concentrations to be lower, which would in turn cause wet deposition to be lower. In this case, the dry deposition would be too high, the lower wet deposition would compensate for this, and the total deposition would be affected less. Uncertainties that affect the rate of dry deposition relative to wet deposition will have less of an effect on total deposition and can be minimized by averaging over time and space. On the other hand, if the emission rates were too high, then concentrations would be higher, and both dry and wet deposition would be higher. Uncertainties that affect air concentrations, such as emissions, will affect both wet deposition and dry deposition, and consequently total deposition (Dennis et al., 2013). Examining both air concentrations and deposition can yield insight into the nature and magnitude of uncertainties in the model results.

Although it is challenging to constrain dry deposition velocities due to the dearth of measurements, previous studies have assessed CMAQ concentration and wet deposition biases relevant to the TDep application of CMAQ concentration fields and deposition velocities.

Because nitrate and sulfate concentrations are bias adjusted in the TDep model-measurement fusion, their concentration errors have less of an effect on the estimate of dry deposition in areas near the measurement stations. Following Appel et al. (2011), CMAQ underestimates NH_4^+ wet deposition over 2002-2006 in comparison with NTN data. Implementing a precipitation correction exacerbates this bias, suggesting that precipitation errors at least partially compensate for an even larger underestimate in NH_3 concentration. On the other hand, incorporating a bi-directional parameterization for NH_3 reduced the bias in annual, precipitation-corrected NH_4^+ wet deposition from a normalized mean bias of -19% to -6% (Appel et al., 2011). More recent CMAQ updates have included additional updates to the NH_3 bi-directional parameterization (Bash et al., 2013; Pleim et al., 2019), while noting that some extent of a model underestimate in NH_3 concentration persists in a more recent CMAQ evaluation (Appel et al., 2021). The model underestimate in NH_3 concentrations has also been supported by short-term field studies in locations outside of NTN, in particular downwind of agricultural areas (e.g., Butler et al., 2015). Because the ammonia concentration and the ammonia dry deposition are not constrained by measurements in the TDep model-measurement fusion calculations, it is likely that the resulting estimates for current conditions reported in this assessment underestimate ammonia dry deposition due to the underestimate in ammonia concentrations.

In addition to assessing the uncertainty of the CMAQ model, it is also necessary to assess the uncertainty in the NADP NTN and CASTNET measurements. The concentration and deposition measurements have a specified level of precision defined in the data quality objectives for each monitoring network. The NADP NTN monitors specify a less than 10% uncertainty and for the CASTNET air concentration measurements the uncertainty is specified as +/- 20%. This is achieved through quality assurance and data management protocols. However, this may not be a complete assessment of the uncertainty. In the case of CASTNET, several studies have collocated reference monitors and inter-compared the different measurement techniques. Differences in sulfate tend to be small. But for nitrate and ammonium in particulate matter, the different sampling methods can yield larger differences (ISA, Appendix 2, section 2.4.5). The differences are thought to be increased by high humidity or influence from coastal airmasses that affect the PM composition, and accordingly may not be relevant everywhere in the U.S. Fully characterizing the differences that arise from different monitoring techniques is beyond the scope of this assessment. Instead, this assessment relies on the data quality objectives as a proxy for uncertainty.

Lastly, the fusion of the model and measurements to a set spatial grid also contributes to uncertainty. The grid representation of the model-measurement fusion may obscure fine resolution variability leading to uncertainty in the deposition to a specific ecosystem. The dry deposition velocity can differ considerably depending on the surface conditions, complex terrain,

elevation, and land cover. For example, the dry deposition velocity of nitric acid (HNO_3) is four times faster over a forest than a lake. In regions with varied terrain, this can create substantial variability in the dry deposition that is not captured at the 4-km horizontal spatial scale of the TDep interpolation. This is also substantial in coastal areas or city-wildland interfaces. A study by Paulot et al. (2018) estimated that grid-based results from models may underestimate deposition to natural vegetation by 30%. Another issue is the spatial resolution may obscure gradients in concentration. This is especially true of compounds such as NO_2 that have high concentrations near emission sources, but degrade quickly, leading to large spatial gradients. Thus, this type of uncertainty is likely less than in other, more populated areas.

2.5.3 National Estimates of Deposition

Total sulfur and total nitrogen deposition estimates for the continental U.S. at 4-km horizontal resolution have been developed for calendar years 2000 through 2021 (NADP, 2021). These data are used in quantitative analyses of ecosystem exposure and risk in the later sections of this document. Figure 2-41 illustrates that nitrogen deposition in 2019-2021 is estimated to be highest in and around regions with large sources. This mostly includes regions of intensive crop and animal livestock production, which are large sources of NH_3 emissions. Total nitrogen deposition results from both the dry and wet deposition pathways as shown in Figures 2-42 (dry) and 2-43 (wet). Dry deposition tends to occur in source-oriented hot spots (e.g., parts of IA, MN, NC, and TX) and is dominated by ammonia (discussed in more detail in 2.5.3.1), while wet deposition tends to be more homogenous, but highest in the central U.S. The wet deposition of N estimates for 2019-2021 have contributions from both ammonium (Figure 2-44) and nitrate (Figure 2-45), with ammonium being larger.

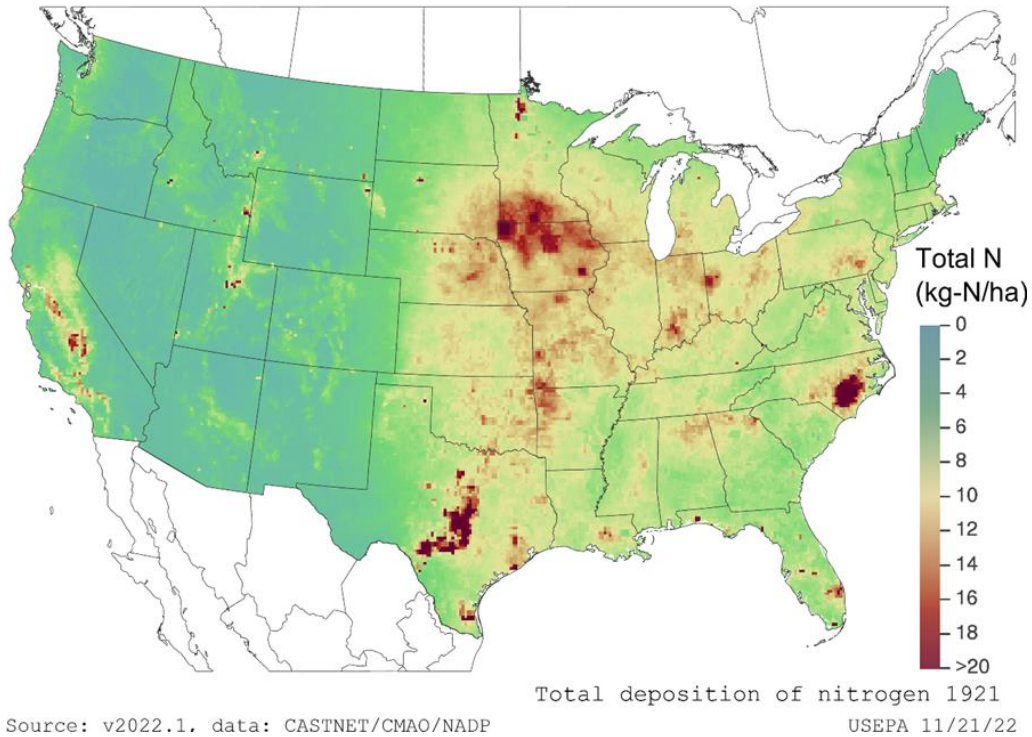

Figure 2-41. Annual average total deposition of nitrogen (2019-2021).

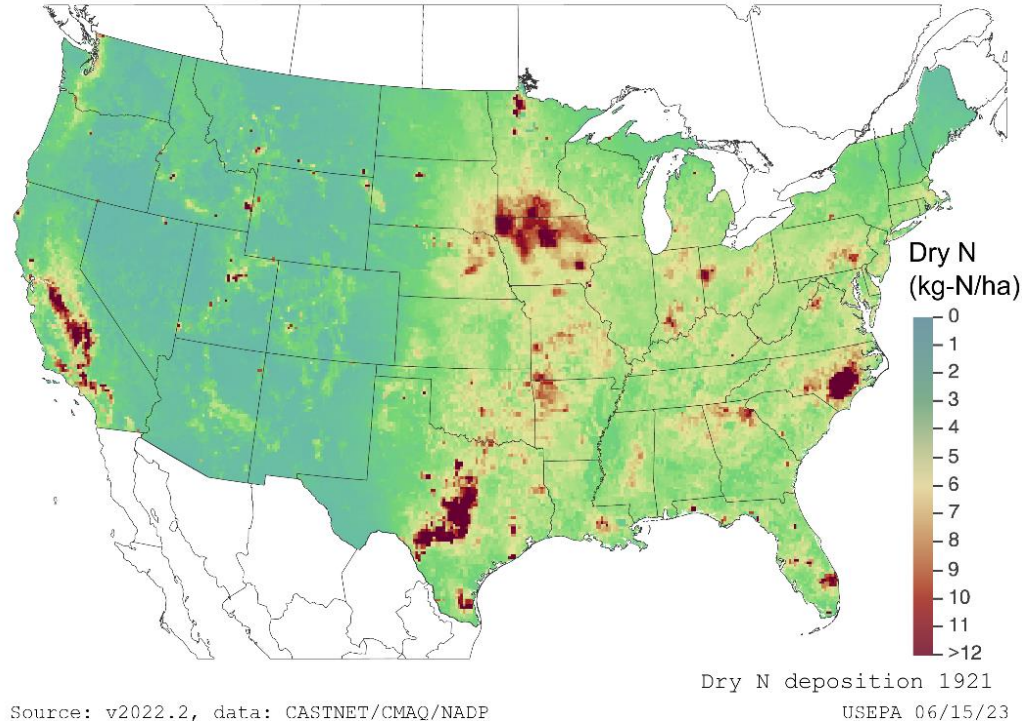

Figure 2-42. Annual average dry deposition of nitrogen (2019-2021).

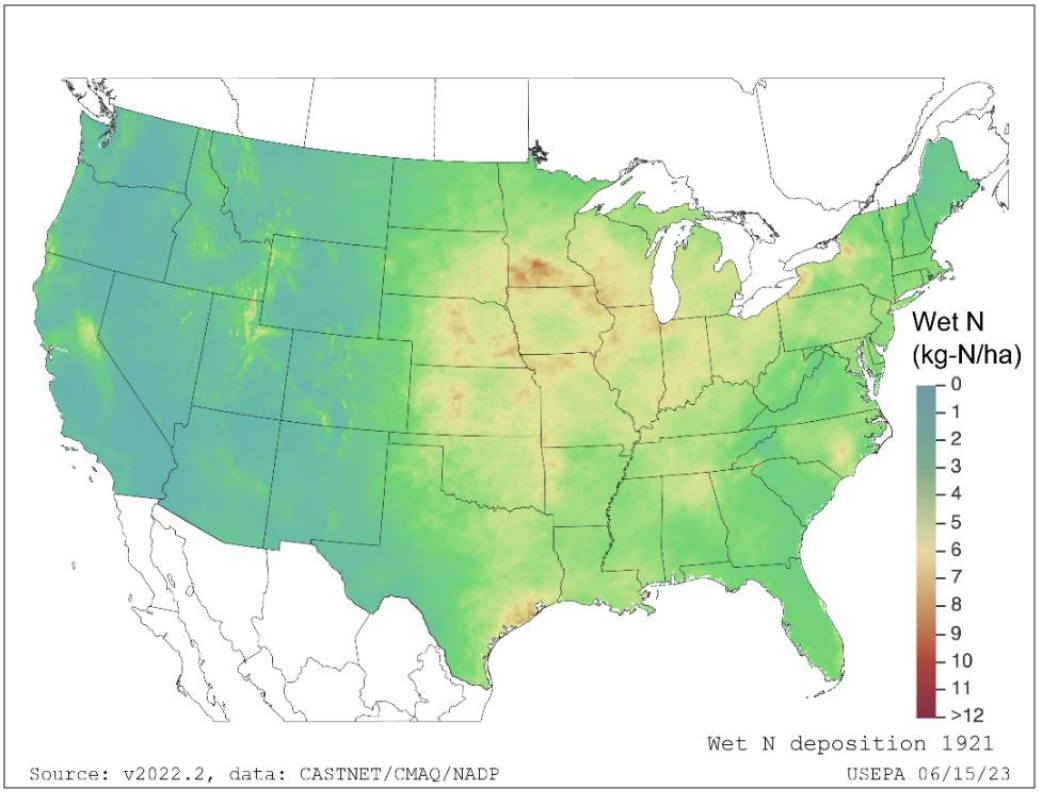

Figure 2-43. Annual average wet deposition of nitrogen (2019-2021).

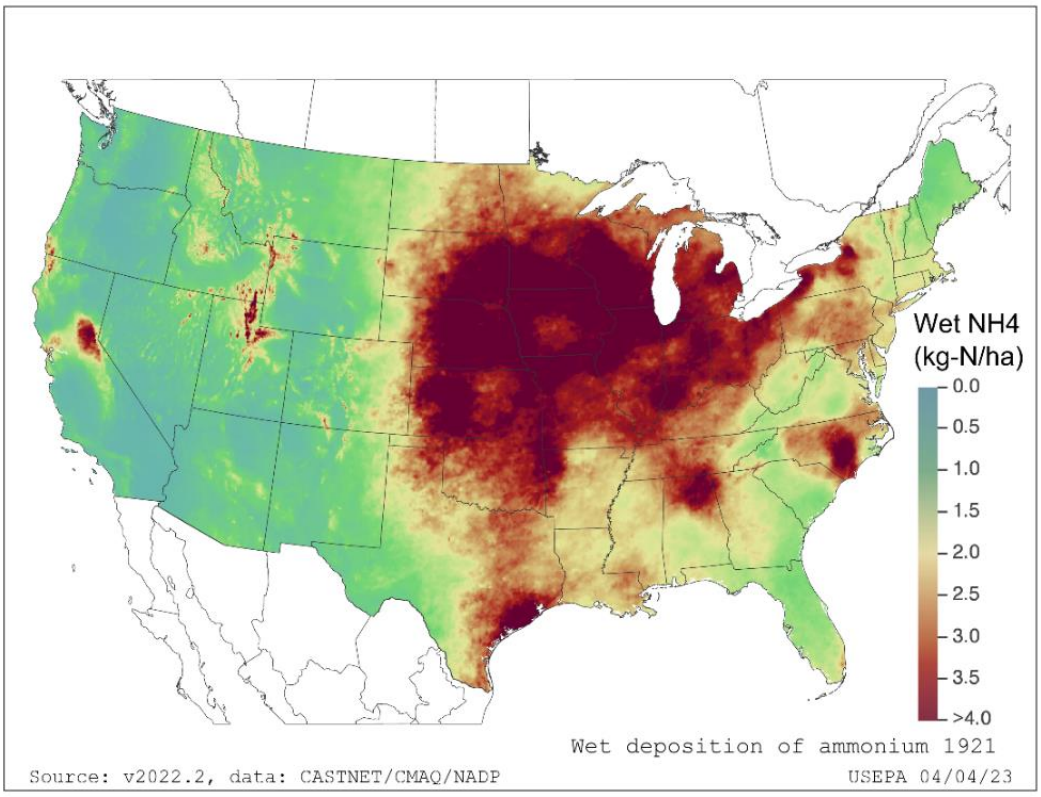

Figure 2-44. Annual average wet deposition of ammonium (2019-2021).

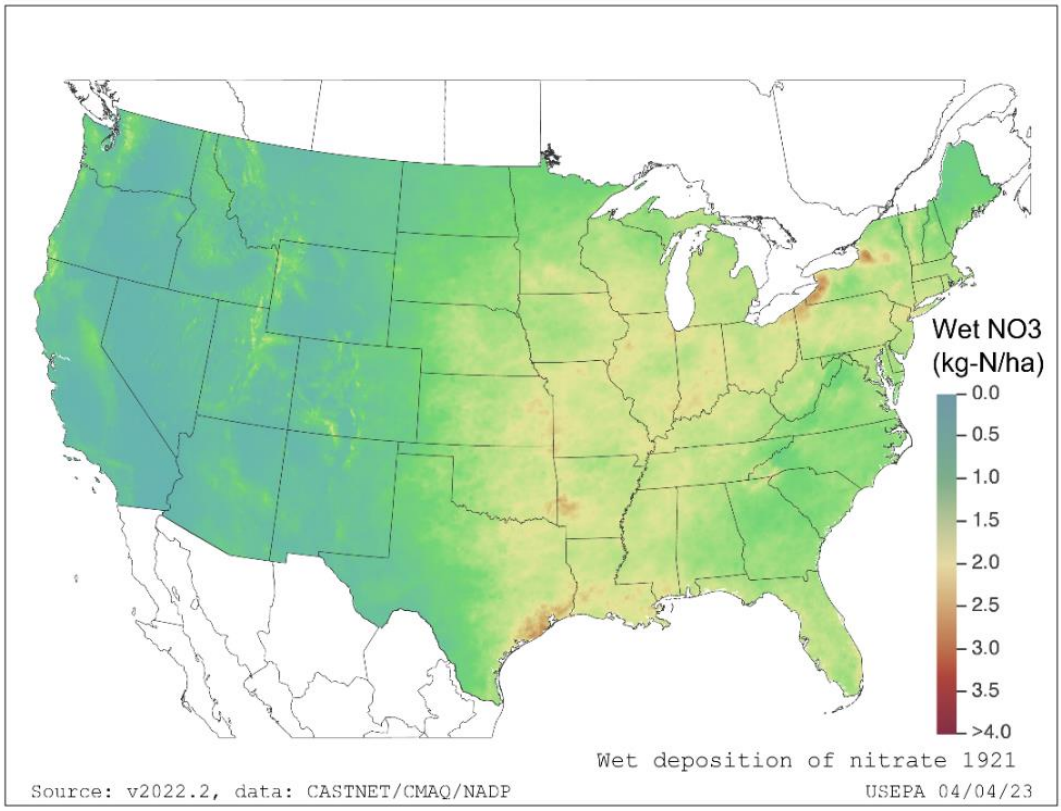

Figure 2-45. Annual average wet deposition of nitrate (2019-2021).

The total sulfur deposition estimates for 2019-2021 are shown in Figure 2-46. For this recent period, sulfur deposition is generally higher in the eastern U.S. (e.g., along the Gulf Coast and in the Mississippi Valley). The large majority of sulfur deposition in the most recent time period is caused by wet deposition, with the exception of a few areas in the western U.S., as shown by Figure 2-47.

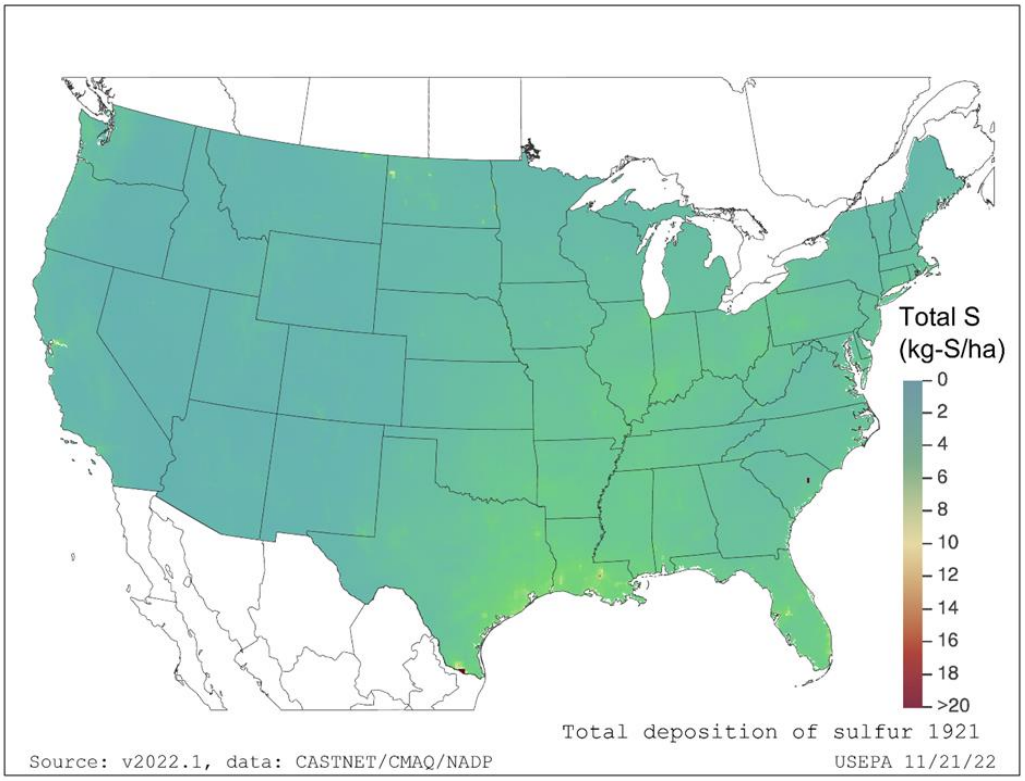

Figure 2-46. Annual average total deposition of sulfur (2019-2021).

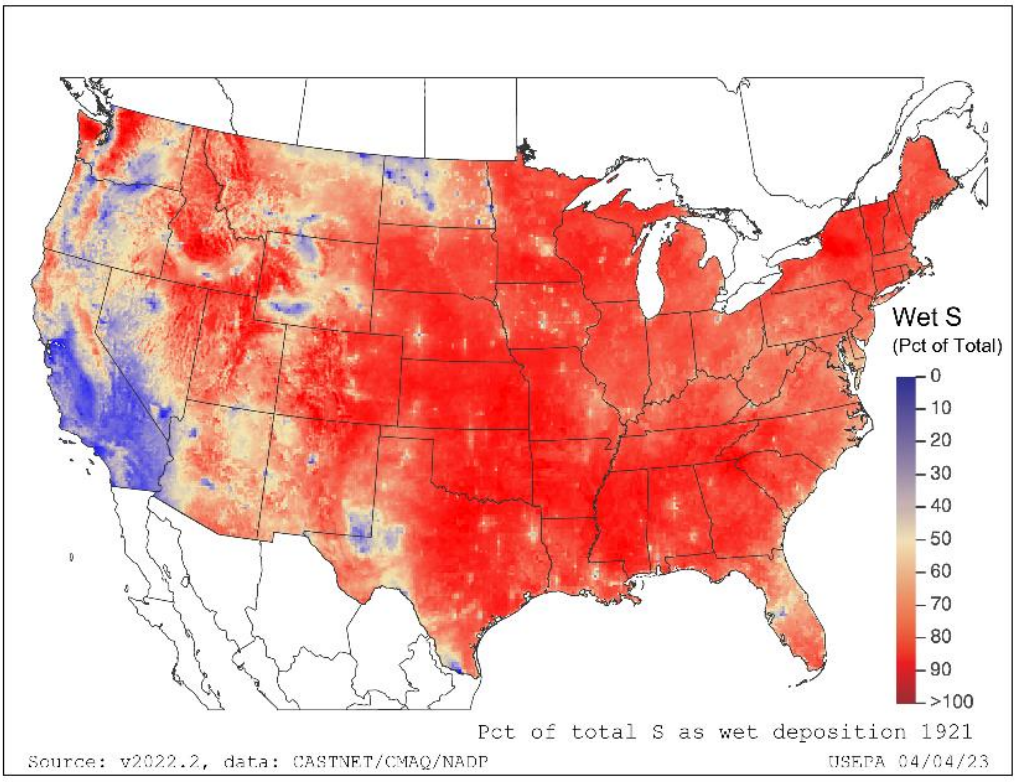

Figure 2-47. Percentage of total deposition of sulfur that occurs as wet deposition across the 2019-2021 period.

2.5.3.1 Contribution from NH_3

Ammonia contributes to total nitrogen deposition, but it is not an oxidized form of nitrogen, so it is not part of the definition of "oxides of nitrogen." In addition, although ammonia is a precursor to PM formation, ammonia is a gas and not a component of particulate matter. Accordingly, ammonia, itself, is not among the criteria pollutants that are part of this review, and therefore we have quantified the contribution of ammonia to nitrogen deposition separately from the other components of nitrogen deposition.

Figure 2-48 shows the dry deposition of ammonia over a recent period (2019-2021). It can be observed, when comparing with Figure 2-42 (note: scales differ), that the majority of dry N deposition is from ammonia (i.e., reduced nitrogen). Figure 2-49 displays the percentage of total N deposition that results from reduced nitrogen. Total nitrogen deposition is the sum of the deposition of ammonia, ammonium, and oxidized nitrogen compounds. The contribution of reduced nitrogen to total N deposition exceeds 70% in areas with high ammonia concentrations, including areas of intensive livestock and crop production in eastern North Carolina, parts of Iowa, Minnesota, Texas, and the Central and Imperial valleys in California (Figure 2-48). In other areas, this contribution more commonly ranges from 40-60% (Figure 2-49).

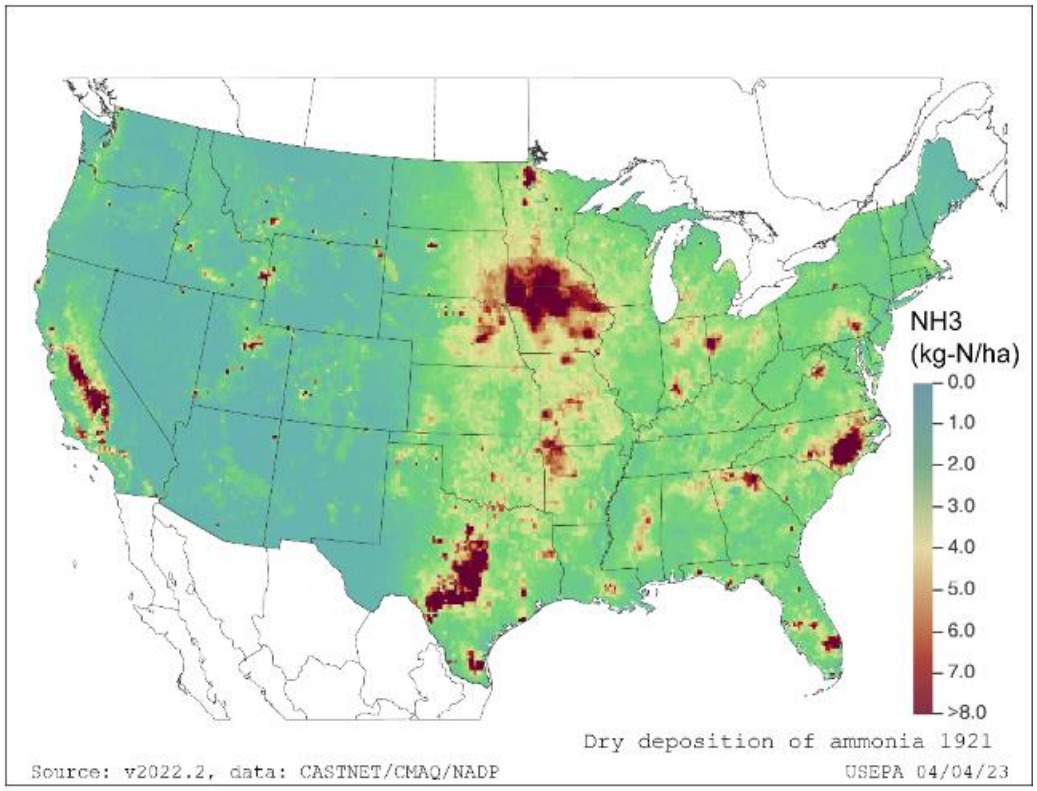

Figure 2-48. Annual average dry deposition of ammonia (2019-2021).

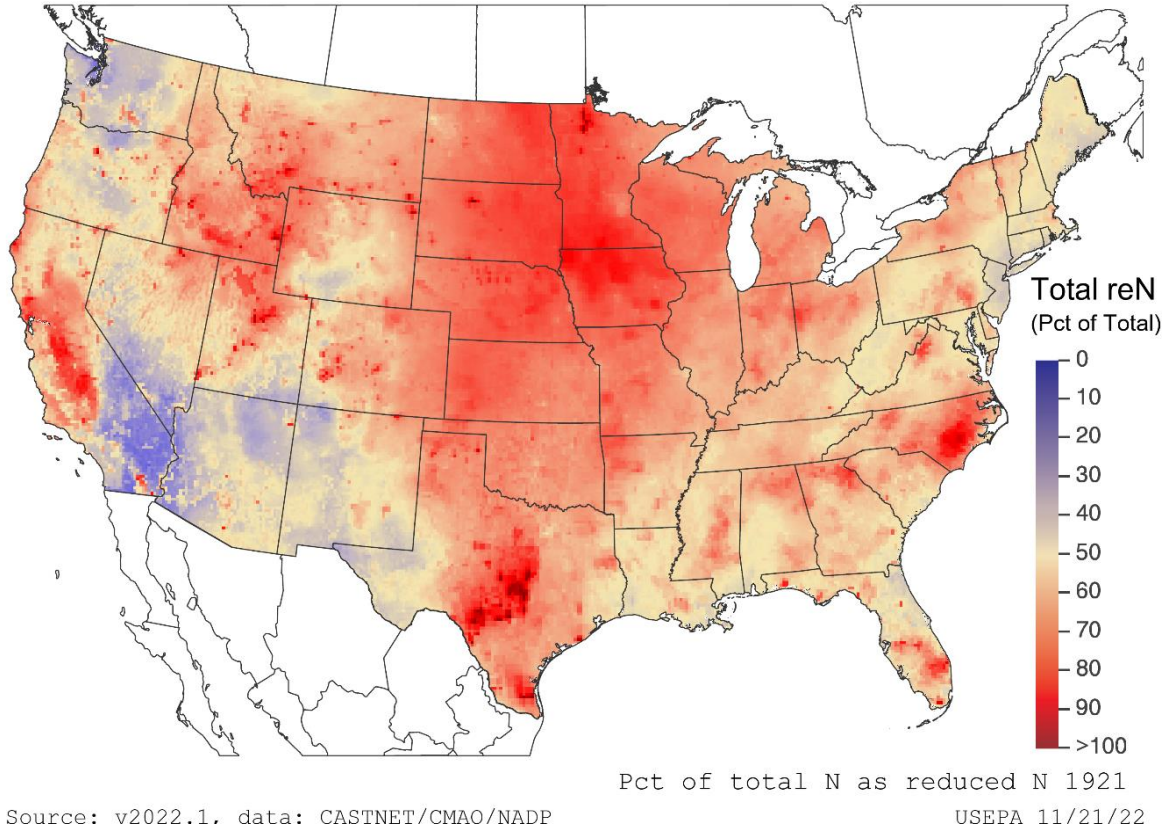

Figure 2-49. Average percent of total N deposition in 2019-2021 as reduced N (gas phase NH_3 and particle phase NH_4^+).

2.5.3.2 Contribution from International Transport

On a national average scale, only a small fraction of sulfur and nitrogen deposition can be attributed to natural emissions or international transport (ISA, Appendix 2, section 2.6.8). Chemical transport models have been used to quantify these contributions (Horowitz et al., 2003; Zhang et al., 2012; Lee et al., 2016). The natural sources of oxidized nitrogen include microbial activity in unfertilized soils and lightning. Natural sources of ammonia include microbial activity in unfertilized soils and wild animals. Chemical transport model simulations have been used to estimate that natural emission sources contribute 16% of the total N deposition in the U.S. Because ammonia and most forms of oxidized N have relatively short atmospheric lifetimes, on the order of hours for gas phase NO_X and NH_3 and days for ammonium and/or nitrate $PM_{2.5}$, international transport contributes just 6% of the N deposition, except within 100 km of the U.S.-Canada or U.S.-Mexico borders, where the contribution is estimated to be at most 20%. U.S. anthropogenic emissions account for 78% of reactive N deposition over the contiguous United States (ISA, Appendix 2, section 2.6.8). Sulfur is naturally emitted from plankton in the ocean and from geologic activity – volcanoes, fumaroles, etc. Like N, relatively little sulfur deposition

can be attributed to international transport. Chemical transport model simulations have been used to estimate that approximately 10% of S in the eastern U.S. can be attributed to natural and international sources. In the western U.S., this increases to 20%, since there is lower S deposition from anthropogenic sources, more geologic emission sources, and closer proximity to long range transport from international sources. In areas with high S deposition, less than 1% can be attributed to natural and international sources (ISA, Appendix 2, section 2.6.8).

2.5.4 Trends in Deposition

With the changes in emissions and air concentrations described above, total deposition of oxidized nitrogen and sulfur have also decreased significantly since 2000 (Feng et al., 2020; McHale et al., 2021). Between the three-year period 2000-2002 and 2018-2020, national average S deposition over the contiguous U.S. has declined by 68% and total N deposition has declined by 15% (U.S. EPA, 2022b). Table 2-2 presents a regional breakout of trends in total S, total N, oxidized N, and reduced N deposition represented as kilograms per hectare (kg/ha). The change in total N deposition is a combination of declining oxidized N and increasing reduced N, which is consistent with the trends in emissions and air concentrations described above. Emissions of NO_X and wet deposition of nitrate have a positive correlation, but because the formation of ammonium is related to the availability of nitrate and sulfate, the correlation between NH_3 emissions and NH_4^+ wet deposition is weaker and negative (Tan et al., 2020). While dry deposition is more uncertain in magnitude, both surface-based and remote-sensing measurements indicate increasing ammonia concentrations, which points to an increasing trend for ammonia dry deposition, especially in areas with significant agricultural emissions in the Midwest and Central Valley of California where ammonia dry deposition has become the largest contributor to inorganic N deposition (Li et al., 2016). As expected, the data suggest that dry deposition of nitric acid has decreased significantly over the past two decades and is likely a key contributor to the decrease in total nitrate deposition and decreasing trends in oxidized nitrogen deposition (ISA, Appendix 2, section 2.7).

Figures 2-50 through 2-56 display the spatial patterns of TDep-estimated deposition across a range of pollutants for two periods (2000-2002 and 2019-2021) to further illustrate the changes in deposition patterns across the U.S. over the past two decades. As shown in Figure 2-50, S deposition has decreased sharply across the U.S. over this period due to the significant decreases in sulfur emissions. Sulfur deposition in the Ohio River Valley region is particularly notable. The trends in N deposition are more heterogeneous. Total N deposition has decreased over parts of the Ohio River Valley and downwind regions in the northeastern U.S. (Figure 2-51), but there are parts of the country where increases in N deposition are estimated to have occurred over the past two decades (e.g., Texas).

Table 2-2. Regional changes in deposition between 2000-2002 and 2019-2021: (a) total S deposition; (b) total, oxidized and reduced N deposition (U.S. EPA, 2022b).

(a) Change in total S deposition

Form of S Deposition	Region	2000-2002	2019-2021	% change
Total Deposition of Sulfur (kg S ha^{-1})	Mid-Atlantic	15.9	2.1	-87
	Midwest	11.2	2.2	-80
	North Central	3.5	1.5	-56
	Northeast	8.7	1.5	-83
	Pacific	1.0	0.6	-38
	Rocky Mountain	1.0	0.6	-46
	South Central	5.4	2.8	-49
	Southeast	10.3	2.6	-74

(b) Change in total, oxidized and reduced N deposition

Form of N Deposition	Region	2000-2002	2019-2021	% change
Total Deposition of Nitrogen (kg N ha^{-1})	Mid-Atlantic	13.4	8.5	-36
	Midwest	12.2	9.8	-20
	North Central	8.5	9.5	+11
	Northeast	10.4	6.2	-40
	Pacific	3.8	3.1	-18
	Rocky Mountain	3.0	3.1	+3
	South Central	7.8	9.0	+16
	Southeast	10.8	8.4	-23
Total Deposition of Oxidized Nitrogen (kg N ha^{-1})	Mid-Atlantic	10.3	4.0	-62
	Midwest	8.0	3.6	-54
	North Central	4.1	2.6	-37
	Northeast	7.7	2.9	-62
	Pacific	2.4	1.4	-42
	Rocky Mountain	1.9	1.3	-35
	South Central	5.0	3.1	-39
	Southeast	7.7	3.4	-56
Total Deposition of Reduced Nitrogen (kg N ha^{-1})	Mid-Atlantic	3.0	4.6	+51
	Midwest	4.3	6.2	+45
	North Central	4.4	6.9	+56
	Northeast	2.7	3.3	+22
	Pacific	1.4	1.7	+22
	Rocky Mountain	1.1	1.8	+72
	South Central	2.8	6.0	+111
	Southeast	3.1	5.0	+63

The states included in each region are as follows: Mid-Atlantic: DE, MD, NJ, PA, VA, WV; Midwest: IL, IN, KY, MI, OH, WI; North Central: IA, KS, MN, MO, ND, NE, SD; Northeast: CT, MA, ME, NH, NY, RI, VT; Pacific: CA, NV, OR, WA; Rocky Mountain: AZ, CO, ID, MT, NM, UT, WY; South Central: AR, LA, OK, TX; Southeast: AL, FL, GA, MS, NC, TN, SC.

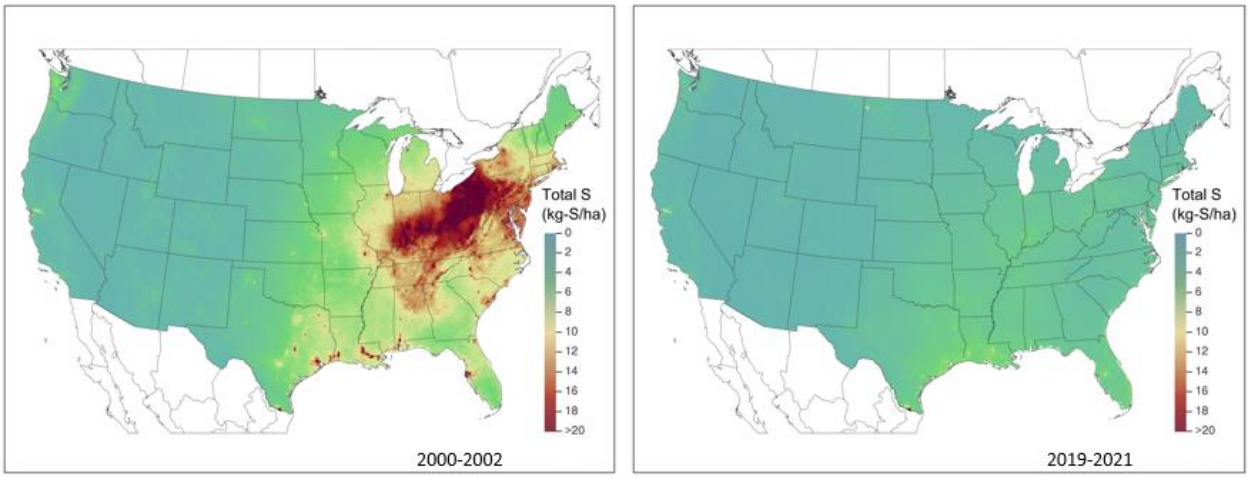

Figure 2-50. TDep-estimated total S deposition: 2000-2002 (left) and 2019-2021 (right).

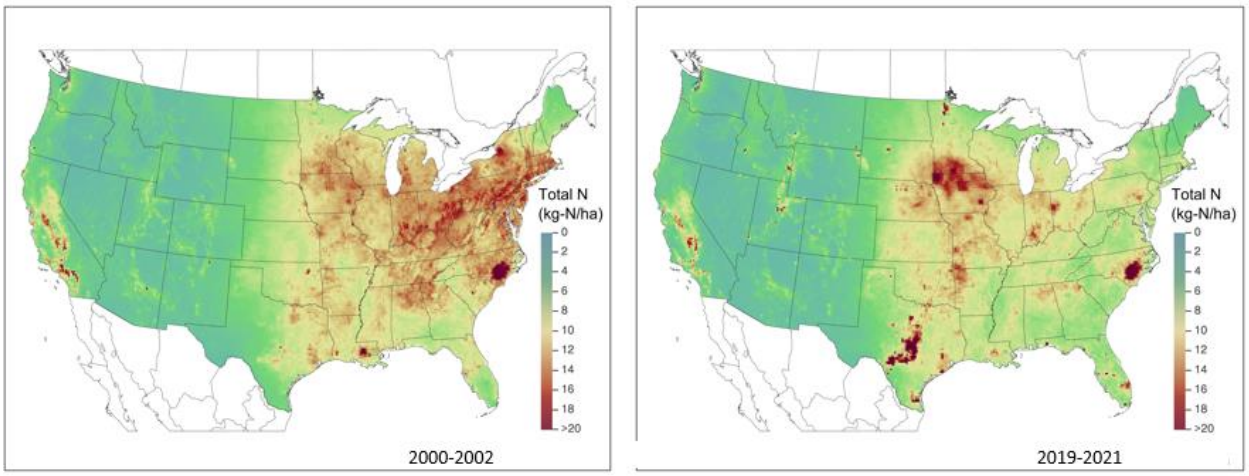

Figure 2-51. TDep-estimated total N deposition: 2000-2002 (left) and 2019-2021 (right).

Looking into the components of these trends in N deposition, it can be seen from Figure 2-52 that most of the widespread changes in N deposition across the U.S., both increases and decreases, are due to changes in dry deposition of N. Figure 2-53 shows that while there have been some changes in wet N deposition over the past 20 years (e.g., decreases near Lake Ontario; increases in parts of southern MN), these levels and patterns have remained relatively unchanged compared to dry N deposition.

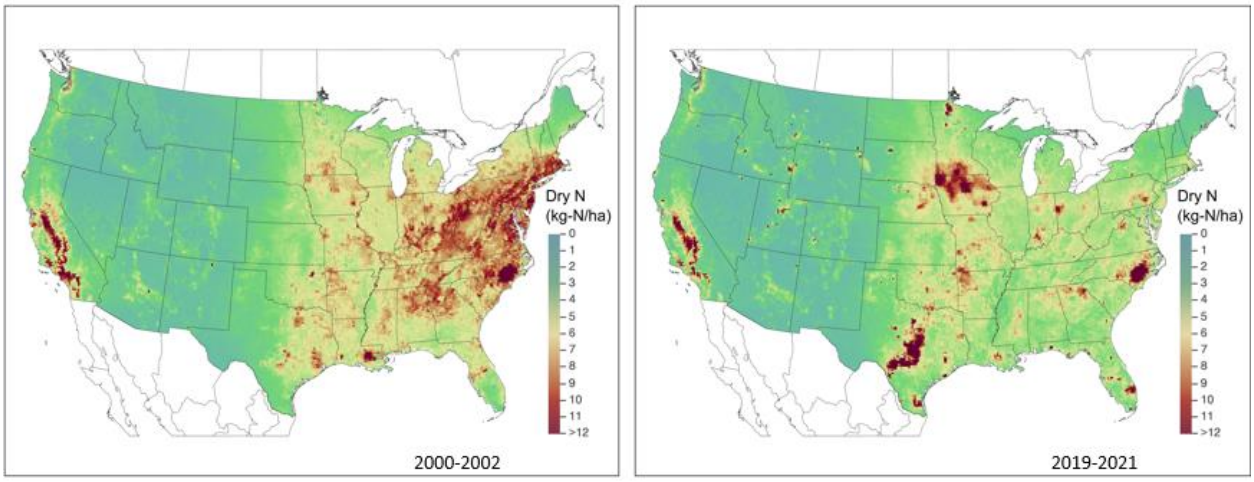

Figure 2-52. TDep-estimated dry N deposition: 2000-2002 (left) and 2019-2021 (right).

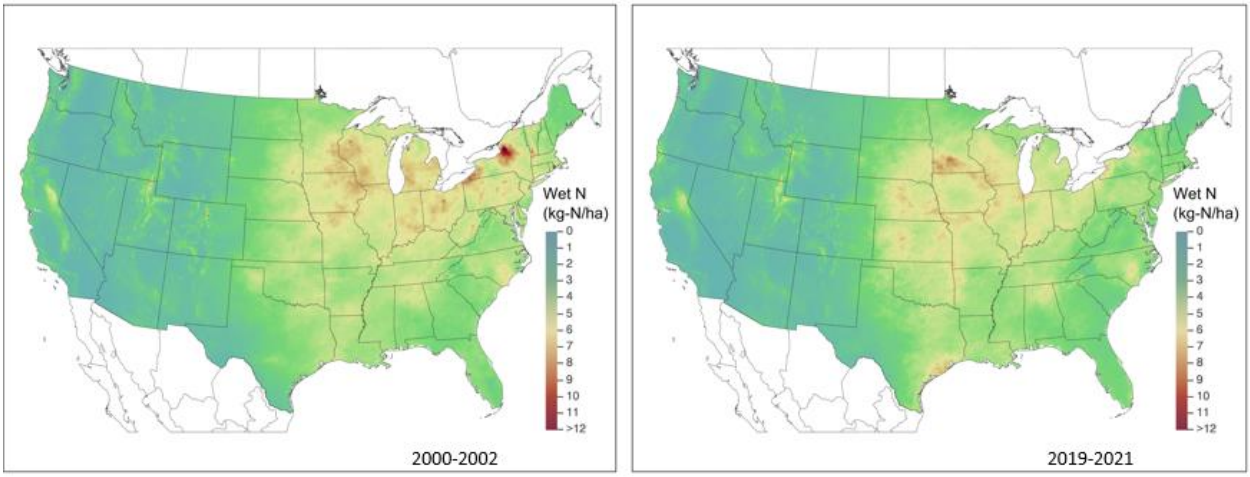

Figure 2-53. TDep-estimated wet N deposition: 2000-2002 (left) and 2019-2021 (right).

The aggregate trends in dry deposition of N are driven by two largely opposing trends in the dry deposition of oxidized nitrogen and reduced nitrogen. Two decades ago, there were large amounts of dry oxidized N deposition (5-10 kg N/ha) over much of the eastern U.S. which are not seen in the more current period (< 5 kg N/ha), as shown in Figure 2-54. Conversely, while there were isolated hotspots or dry reduced N deposition in the 2000-2002 timeframe, the number and magnitude of these hotspots has increased significantly in the more recent 2019-2021 period, as shown by Figure 2-55, especially in places like AR, IA, MN, MO and TX. Figure 2-56 confirms that the increases in dry deposition of reduced N are closely linked to increases in NH_3 deposition.

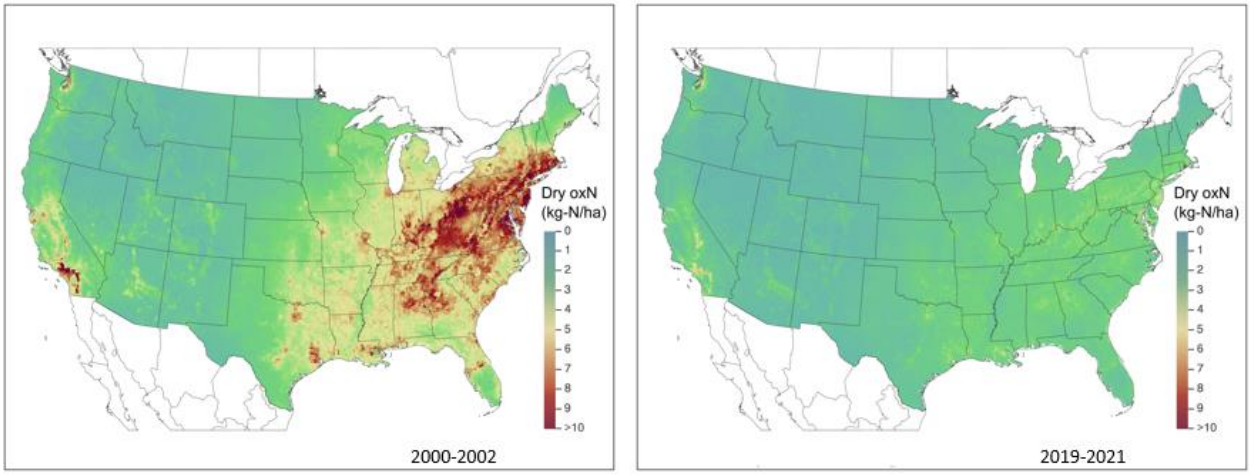

Figure 2-54. TDep-estimated dry oxidized N deposition: 2000-2002 (left) and 2019-2021 (right).

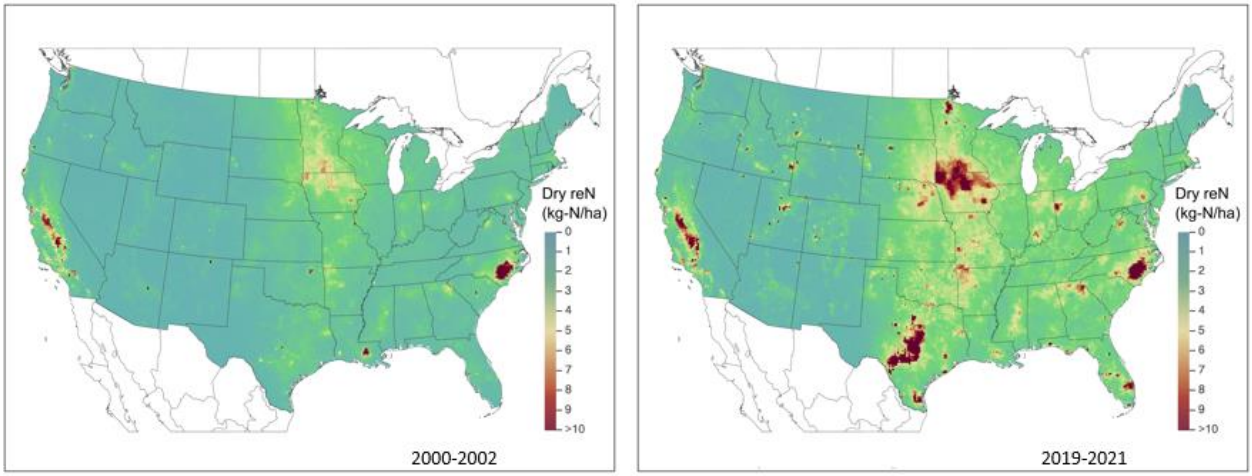

Figure 2-55. TDep-estimated dry reduced N deposition: 2000-2002 (left) and 2019-2021 (right).

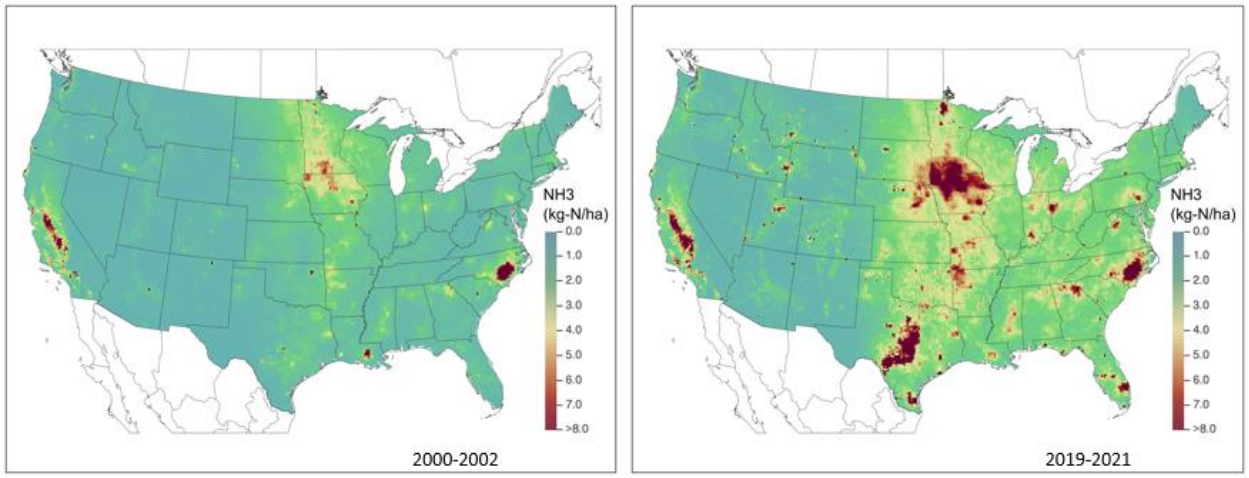

Figure 2-56. TDep-estimated NH_3 deposition: 2000-2002 (left) and 2019-2021 (right).

The trends in deposition of reduced nitrogen should be viewed with some caution, in part because before 2011, ambient air NH_3 monitoring was rare. For particulate matter, the trend in ammonium (NH_4^+) has followed the downward trends in sulfate and nitrate, because in order for NH_3 to partition into the particle phase, an anion, such as sulfate or nitrate, is needed to neutralize it. Satellite-based measurements and chemical transport models have been used to augment the surface-based measurements of ammonia and ammonium to better understand trends. These studies also show increasing ammonia concentrations, especially in parts of the Midwest, South-east, and West near agricultural sources (Warner et al., 2016; Warner et al., 2017; Yu et al., 2018; Nair et al., 2019; He et al., 2021). These trends are attributed to a combination of warmer temperatures causing greater emissions, increasing agricultural activity, and less available sulfate and nitrate, shifting particle ammonium to gas-phase ammonia.

While there is always uncertainty in projecting future trends, the EPA generally expects reductions in total national N and S deposition over the next decade, although this will depend on trends in reduced N deposition. In a recent regulatory impact assessment for the proposed revisions to the PM NAAQS, the EPA used the CMAQ model to simulate an illustrative implementation scenario that included additional emissions reductions of NO_X and SO_2 (U.S. EPA, 2022a). The emission scenarios for these simulations included impacts projected for the Inflation Reduction Act of 2022 Tax Incentive Provisions, the 2023 Good Neighbor Plan, and the 2022 Control of Air Pollution from New Motor Vehicles: Heavy-Duty Engine and Vehicle Standards, among other finalized rules. Rules that were not yet finalized at the time of the Inflation Reduction Act's release (e.g., 2023 111b and d and MATS proposals) were not included. The percent change in total N and total S deposition projected to occur by the model in 2032 (from a baseline 2016 scenario) within Class I areas is shown in Figure 2-57 and Figure 2-58, respectively. In this scenario, deposition in Class I Areas is expected to continue to decline as existing regulations are implemented, due to reductions in NO_X and SO_2 emissions. While national NH_3 emissions were projected to increase between 2016 and 2032 based on anticipated changes in activity (e.g., growth in livestock), these increases were insufficient to offset the reductions in deposition associated with NO_X and SO_2 emission reductions (U.S. EPA, 2022a). The projected average deposition reduction for N and S is about 10%, with largest reductions occurring in the East. The projected reduction in S emissions in the Pacific Coast states is relatively minor, but there is already very little S deposition and very few SO_2 emission sources in this region. It should be noted that there is considerable uncertainty in the change in future deposition related to the potential for revision to the annual average $PM_{2.5}$ primary standard (88 FR 5558, January 27, 2023). The emission sources that typically contribute most to the areas of highest $PM_{2.5}$ concentrations can be located relatively far from more remote Class I Areas and can have a highly variable effect on deposition in those areas. Second, as part of implementation

of $PM_{2.5}$ standards, States can elect to reduce emission sources that contribute to organic carbon $PM_{2.5}$ which would be expected to have little impact on deposition.

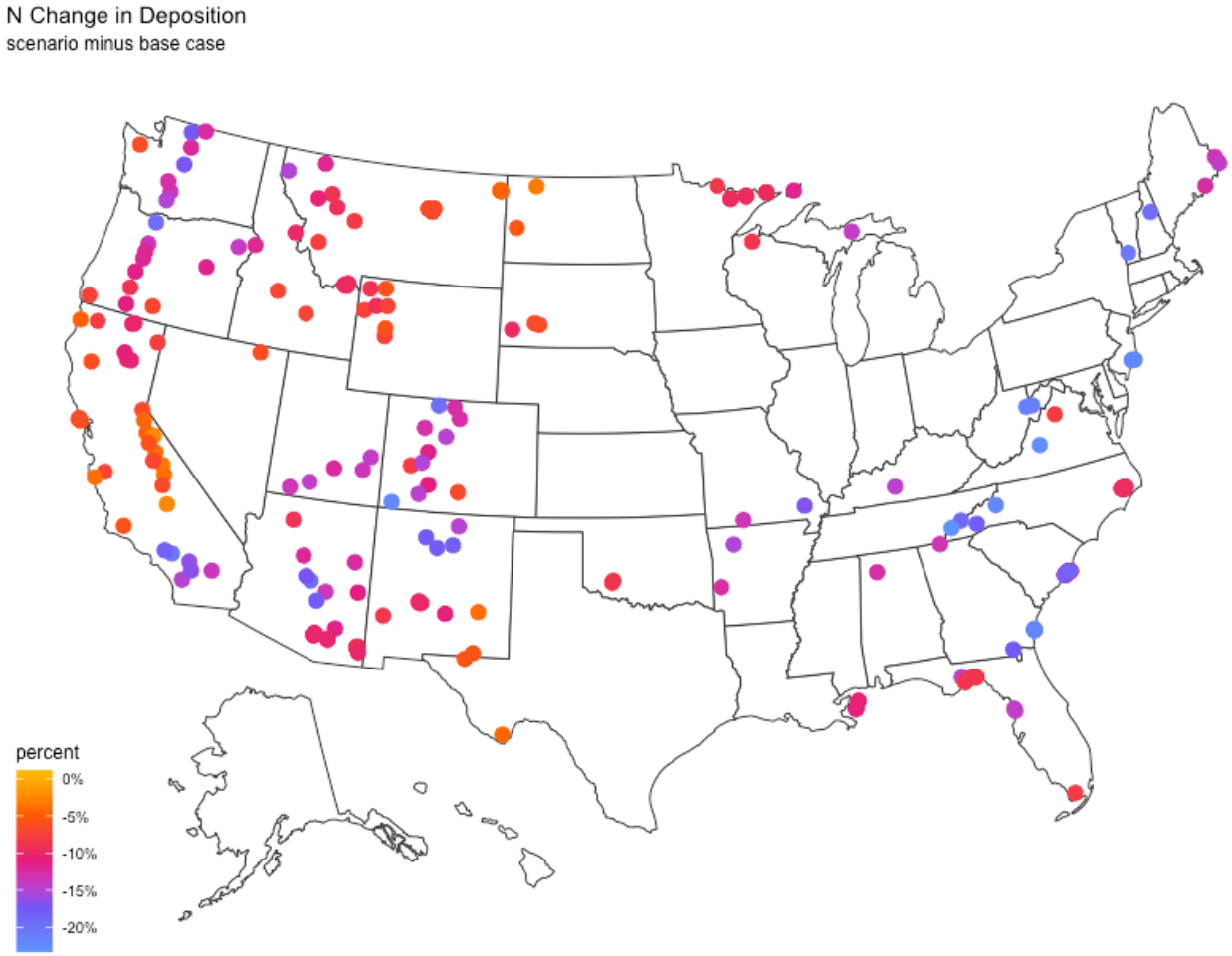

Figure 2-57. Projected percent change in total N deposition in Class 1 areas from 2016, based on a scenario for 2032 that includes implementation of existing national rules on mobile and stationary sources (U.S. EPA, 2022a).

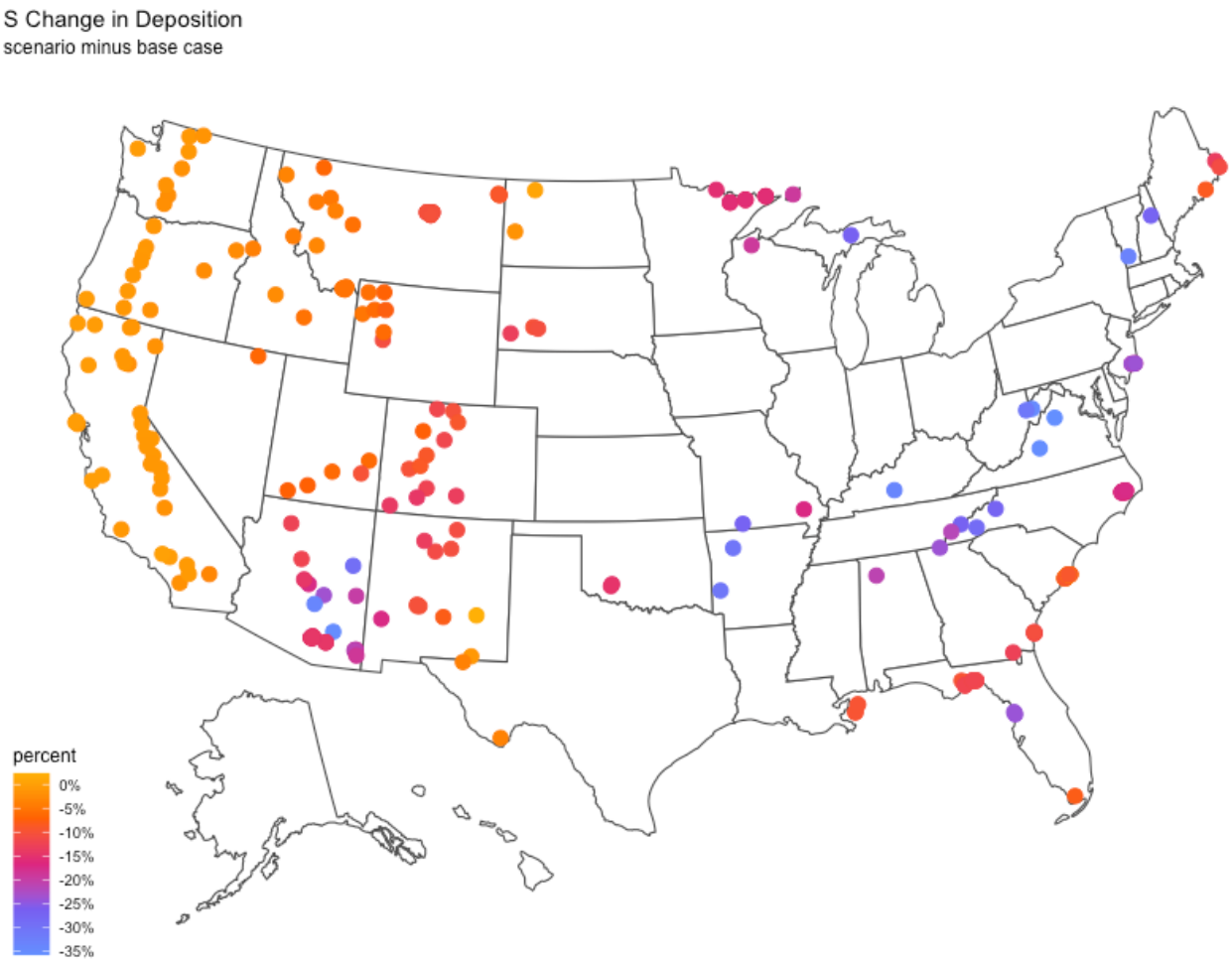

Figure 2-58. Projected percent change in total S deposition in Class 1 areas from 2016, based on a scenario for 2032 that includes implementation of existing national rules on mobile and stationary sources (U.S. EPA, 2022a).

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3 CURRENT STANDARDS AND GENERAL APPROACH FOR THIS REVIEW

This review focuses on evaluation of the currently available evidence and quantitative analyses related to the welfare effects of oxides of S and N (also referred to as SO_X and N oxides) and the ecological effects of PM[1] in consideration of several overarching policy-relevant questions. The first such question considers whether the currently available scientific evidence and quantitative information support or call into question the adequacy of the public welfare protection for these effects afforded by the current secondary standards for these pollutants. In this context we consider two categories of effects: (1) effects associated with the airborne pollutants (sometimes referred to as "direct effects" of the pollutants in ambient air), and (2) effects associated with deposition of the pollutants or their transformation products into aquatic and terrestrial ecosystems.

This chapter describes the basis for the existing secondary standards (section 3.1) and the approach taken in the 2012 review of deposition-related effects (section 3.2) and also outlines the approach being taken in this review of the current NO_2, SO_2 and PM secondary standards (section 3.3).

3.1 BASIS FOR THE EXISTING SECONDARY STANDARDS

The existing secondary standards for SO_X and N oxides were established in 1971 (36 FR 8186, April 30, 1971). The secondary standard for SO_2 is 0.5 ppm, as a 3-hour average, not to be exceeded more than once per year (40 CFR §50.5). The secondary standard for N oxides is 0.053 ppm NO_2 (100 micrograms per cubic meter [$\mu g/m^3$] of air), as the arithmetic mean of the 1-hour NO_2 concentrations over the course of a year (40 CFR §50.11). Both standards were selected to provide protection to the public welfare related to effects on vegetation (U.S. DHEW, 1969; U.S. EPA, 1971).

The welfare effects evidence for SO_X in previous reviews indicates a relationship between short- and long-term SO_2 exposures and foliar damage to cultivated plants, reductions in productivity, species richness, and diversity (U.S. DHEW, 1969; U.S. EPA, 1982a; U.S. EPA, 2008). At the time the standard was set, concentrations of SO_2 in the ambient air were also associated with other welfare effects, including effects on materials and visibility (U.S. DHEW,

[1] As noted in Chapter 1, other welfare effects of PM, such as visibility and materials damage were addressed in the separate PM NAAQS review completed in 2020 and are part of the reconsideration of that 2020 decision, a proposed decision for which was published earlier this year (88 FR 5558, January 27, 2023). Given the presence of S and N compounds in PM, the ecological effects of PM are included in this review.

1969). However, the available data were not sufficient to establish a quantitative relationship between specific SO_2 concentrations and such effects (38 FR 25679, September 14, 1973). These two categories of effects have more recently been considered in the PM secondary NAAQS reviews (e.g., 85 FR 82684, December 18, 2020). Accordingly, direct effects on vegetation of SO_X in ambient air is the basis for the existing secondary standard for SO_X.

The welfare effects evidence for N oxides in previous reviews includes foliar injury, leaf drop, and reduced yield of some crops (U.S. EPA, 1971; U.S. EPA, 1982a; U.S. EPA, 1993; U.S. EPA, 2008). Since it was established in 1971, the secondary standard for N oxides has been reviewed three times, in 1985, 1996, and 2012 (50 FR 25532, June 19, 1985; 61 FR 52852; October 8, 1996; 77 FR 20218, April 3, 2012). Although those reviews identified additional effects related to N deposition, they all have concluded that the existing standard provided adequate protection related to the vegetation effects of airborne N oxides (i.e., the "direct" effects of N oxides in ambient air).

The existing secondary standards for PM include two $PM_{2.5}$ standards and one PM_{10} standard. The $PM_{2.5}$ standards are 35 ug/m^3 as the average of three consecutive annual 98th percentile 24-hour averages and 15.0 ug/m^3, as an annual mean concentration, averaged over three years (40 CFR 50.13). The PM_{10} standard is 150 ug/m^3 as a 24-hour average, not to be exceeded more than once per year on average over three years (40 CFR §50.6). These PM mass-based standards were most recently reviewed in the PM NAAQS review completed in 2013 with regard to protection for an array of effects that include effects on visibility, materials damage, and climate effects, as well as ecological effects. It is only the latter – ecological effects, including those related to deposition – that fall into this current review that combines consideration of these effects with the welfare effects of N oxides and SO_X. In the 2013 review, with the revision made to the form of the annual $PM_{2.5}$ standard, it was concluded that those standards provided protection for ecological effects (e.g., 78 FR 3225-3226, 3228, January 15, 2013). In reaching this conclusion, it was noted that the PA for the review explicitly excluded discussion of the effects associated with deposited PM components of N oxides and SO_X and their transformation products which were being addressed in the joint review of the secondary NO_2 and SO_2 NAAQS (78 FR 3202, January 15, 2013). The ecological effects of PM considered include direct effects on plant foliage; effects of the ecosystem loading of PM constituents such as metals or organic compounds (2009 ISA [U.S. EPA, 2009b], section 2.5.3). For all of these effects, the 2013 decision recognized an absence of information that would support any different standards and concluded the existing standards, with the revision to the form of the annual $PM_{2.5}$ standard (to remove the option for spatial averaging consistent with this change to the primary annual $PM_{2.5}$ standard), provided the requisite protection (78 FR 3086, January 15, 2013).

Table 3-1. Existing secondary standards for S oxides, PM, and N oxides.

Pollutant	Indicator	Averaging Time	Level	Form
S Oxides	SO_2	3 hours	0.5 ppm	Not to be exceeded more than once per year
PM	$PM_{2.5}$	1 year	15 μg m^{-3}	Annual mean, averaged over 3 years
		24 hours	35 μg m^{-3}	98th percentile, averaged over 3 years
	PM_{10}	24 hours	150 μg m^{-3}	Not to be exceeded more than once per year on average over 3 years
N Oxides	NO_2	1 year	53 ppb	Annual mean

3.2 PRIOR REVIEW OF DEPOSITION-RELATED EFFECTS

The most recent review of the NO_2 and SO_2 secondary standards was completed in 2012. In that review, the EPA recognized that a significant increase in understanding of the effects of N oxides and SO_X had occurred since the prior secondary standards reviews for those pollutants, reflecting the large amount of research that had been conducted on the effects of deposition of nitrogen and sulfur to ecosystems (77 FR 20236, April 3, 2012). Considering the extensive evidence available at that time, the Agency concluded that the most significant current risks of adverse effects to public welfare associated with those pollutants are those related to deposition of N and S compounds to both terrestrial and aquatic ecosystems (77 FR 20236, April 3, 2012). Accordingly, in addition to evaluating the protection provided by the secondary standards for N oxides and SO_X from effects associated with the airborne pollutants, the 2012 review also included extensive analyses of the welfare effects associated with nitrogen and sulfur deposition to sensitive aquatic and terrestrial ecosystems (77 FR 20218, April 3, 2012).

Based on the available evidence, the risks of atmospheric deposition analyzed in the 2009 REA related to two categories of ecosystem effects, acidification and nutrient enrichment (U.S. EPA, 2009a). The analyses included assessment of risks of both types of effects in both terrestrial and aquatic ecosystems. While the available evidence supported conclusions regarding the role of atmospheric deposition of S and N compounds in acidification and nutrient enrichment of aquatic and terrestrial ecosystems, there was variation in the strength of the evidence and of the information supporting the multiple quantitative linkages between the pollutants in ambient air and responses of terrestrial and aquatic ecosystems, their associated biota, and potential public welfare implications. As a result, the focus in the 2012 review with regard to consideration of a secondary standard to provide protection from deposition-related effects of was on the information related to aquatic acidification (U.S. EPA, 2011, Chapter 7).

With regard to acidification-related effects in terrestrial ecosystems, the 2009 REA had analyzed risks to sensitive tree species in the northeastern U.S. using the ecological indicator, soil BC:Al (base cations to aluminum) ratio, which has links to tree health and growth (U.S. EPA, 2009a). While the analyses indicated results of potential concern with regard to 2002 levels

of acid deposition, several uncertainties affected the strength of associated conclusions. As noted in the 2012 decision, an important drawback in understanding terrestrial acidification is related to the sparseness of available data for identifying appropriate BC:Al ratio target levels, and that the then-available data were based on laboratory responses rather than on field measurements (77 FR 20229, April 3, 2012). The 2012 decision also recognized uncertainties with regard to empirical case studies in the ISA noting that other stressors present in the field that are not present in the laboratory may confound the relationship between N oxides and SO_X deposition and terrestrial acidification effects (2008 ISA, section 3.2.2.1; 77 FR 20229, April 3, 2012). The REA analyses of aquatic acidification (which involved water quality modeling of acid deposition in case study watersheds and prediction of waterbody acid neutralizing capacity [ANC] response), however, provided strong support to the evidence for a relationship between atmospheric deposition of N and S compounds and loss of acid neutralizing capacity in sensitive ecosystems, with associated aquatic acidification effects.

Consideration of the nutrient enrichment-related effects of atmospheric N and S deposition with regard to identification of options to provide protection for deposition-related effects was limited by several factors. For example, while there is extensive evidence of deleterious effects of excessive nitrogen loadings to terrestrial ecosystems, the co-stressors affecting forests, including other air pollutants such as ozone, and limiting factors such as moisture and other nutrients, confound the assessment of marginal changes in any one stressor or nutrient in a forest ecosystem, leaving the information on the effects of changes in N deposition on forestlands and other terrestrial ecosystems limited (U.S. EPA, 2011, section 6.3.2). Further, the 2008 ISA noted that only a fraction of the deposited N is taken up by the forests, with most of the N retained in the soils (2008 ISA, section 3.3.2.1), and that forest management practices can significantly affect the nitrogen cycling within a forest ecosystem. Accordingly, the response of managed forests to N oxides deposition will be variable depending on the forest management practices employed in a given forest ecosystem (2008 ISA, Annex C, section C.6.3). Factors affecting consideration of aquatic eutrophication effects included the appreciable contributions of non-atmospheric sources to waterbody nutrient loading which affected our attribution of specific effects to atmospheric sources of N, and limitations in the ability of the available data and models to characterize incremental adverse impacts of N deposition (U.S. EPA, 2011, section 6.3.2).

Thus, in light of the evidence and findings of these analyses, and advice from the CASAC, the PA concluded it appropriate to place greatest confidence in findings related to the aquatic acidification-related effects of N oxides and SO_X relative to other deposition-related effects. Therefore, the PA focused on aquatic acidification effects from deposition of N and S compounds in identifying policy options for providing public welfare protection from

deposition-related effects of N oxides and SO_X, concluding that the available information and assessments were only sufficient at that time to support development of a standard to address aquatic acidification. Consistent with this, the PA concluded it was appropriate to consider a secondary standard in the form of an aquatic acidification index (AAI) and identified a range of AAI values (which correspond to minimum ANC levels) for consideration (U.S. EPA, 2011, section 7.6.2).

Conceptually, the AAI is an index that utilizes the results of ecosystem and air quality modeling to estimate waterbody ANC. Thus, the standard level for an AAI-based standard is a national minimum target ANC for waterbodies in the ecoregions of the U.S. for which the data were considered adequate for these purposes. While the NAAQS have historically been set in terms of an ambient air concentration, an AAI-based standard was envisioned to have a single value established for the AAI, but the concentrations of SO_X and N oxides would be specific to each ecoregion, taking into account variation in several factors that influence waterbody ANC, and consequently could vary across the U.S. The factors, specific to each ecoregion, which it was envisioned would be established as part of the standard, include: surface water runoff rates and so-called "transference ratios," which are factors applied to back-calculate or estimate the concentrations of SO_X and N oxides corresponding to target deposition values that would meet the AAI-based standard level, which is also the target minimum ANC (U.S. EPA, 2011, Chapter 7).[2] The ecoregion-specific values for these factors would be specified based on then available data and simulations of the CMAQ model, and codified as part of such a standard. As part of the standard, these factors would be reviewed in the context of each periodic review of the NAAQS.

After consideration of the PA conclusions, the Administrator concluded that while the conceptual basis for the AAI was supported by the available scientific information, there were limitations in the available relevant data, and uncertainties associated with specifying the elements of the AAI, specifically those based on modeled factors, that posed obstacles to establishing such a standard under the CAA. It was recognized that the general structure of an AAI-based standard addressed the potential for contributions to acid deposition from both N oxides and of SO_X, and quantitatively described linkages between ambient air concentrations, deposition, and aquatic acidification, considering variations in factors affecting these linkages across the country. However, the Administrator judged that the limitations and uncertainties in the available information were judged to be too great to support establishment of a new standard

[2] These were among the ecoregion-specific factors that comprised the parameters F1 through F4 in the AAI equation (2011 PA, p. 7-37). The parameter F2 represented the ecoregion-specific estimate of acidifying deposition associated with reduced forms of nitrogen, NH_X (2011 PA, p. 7-28 and ES-8 to ES-9). The 2011 PA suggested that this factor could be specified based on a 2005 CMAQ model simulation over 12-km grid cells or monitoring might involve the use of monitoring data for NH_X applied in dry deposition modeling. It was recognized that appreciable spatial variability, as well as overall uncertainty, were associated with this factor.

that could be concluded to provide the requisite protection for such effects under the Act (77 FR 20218, April 3, 2012).

These uncertainties generally related to the quantification of the various elements of the standard (the "F factors"), and their representativeness at an ecoregion scale. These uncertainties and the complexities in this approach were recognized to be unique to the 2012 review of the NAAQS for N and S oxides and were concluded to preclude the characterization and degree of protectiveness that would be afforded by an AAI-based standard, within the ranges of levels and forms identified in the PA, and the representativeness of F factors in the AAI equation described in the 2011 PA (77 FR 20261, April 3, 2012).

> "… the Administrator recognizes that characterization of the uncertainties in the AAI equation as a whole represents a unique challenge in this review primarily as a result of the complexity in the structure of an AAI based standard. In this case, the very nature of some of the uncertainties is fundamentally different than uncertainties that have been relevant in other NAAQS reviews. She notes, for example, some of the uncertainties uniquely associated with the quantification of various elements of the AAI result from limitations in the extent to which ecological and atmospheric models, which have not been used to define other NAAQS, have been evaluated. Another important type of uncertainty relates to limitations in the extent to which the representativeness of various factors can be determined at an ecoregion scale, which has not been a consideration in other NAAQS." [77 FR 20261, April 3, 2012]

The Administrator concluded that while the existing secondary standards were not adequate to provide protection against potentially adverse deposition-related effects associated with N oxides and SO_X, it was not appropriate under Section 109 to set any new or additional standards at that time to address effects associated with deposition of N and S compounds on sensitive aquatic and terrestrial ecosystems (77 FR 20262-20263, April 3, 2012).

3.3 GENERAL APPROACH FOR THIS REVIEW

As is the case for all NAAQS reviews, this secondary standards review is fundamentally based on using the Agency's assessment of the current scientific evidence and associated quantitative analyses to inform the Administrator's judgments regarding secondary standards that are requisite to protect the public welfare from known or anticipated adverse effects. The approach planned for this review of the secondary N oxides, SO_X, and PM standards will build on the last reviews, including the substantial assessments and evaluations performed over the course of those reviews, and considering the more recent scientific information and air quality data now available to inform understanding of the key policy-relevant issues in the current review.

The evaluations in the PA, including the scientific assessments in the ISA (building on prior such assessments) augmented by quantitative air quality, exposure and risk analyses, are intended to inform the Administrator's public welfare policy judgments and conclusions, including his decisions as to whether to retain or revise the standards. The PA considers the potential implications of various aspects of the scientific evidence, the air quality, exposure, or risk-based information, and the associated uncertainties and limitations. In so doing, the approach for this PA involves evaluating the available scientific and technical information to address a series of key policy-relevant questions using both evidence- and exposure/risk-based considerations.[3] Together, consideration of the full set of evidence and information available in this review will inform the answer to the following initial overarching question for the review:

- **Do the currently available scientific evidence and exposure-/risk-based information support or call into question the adequacy of the public welfare protection afforded by the current secondary standards?**

In reflecting on this question in Chapter 7 of this PA, we consider the available body of scientific evidence, assessed in the ISA (summarized in Chapters 4 and 5), and considered as a basis for developing or interpreting the quantitative information, including air quality and exposure analyses (summarized in Chapters 5 and 6), including whether it supports or calls into question the scientific conclusions reached in the last review regarding welfare effects related to SO_X, N oxides and PM in ambient air. Information available in this review that may be informative to public policy judgments on the significance or adversity of key effects on the public welfare is also considered. Additionally, the currently available exposure and risk information, whether newly developed in this review or predominantly developed in the past and interpreted in light of current information, is considered. Further, in considering this question with regard to these secondary standards, we give particular attention to exposures and risks for effects with the greatest potential for public welfare significance.

The approach to reaching conclusions on the current secondary standards and, as appropriate, on potential alternative standards, including consideration of policy-relevant questions that frame the current review, is illustrated in Figure 3-1.

[3] Generally in NAAQS reviews, the term "evidence" refers to the scientific information evaluated and interpreted in the ISA, and the term "exposure/risk" refers to quantitative analyses of air quality, exposure and risk which have also been described as Risk and Exposure Assessments. The quantitative exposure/risk analyses are developed based on the scientific information in the ISA. In this review, the exposure/risk assessment (aka REA) is focused on aquatic acidification. It is summarized in Chapter 5 and described in detail in Appendix 5A. Other quantitative information drawn from the ISA and studies assessed in the ISA is also presented in Chapter 5 and Appendix 5B.

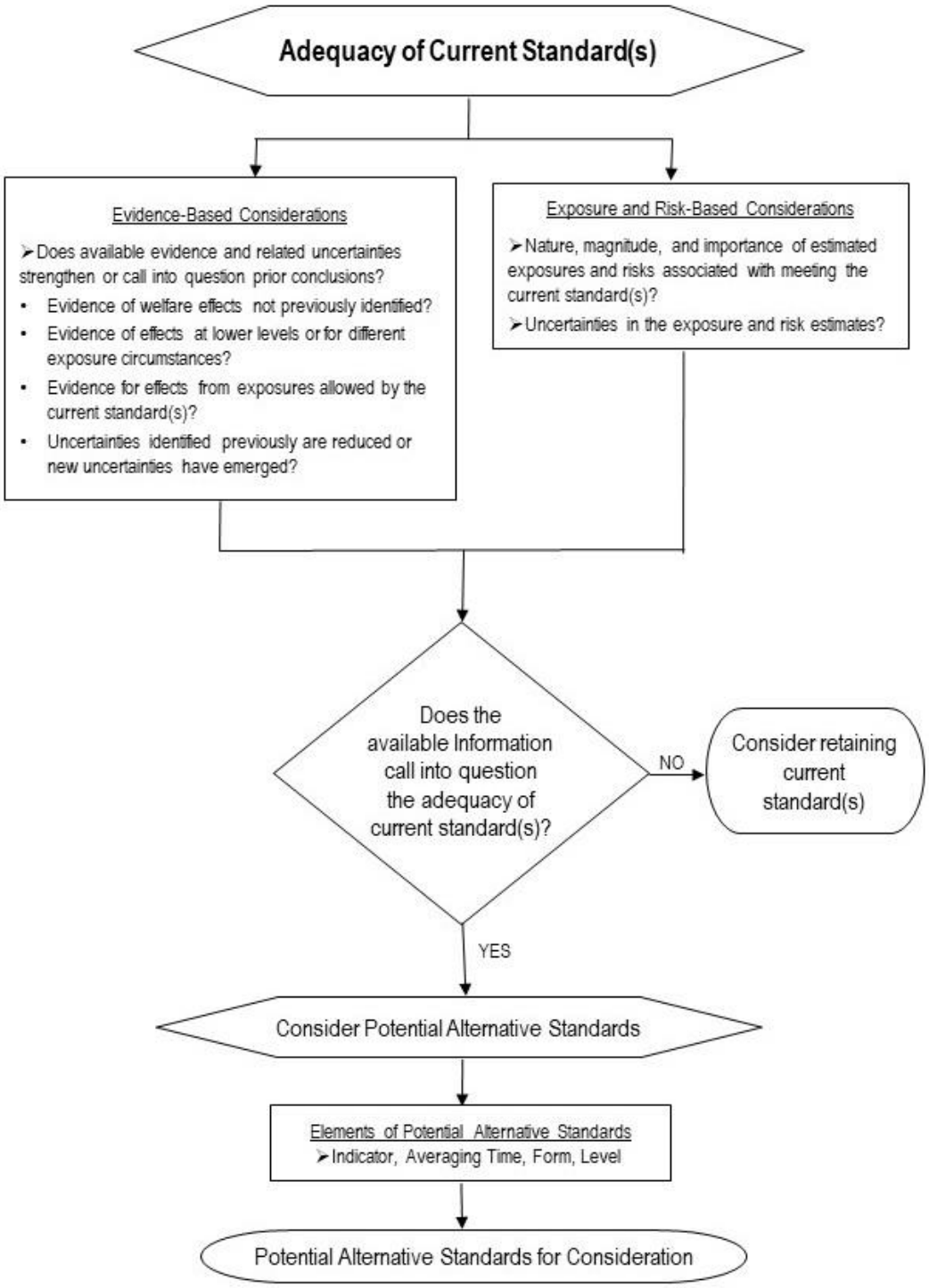

Figure 3-1. Overview of general approach for review of the secondary N oxides, SOx, and PM standards.

The Agency's approach in its review of secondary standards is consistent with the requirements of the provisions of the CAA related to the review of NAAQS and with how the EPA and the courts have historically interpreted the CAA. As discussed in section 1.2 above, these provisions require the Administrator to establish secondary standards that, in the Administrator's judgment, are requisite (i.e., neither more nor less stringent than necessary) to protect the public welfare from known or anticipated adverse effects associated with the presence of the pollutant in the ambient air. In so doing, the Administrator considers advice from the CASAC and public comment.

Consistent with the Agency's approach across all NAAQS reviews, the approach of this PA informs the Administrator's judgments based on a recognition that the available welfare effects evidence generally reflects a range of effects that include ambient air exposure circumstances for which scientists generally agree that effects are likely to occur as well as lower levels at which the likelihood and magnitude of response become increasingly uncertain. The four basic elements of the NAAQS (i.e., indicator, averaging time, form, and level) are considered collectively in evaluating the protection afforded by the current standard, or any alternative standards considered. The CAA does not require that standards be set at a zero-risk level, but rather at a level that reduces risk sufficiently so as to protect the public welfare from known or anticipated adverse effects.

The Agency's decisions on the adequacy of the current secondary standards and, as appropriate, on any potential alternative standards considered in a review, are largely public welfare policy judgments made by the Administrator. In general, conclusions reached by the Administrator in secondary NAAQS reviews on the amount of public welfare protection from the presence of the pollutant(s) in ambient air that is appropriate to be afforded by a secondary standard take into account a number of considerations, among which are the nature and degree of effects of the pollutant, including his judgments as to what constitutes an adverse effect to the public welfare, as well as, the strengths and limitations of the available and relevant information, with its associated uncertainties. Across reviews, it is generally recognized that such judgments should neither overstate nor understate the strengths and limitations of the evidence and information nor the appropriate inferences to be drawn as to risks to public welfare, and that the choice of the appropriate level of protection is a public welfare policy judgment entrusted to the Administrator under the CAA taking into account both the available evidence and the uncertainties (80 FR 65404-05, October 26, 2015). Thus, the Administrator's final decisions in such reviews draw upon the scientific information and analyses about welfare effects, environmental exposures and risks, and associated public welfare significance, as well as judgments about how to consider the range and magnitude of uncertainties that are inherent in the scientific evidence and quantitative analyses.

3.3.1 Approach for Direct Effects of the Pollutants in Ambient Air

As in past reviews of secondary standards for SO_X, N oxides and PM, this review will continue to assess the protection provided by the standards from effects of the airborne pollutants. Accordingly, this PA draws on the currently available evidence as assessed in the ISA, including the determinations regarding the causal nature of relationships between the airborne pollutants and ecological effects, which focus most prominently on vegetation, and quantitative exposure and air quality information (summarized in Chapters 4 and 5). Based on this information, we will consider the policy implications, most specifically in addressing the overarching question articulated in section 3.3 above. Building from these considerations, the PA concludes whether the evidence supports the retention or revision of the current NO_2 and SO_2 secondary standards. With regard to the effects of PM, we will take a similar approach, based on the evidence presented in the current ISA and conclusions from the review of the PM NAAQS concluded in 2013 (in which ecological effects were last considered) to assess the effectiveness of the current PM standard to protect against these types of impacts.

3.3.2 Approach for Deposition-Related Ecological Effects

In addition to evaluating the standards as to protection for effects of the airborne pollutants, we are also evaluating the standards as to protection from deposition-related effects. In so doing, we have considered the quantitative analyses conducted in the last review of the relationships between N oxides and SO_X and deposition related effects and considerations for secondary standards. The overall approach we are employing takes into account the nature of the welfare effects and the exposure conditions associated with effects in order to identify deposition-level benchmarks appropriate to consider in the context of public welfare protection. To identify metrics relevant to air quality standards (and their elements), we apply relationships developed from air quality measurements near pollutant sources and deposition estimates in sensitive ecoregions. From these, we identify an array of policy options that might be expected to provide protection from adverse effects to the public welfare. This approach is illustrated in Figure 3-2 below.

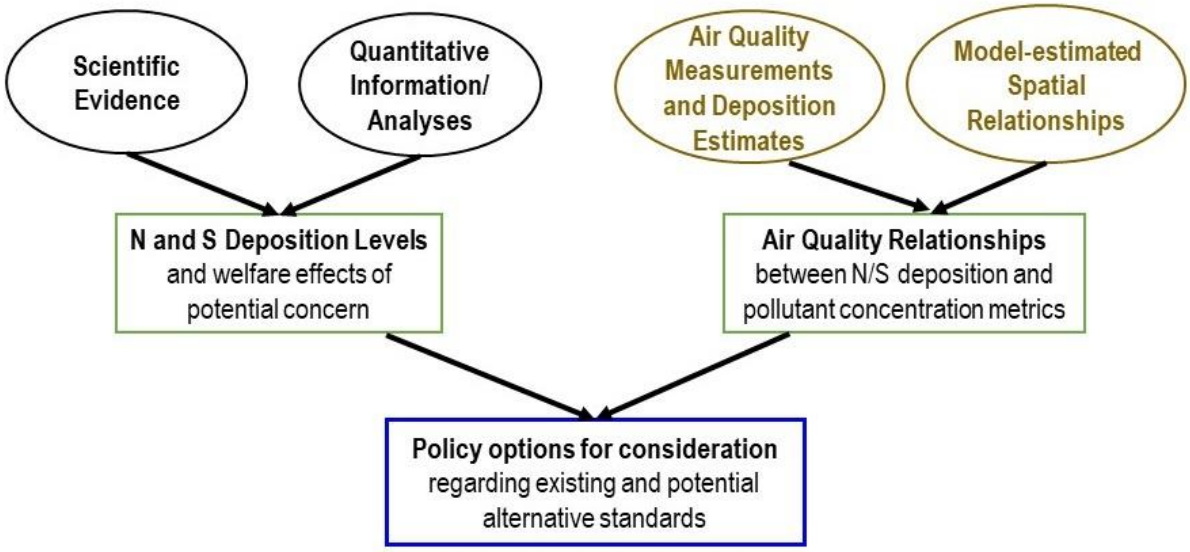

Figure 3-2. General approach for assessing the currently available information with regard to consideration of protection provided for deposition-related ecological effects on the public welfare.

Our consideration of the nature of the welfare effects draws on the overview provided in Chapter 4, based on the evidence presented in the ISA, key limitations in this evidence, and the associated uncertainties. These effects encompass both effects of airborne N oxides and SO_X, as well as deposition-related effects, including terrestrial and aquatic acidification effects, as well as effects from N enrichment. In so doing, we take note of the public welfare implications of such effects (as summarized in section 4.3).

Next, we consider the current information on exposure conditions associated with effects (Chapter 5) in order to identify deposition levels appropriate to consider in the context of public welfare protection. We investigate the extent to which the available evidence provides quantitative information linking N oxides, SO_X, and PM to deposition-related effects that can inform judgements on the likelihood of occurrence of such effects under air quality that meets the current standards. In critically assessing the available quantitative information, we recognize that the impacts of N and S deposition, which include ecosystem acidification and nutrient enrichment, are influenced by past deposition. The historical deposition associated with N oxides, SO_X, and PM in ambient air has modified soil and waterbody chemistry with associated impacts on terrestrial and aquatic ecosystems and organisms (U.S. EPA, 2020; U.S. EPA, 2008; U.S. EPA, 1982b).[4]

[4] The role of historical deposition in current ecosystem circumstances (e.g., waterbody acidification and loss of aquatic species, terrestrial acidification, and aquatic eutrophication) and the complications affecting recovery

These impacts from the dramatically higher deposition of the past century can affect how ecosystems and biota respond to more recent lower deposition rates, complicating interpretation of impacts related to more recent, lower deposition levels. This complexity is illustrated by findings of some studies that compared soil chemistry across 15–30-year intervals (1984-2001 and 1967-1997) and reported that although atmospheric deposition in the Northeast declined across those intervals, soil acidity increased (ISA, Appendix 4, section 4.6.1). As noted in the ISA, "[i]n areas where N and S deposition has decreased, chemical recovery must first create physical and chemical conditions favorable for growth, survival, and reproduction" (ISA, p. IS-102). Thus, the extent to which S and N compounds are retained in soil matrices, once deposited, with potential effects on soil chemistry, as well as ambient air concentrations and associated deposition, influence the dynamics of the response of the various environmental pathways to changes in air quality.

Based on the information summarized in Chapter 5 for aquatic and terrestrial systems, we seek to identify deposition levels associated with welfare effects of potential concern for consideration with regard to secondary standard protection. In so doing, one objective is to discern for what effects the evidence is most robust with regard to established quantitative relationships between deposition and ecosystem effects. In this context, we present an analysis of the findings in the currently available evidence, as well as additional quantitative analyses as they relate to effects of airborne N oxides, SO_X, and PM and deposition-related effects. The information for terrestrial ecosystems is derived primarily from analysis of the evidence presented in the ISA. For aquatic ecosystems, we give primary focus to aquatic acidification, for which we have conducted quantitative risk and exposure analyses based on available modeling applications (primarily based on steady-state, rather than dynamic, models) that relate acid deposition and acid neutralizing capacity in U.S. waterbodies (see section 5.1 and Appendix 5A).

In parallel with the assessments described in Chapter 5, we have utilized air quality data and trajectory-based air quality modeling to characterize atmospheric transport of the pollutants from their occurrence at monitors near their point of release to distant ecoregions where they might be expected to deposit (Chapter 6). Based on these analyses which inform an understanding of the relative contributions of source locations to individual ecoregions in the U.S., we evaluate quantitative relationships of air pollutant concentrations with atmospheric deposition rates. This includes consideration of air quality measurements near pollutant sources

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have been noted in scientific assessments for NAAQS reviews ranging from the 1982b AQCD for PM and SO_X to the current ISA (ISA, sections IS.2.3, IS.5.1.2, IS.6.1.1.1, and IS.11, Appendix 4, section 4.8.5, Appendix 6, section 6.6.3, Appendix 7, sections 7.1.5, 7.1.7, and 7.2.7, Appendix 8, sections 8.3.1.1, 8.4.1,8.4.4, 8.4.5 8.6.6, and 8.6.8, Appendix 9, 9.3.2.1, Appendix 10, section 10.2.5, Appendix 12, section 12.3.3.4; 2008 ISA, sections 3.2.1.2, 3.2.3, 3.2.4.3 and 3.2.4.4; 1982b AQCD, section 1.7 and Chapter 7).

and deposition estimates in sensitive ecoregions. We have considered existing standard metrics, as well as other potential metrics that might effectively control deposition. In so doing, we also recognize key uncertainties and limitations in relating deposition to measurements of air quality, as well as uncertainties and limitations associated with various exposure metrics. Thus, in combination with the identified deposition levels of interest, we consider the extent to which existing standards provide protection from these levels and seek to identify potential alternative standards that might afford such protection and identify an array of policy options for consideration in this review (Chapter 7).

3.3.3 Identification of Policy Options

This PA provides a range of potential policy options, supported by the science, to inform the Administrator's decisions regarding secondary standards that provide the "requisite" public welfare protection from these pollutants in ambient air. In so doing, this PA considers the evidence and quantitative analyses for direct effects of the pollutants in ambient air as well as the effects of the pollutants deposited into aquatic and terrestrial ecosystems, as described in sections 3.3.1 and 3.3.2 above, with regard to the policy-relevant questions identified for the review. Based on those considerations (discussed in Chapter 7), we consider the overarching questions for the review with regard to the extent to which the current information calls into question any of the existing standards, and the extent to which new or revised standards may be appropriate to consider. Key aspects of the available information, its limitations and associated uncertainties are discussed and conclusions reached with regard to protection from effects of the airborne pollutants and deposition-related effects. We note that the recent lower air concentrations and deposition estimates may lead to additional uncertainty in linking air quality to deposition than was the case with the higher concentrations and deposition of the past.

In considering potential alternative standards, as appropriate, we evaluate what the current information, including emissions and air quality analyses available in Chapters 2 and 6, may indicate regarding the relationships between N oxides, SO_X, and PM and N and S deposition, the influence of different averaging times, and what the quantitative analyses indicate regarding the extent to which one or more standards may have potential for controlling deposition-related and other effects of concern (Chapter 7). In so doing, we consider potential alternative standards of the same indicator and averaging time as existing standards, as well as options involving different averaging times and/or indicators, in order to inform the Administrator's judgements on the currently available information and what the available information indicates regarding what control of air quality (and as appropriate, associated deposition) may be exerted by alternative standards. Finally, the PA presents staff conclusions on whether the current evidence and quantitative analyses call into question the adequacy of

protection from ecological effects afforded by the SO_2, NO_2, and PM secondary standards, and what alternative standards may be appropriate for the Administrator to consider.

In identifying policy options appropriate to consider for providing protection from deposition-related effects, we are mindful of the long history of greater and more widespread atmospheric emissions that occurred in previous years (both before and after establishment of the existing NAAQS) and that has contributed to acidification and/or nutrient enrichment of aquatic and terrestrial ecosystems, the impacts of which exist to some extent in some ecosystems today. This historical backdrop additionally complicates policy considerations related to deposition-related effects and the identification of appropriate targets for protection in ecosystems today that might be expected to protect key ecosystem functions in the context of changing conditions over time.

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4 NATURE OF WELFARE EFFECTS

In this chapter we summarize the current evidence on the ecosystem effects of oxides of nitrogen, oxides of sulfur and particulate matter in ambient air. We consider both the evidence for direct effects of the pollutants in ambient air and for the effects of the associated atmospheric deposition into aquatic and terrestrial ecosystems. Of the welfare effects categories listed in section 302(h) of the Clean Air Act, the effects of oxides of nitrogen, oxides of sulfur and particulate matter on aquatic and terrestrial ecosystems, which encompass soils, water, vegetation, and wildlife, are the focus of this review.

In addition to direct effects of the pollutants in ambient air, oxides of N and S, and PM in ambient air contribute to deposition of N and S, as summarized in section 2.5 above, which can affect ecosystem biogeochemistry, structure, and function in multiple ways. These effects include nutrient enrichment, primarily associated with excess N, and acidification, due to N and S deposition. Both N and S are essential nutrients. Nitrogen availability, however, is sometimes the limiting factor for plant growth and productivity in aquatic and terrestrial ecosystems.[1] Accordingly, increases in the inputs of N-containing compounds to an ecosystem can affect vegetation growth and productivity, which in natural systems (both aquatic and terrestrial) can affect the relative representation and abundance of different species as a result of differing N requirements and growth characteristics among different species. Sulfur and N compounds can contribute to the acidity of terrestrial and aquatic ecosystems. The extent to which S and N deposition contribute to ecosystem acidification or to which N deposition contributes to nitrogen enrichment, and associated ecological effects, depends on characteristics of the deposited compounds and the receiving ecosystem.

Ecosystem effects considered in the currently available evidence include effects on the presence and abundance of different species, with the associated potential for changes in ecosystem function (ISA, section IS.2.2.4). The ecological metrics that have commonly been assessed, and for which there are effects related to atmospheric deposition, include species richness, community composition and biodiversity. Species richness is the number of species in a particular community and community composition additionally accounts for the number of individuals of each species. For example, two sites may both have 10 species of trees but differ in tree community composition because one may have nearly all individuals from one species and the second may have equal representation by all 10 species (ISA, section IS.2.2.4). The term

[1] In addition to N, phosphorus is the other essential nutrient for which availability sometimes is the limiting factor in plant growth and productivity, e.g., in many aquatic systems. Sulfur is rarely limiting in natural systems (ISA, Appendix 7, section 7.1 and Appendix 4, section 4.3).

biodiversity has a broader meaning intended to encompass ecosystem function and services that relate to the species composition and population sizes of the community. As numerous studies demonstrate, "the number and diversity of organisms in a system control the abundance of habitat for other species, the biogeochemical cycling of nutrients and carbon, and the efficiency at which biotic systems are able to transform limited resources into biomass" (ISA, p. IS-16).

This PA focuses on the evidence described in the 2020 ISA, and prior ISAs and AQCDs for the three criteria pollutants and focuses on effects on specific ecosystems and biological receptors from N and S deposition and both the confidence and key uncertainties associated with those effects. The summaries of this evidence below are organized to address the following questions.

- **What is the nature of the welfare effects associated with N and S and PM? Is there new evidence on welfare effects beyond those identified in the last reviews? Does the newly available evidence alter prior conclusions?**
- **What does the available evidence indicate regarding ecosystems at particular risk from deposition-related effects, and what are associated important, or key, uncertainties?**
- **What are important uncertainties in the evidence? To what extent have such uncertainties identified in the evidence in the past been reduced and/or have new uncertainties been recognized?**

The summaries in this chapter begin with the direct effects of oxides of N and S in ambient air in section 4.1, followed by subsections regarding deposition-related effects. Section 4.2 focuses on effects of deposition-related aquatic acidification, while 4.3 focuses on effects related to nitrogen enrichment. Other deposition-related effects, including those associated with PM in ambient air, are summarized in section 4.4. Lastly, section 4.5 addresses considerations of the public welfare effects given that the public welfare implications of the evidence regarding S and N related welfare effects are dependent on the type and severity of the effects, as well as the extent of the effect at a particular biological or ecological level of organization. In section 4.5, we discuss such factors here in light of judgments and conclusions made in NAAQS reviews regarding effects on the public welfare.

4.1 DIRECT EFFECTS OF OXIDES OF N AND S IN AMBIENT AIR

There is a well-established body of scientific evidence that has shown that acute and chronic exposures to oxides of N and S, such as SO_2, NO_2, NO, HNO_3 and PAN in the air, are associated with negative effects on vegetation. Such scientific evidence, as was available in 1971, was the basis for the current secondary NAAQS for oxides of sulfur and oxides of nitrogen, as summarized in section 3.1 above. The current scientific evidence continues to

demonstrate such effects, with the ISA specifically concluding that the evidence is sufficient to infer a causal relationship between gas-phase SO_2 and injury to vegetation (ISA, Appendix 3, section 3.6.12), and between gas-phase NO, NO_2 and PAN and injury to vegetation (ISA, Appendix 3, section 3.6.2). The ISA additionally concluded the evidence to be sufficient to infer a causal relationship between exposure to HNO_3 and changes to vegetation, noting that experimental exposure can damage leaf cuticle of tree seedlings and HNO_3 concentrations have been reported to have contributed to declines in lichen species in the Los Angeles basis (ISA, Appendix 3, section 3.6.3).

Uptake of gas phase N and S pollutants in a plant canopy is a complex process involving adsorption to surfaces (leaves, stems and soil) and absorption into leaves (ISA, Appendix 3, sections 3.1, 3.2 and 3.3). Several factors affect the extent to which ambient air concentrations of gas-phase N and S pollutants elicit specific plant responses. These include rate of stomatal conductance and plant detoxification mechanisms, and external factors such as plant water status, light, temperature, humidity, and pollutant exposure regime (ISA Appendix 3, sections 3.2 and 3.3). The entry of gases into a leaf depends on atmospheric chemical processes and physical characteristics of the surfaces, including the stomatal aperture. Stomatal opening is controlled largely by environmental conditions, such as water availability, humidity, temperature, and light intensity. When the stomata are closed, resistance to gas uptake is high and the plant has a very low degree of susceptibility to injury (ISA, Appendix 3, section 3.1). However, "unlike vascular plants, mosses and lichens do not have a protective cuticle barrier to gaseous pollutants, which is a major reason for their sensitivity to gaseous S and N" (ISA, Appendix 3, p. 3-2).

Specifically for SO_X, we note that high concentrations in the first half of the twentieth century have been blamed for severe damage to plant foliage that occurred near large ore smelters during that time (ISA, Appendix 3, section 3.2). In addition to foliar injury, which is usually a rapid response, SO_2 exposures have also been documented to reduce plant photosynthesis and growth. The appearance of foliar injury can vary significantly among species and growth conditions (which affect stomatal conductance). The research activity on SO_2 effects on vegetation has declined since the 1980s, especially in the U.S., due to the appreciable reductions in ambient air concentrations of SO_2 (ISA, Appendix 3, section 3.2). For lichens, damage from SO_2 exposure has been observed to include decreases in photosynthesis and respiration, damage to the algal component of the lichen, leakage of electrolytes, inhibition of nitrogen fixation, decreased potassium absorption, and structural changes (ISA, Appendix 3, section 3.2; Belnap et al., 1993; Farmer et al., 1992, Hutchinson et al., 1996).

Although there is evidence of plant injury associated with SO_2 exposures dating back more than a century (ISA, Appendix 3, section 3.2), as exposures have declined in the U.S., some studies in the eastern U.S. have reported increased growth in some SO_2-sensitive tree

species. For example, studies by Thomas et al. (2013) with eastern red cedar in West Virginia have reported significant growth rate increases in more recent years. In this study, the authors conducted a multivariate correlation analysis using historical climate variables, atmospheric CO_2 concentrations, and estimated emissions of SO_2 and NO_X in the U.S. and found that the growth of eastern red cedar trees (assessed through 100-year tree ring chronology) is explained best by increases in atmospheric CO_2 and NO_X emissions and decreases in SO_2 emissions. Although the authors attributed the growth response to reductions in SO_2-associated acid deposition, and related recovery from soil acidification, the relative roles of different pathways is unclear as a historical deposition record was not available (ISA, Appendix 3, section 3.2). Other researchers have suggested that the observed red cedar response was related to the fact that the trees were growing on a limestone outcrop that could be well buffered from soil acidification (Schaberg et al., 2014). This seems to suggest a somewhat faster recovery than might be expected from deposition-related soil acidification which may indicate a relatively greater role for changes in ambient air concentrations of SO_2, in combination with changes in other gases than was previously understood (ISA, Appendix 3, section 3.2 and Appendix 5, section 5.2.1.3).

The evidence base evaluated in the 1993 AQCD for Oxides of N included evidence of phytotoxic effects of NO, NO_2, and PAN on plants through decreasing photosynthesis and induction of visible foliar injury (U.S. EPA, 1993). The 1993 AQCD additionally concluded that concentrations of NO, NO_2, and PAN in the atmosphere were rarely high enough to have phytotoxic effects on vegetation. Little new information is available since that time on these phytotoxic effects at concentrations currently observed in the U.S. (ISA, Appendix 3, section 3.3).

The evidence for HNO_3 indicates a role in lichen species declines observed in the 1970s in the Los Angeles basin (ISA, Appendix 3, section 3.3; Boonpragob and Nash 1991; Nash and Sigal, 1999; Riddell et al., 2008). A 2008 resampling of areas shown to be impacted in the past by HNO_3 found community shifts, declines in the most pollutant-sensitive lichen species, and increases in abundance of nitrogen-tolerant lichen species compared to 1976−1977, indicating that these lichen communities have not recovered and had experienced additional changes (ISA, Appendix 3, section 3.4; Riddell et al., 2011). The recently available evidence on this topic also included a study of six lichen species that reported decreased chlorophyll content and chlorophyll fluorescence, decreased photosynthesis and respiration, and increased electrolyte leakage from HNO_3 exposures for 2-11 weeks (daily peak levels near 50 ppb) in controlled chambers. (ISA, Appendix 3, section 3.4; Riddell et al., 2012).

4.2 ACID DEPOSITION-RELATED ECOLOGICAL EFFECTS

Deposited S and N compounds can both act as acidifying agents. Acidifying deposition can affect biogeochemical processes in soils, with ramifications for terrestrial biota and for the chemistry and biological functioning of associated surface waters (ISA, Appendix 7, section 7.1). Soil acidification is influenced by the deposition of inorganic acids (HNO_3 and H_2SO_4), and by chemical and biological processes, which can also be influenced by atmospheric deposition of other chemicals. For example, NH_3 or NH_4^+ can stimulate soil bacteria that produce NO_3^- (ISA, Appendix 4, section 4.3). In this process, hydrogen ions are produced and the extent to which this changes soil acidity depends on the fate of the NO_3^-. When NO_3^-, or SO_4^{2-}, leach from soils to surface waters, an equivalent number of positive cations, or countercharge, is also transported. If the countercharge is provided by a base cation (e.g., calcium, [Ca^{2+}], magnesium [Mg^{2+}], sodium [Na^+], or potassium [K^+]), rather than hydrogen (H^+), the leachate is neutralized, but the soil becomes more acidic from the H^+ left behind and the base saturation of the soil is reduced by the loss of the base cation. Depending on the relative rates of soil processes that contribute to the soil pools of H^+ and base cations, such as weathering, continued SO_4^{2-} or NO_3^- leaching can deplete the soil base cation pool which contributes to increased acidity of the leaching soil water, and by connection, the surface water. Accordingly, the ability of a watershed to neutralize acidic deposition is determined by a variety of biogeophysical factors including weathering rates, bedrock composition, vegetation and microbial processes, physical and chemical characteristics of soils, and hydrology (ISA Appendix 4, section 4.3).

This connection between SO_2 and NO_X emissions, atmospheric deposition of N and/or S, and the acidification of acid-sensitive soils and surface waters is well documented with several decades of evidence, particularly in the eastern U.S. (ISA, section IS.5; Appendix 8, section 8.1). While there is evidence newly available since the 2008 ISA, in general, the fundamental understanding of mechanisms and biological effects has not changed. Rather, the more recent studies further support the 2008 ISA findings on these broad conclusions and provide updated information on specific aspects. An overview of the ISA findings is provided for aquatic acidification in section 4.2.1 below, and for terrestrial acidification in section 4.2.2 below.

4.2.1 Freshwater Ecosystems

Surface water processes integrate the chemicals deposited directly onto waterbodies with those released from hydrologically connected terrestrial ecosystems as a result of deposition within the watershed (ISA, Appendix 7, section 7.1). As was the case in the last review, the body of evidence regarding such processes available in this review, including that newly available, is sufficient to infer a causal relationship between N and S deposition and the alteration of freshwater biogeochemistry (ISA, section IS.6.1). Additionally, based on the previously

available evidence, the current body of evidence is also sufficient to conclude that a causal relationship exists between acidifying deposition and changes in biota, including physiological impairment and alteration of species richness, community composition, and biodiversity in freshwater ecosystems (ISA, section IS.6.3).

In addition to the acidity of surface waters quantified over weeks or months, waterbodies can also experience spikes in acidity in response to episodic events such as precipitation or rapid snowmelt that may elicit a pulse of acidic leachate over shorter periods such as hours or days. In these situations, sulfate and nitrate in snowpack (or downpours) can provide a surge or pulse of drainage water, containing acidic compounds, that is routed through upper soil horizons rather than the deeper soil horizons that usually would provide buffering for acidic compounds (ISA, Appendix 7, section 7.1). During these episodes, N and S sources other than atmospheric deposition, such as acid mine drainage or road salt applications can also be important. While some streams and lakes may have chronic or base flow chemistry that provides suitable conditions for aquatic biota, they may experience occasional acidic episodes with the potential for deleterious consequences to sensitive biota (ISA, Appendix 8, section 8.5).

4.2.1.1 Nature of Effects and New Evidence

Longstanding evidence has well characterized the changes in biogeochemical processes and water chemistry caused by N and S deposition to surface waters and their watersheds and the ramifications for biological functioning of freshwater ecosystems (ISA, Appendix 8, section 8.1). The 2020 ISA found that the newly available scientific research "reflects incremental improvements in scientific knowledge of aquatic biological effects and indicators of acidification as compared with knowledge summarized in the 2008 ISA" (ISA, Appendix 8, p. 8-80). Previously and newly available studies "indicate that aquatic organisms in sensitive ecosystems have been affected by acidification at virtually all trophic levels and that these responses have been well characterized for several decades" (ISA, Appendix 8, p. 8-80). For example, information reported in the previous 2008 ISA "showed consistent and coherent evidence for effects on aquatic biota, especially algae, benthic invertebrates, and fish that are most clearly linked to chemical indicators of acidification" (ISA, Appendix 8, p. 8-80). These indicators are surface water pH, base cation ratios, acid neutralizing capacity (ANC), and inorganic aluminum (Al_i) concentration (ISA, Appendix 8, Table 8-9).

The effects of waterbody acidification on fish species are especially well understood in the scientific literature, and many species have been documented to have experienced negative effects from acidification (ISA, Appendix 8, section 8.3). Research conducted in fresh waterbodies of Europe and North America before 1990 documented the adverse biological effects on various fish species associated with acidification (ISA, Appendix 8, section 8.3.6).

Some of the most frequently studied fish species are brown and brook trout, and Atlantic salmon, among these species the earliest lifestages are most sensitive to acidic conditions. Many effects of acidic surface waters on fish, particularly effects on gill function or structure, relate to the combination of low pH and elevated dissolved inorganic Al (ISA, Appendix 8, section 8.3.6.1).

Based on studies in the 1980s and 1990s of waterbodies affected by acidic deposition, researchers have summarized the evidence of effects on fish populations in relation to the pH and ANC of the studied waterbodies. Such effects include reduced presence of some species in acidified lakes in the Adirondacks of New York or the Appalachian Mountains (ISA, Appendix 8, section 8.3.6). Such studies have been used to characterize ranges of ANC as to potential risk to aquatic communities. The use of ANC as an indicator of waterbody acidification is described in section 4.2.1.2 below.

Despite the reductions in acidifying deposition, as summarized in section 2.5 above, aquatic ecosystems across the U.S. are still experiencing effects from historical contributions of N and S (ISA, Appendix 8, section 8.6). Long-term monitoring programs in several acid-sensitive regions of the U.S., including the Adirondacks and the northeastern U.S. have documented temporal trends in surface water chemistry that include evidence for chemical recovery in the northeastern and southeastern U.S. suggesting that full chemical recovery may take many decades or not occur at all due to the dynamics of S adsorption and desorption and long-term Ca depletion of soils (ISA, Appendix 7, section 7.1.5.1, Appendix 11, section 11.2 and Appendix 16, section 16.3.4). As reported in the 2008 ISA, biological recovery of aquatic systems lags chemical recovery due to a number of physical and ecological factors (including the time for populations to recover), as well as other environmental stressors, which make the time required for biological recovery uncertain (ISA, Appendix 8, section 8.4). Some recent studies report on waterbodies showing signs of recovery from the impacts of many decades of substantially elevated acidic deposition. One example is the successful reintroduction and re-establishment of a naturalized native fish species (brook trout) in an Adirondack Lake from which the species had been previously lost. Based on reconstruction of the historical record, the study reported ANC had increased from -2 microequivalents per liter (μeq/L) during the 1980s to 12 μeq/L during the period 2010-2012 when the trout were reintroduced. By 2012, young fish were observed, documenting successful reproduction in or in tributary streams near, the lake (ISA, Appendix 8, section 8.4.4; Sutherland et al., 2015). Another recent study in the Adirondack Lake region however, found no evidence of widespread or substantial brook trout recovery, although water quality had improved, indicating the impact of the factors mentioned above that can contribute to lags of biological recovery behind chemical recovery (ISA, Appendix 8, sections 8.4 and 8.4.4).

4.2.1.2 Freshwater Ecosystem Sensitivity

The effects of acid deposition on aquatic systems depend largely upon the ability of the system to neutralize additional acidic inputs from the environment, whether from the atmosphere or from surface inputs. There is a large amount of variability between freshwater systems in this regard which reflects their underlying geology as well as previous acidic inputs. Accordingly, different freshwater systems (e.g., in different geographic regions) respond differently to similar amounts of acid deposition. The main factor in determining sensitivity is the underlying geology of an area and its ability to provide soil base cations through weathering to buffer acidic inputs (ISA, Appendix 8, section 8.5.1). As noted in the ISA, "[g]eologic formations having low base cation supply, due mainly to low soil and bedrock weathering, generally underlie the watersheds of acid-sensitive lakes and streams" (ISA, Appendix 8, p. 8-58). Consistent with this, studies have indicated that the thickness of the till (the sediment layer deposited by action of receding glaciers) "has been shown to be a key control on the pH and ANC of Adirondack lakes" (ISA, Appendix 8, p. 8-58). Other factors identified as contributing to the sensitivity of surface waters to acidifying deposition, include topography, soil chemistry and physical properties, land use and history, and hydrologic flowpath, as well as impacts of historic, appreciably higher, deposition (ISA, Appendix 8, p. 8-58).

Acid neutralizing capacity is commonly used to describe the potential sensitivity of a freshwater system to acidification-related effects and has been found in various studies to be the single best indicator of the biological response and health of aquatic communities in acid sensitive systems (ISA, Appendix 8, section 8.6). The parameter ANC is an indicator of the buffering capacity of natural waters against acidification. Although ANC does not directly affect biota, it is a indicator of acidification that relates to pH and aluminum levels, and biological effects in aquatic systems are primarily attributable to low pH and high inorganic aluminum concentration (ISA, p. ES-14). Acid neutralizing capacity is parameter that can be measured in water bodies. It is also often estimated for use in water quality modeling, as the molar sum of strong base cations minus the molar sum of strong acid anions (specifically including SO_4^{2-} and NO_3^-) (e.g., Driscoll et al., 1994). Water quality models are generally better at estimating ANC than at estimating other indicators of acidification-related risk. While ANC is not the direct cause of acidification-related effects on aquatic biota, it serves as an indicator of acidification-related risk, since it has been related to the health of biota and to other surface water constituents like pH and Al or watershed characteristics like base cation weathering (BCw) rate (ISA, Appendix 8, sections 8.1 and 8.3.6.3). Waterbody pH largely controls the bioavailability of Al, which is toxic to fish (ISA, Appendix 8, section 8.6.4). Values of ANC can also be influenced by high concentrations of naturally occurring organic acids (Waller et al. 2012). In waters where that occurs, ANC may not be a good indicator of risk to biota as the organic compounds can reduce

bioavailability of Al, buffering effects usually associated with low pH and high total Al concentrations (ISA, Appendix 8, section 8.3.6.4).

In its role as an indicator, ANC levels are commonly used to categorize waterbody sensitivity. Waterbodies with annual average levels above 100 are generally not considered sensitive or at risk of acidification-related effects. There is potential for risk at lower levels, at which consideration of other factors can inform interpretation. National survey data dating back to the early 1980s that were available for the 2008 ISA indicated acidifying deposition had acidified surface waters in the southwestern Adirondacks, New England uplands, eastern portion of the upper Midwest, forested Mid-Atlantic highlands, and Mid-Atlantic coastal plain (2008 ISA, section 4.2.2.3; ISA, Appendix 8, section 8.5.1). As noted in section 4.2.1 above, events such as spring snowmelt and heavy rain events can contribute to episodic acidification events. For example, in some impacted northeastern waterbodies, ANC levels may dip below zero for hours to days or weeks in response to such events, while waterbodies labeled chronically acidic have ANC levels below zero throughout the year (ISA, Appendix 6, section 6.1.1.1; Driscoll et al., 2001). Accordingly, headwater streams in both the eastern and western U.S. tend to be more sensitive to such episodes due to their smaller size (ISA, Appendix 8, section 8.5.1).

Fish and water quality surveys as well as *in situ* bioassays inform our understanding of risk posed to fish species across a range of ANC. For example, surveys in the heavily impacted Adirondack mountains found that waterbodies with ANC levels near/below zero[2] and pH near/below 5.0 generally had few or no fish species (Sullivan et al., 2006; ISA, Appendix 8, section 8.6). Waterbodies with levels of ANC above zero differed in the types and numbers of species present. At relatively lower ANC levels such as below 20 μeq/L, comparatively acid tolerant species such as brook trout can have healthy populations, but sensitive fish species such as Atlantic salmon smolts, blacknose shiner, and other fish can be absent, or their population can be greatly reduced. While most sensitive species were not lost from the aquatic system, their fitness (population size and growth) declined; plankton and macroinvertebrate assemblages were also impacted somewhat; and fish species richness in some areas was lower, with fewer of the most sensitive species present. Some sites with ANC levels above 80 μeq/L have appeared unimpaired (Bulger et al., 1999; Driscoll et al., 2001; Kretser et al., 1989; Sullivan et al., 2006). An ANC level of 100 μeq/L is often identified as a benchmark at/below which waterbodies may be considered at increased sensitivity.

Surveys conducted from the 1980s through 2004, available in the last review, indicated that the surface waters in the southwestern Adirondacks, New England uplands, eastern portion

[2] A survey of waterbodies in the Adirondacks in 1984-1987 found 27% of streams to have ANC values below zero, with a minimum value of -134 μeq /L (Sullivan et al., 2006). Values of ANC below 20 in Shenandoah stream sites were associated with fewer fish of sensitive species compared to sites with higher ANC (Bulger et al., 1999).

of the upper Midwest, forested Mid-Atlantic highlands, and Mid-Atlantic coastal plain had been acidified as a result of acidifying deposition (ISA, Appendix 8, section 8.5.1). A compilation of historical water quality measurements of ANC from 1980 to 2011 (nearly 200,000 measurements at nearly 20,000 spatially unique sites) is presented in Figure 4-1 below (Sullivan, 2017).[3] As described in the ISA, "[a]cidic waters were mostly restricted to northern New York, New England, the Appalachian Mountain chain, upper Midwest, and Florida" (ISA, Appendix 8, p. 8-60). Additionally, the figure indicates low, but positive, ANC values for these same regions, as well as high-elevation western waterbodies (e.g., in the Sierra and Cascades mountains) and parts of Arkansas and the Gulf states (Figure 4-1; ISA, Appendix 8, section 8.5.2). The findings for high-elevation portions of the West and parts of Arkansas and the Gulf states are thought to largely reflect base cation supply in soils, as levels of acidifying deposition have been low in most areas of the West, and acidic surface waters there are rare (ISA, Appendix 8, section 8.5.2).

[3] Samples expected to be strongly influenced by acid mine drainage, sea salt spray, or road salt application were excluded. Among the full dataset, 6,065 sites had ANC < 100 μeq/L.

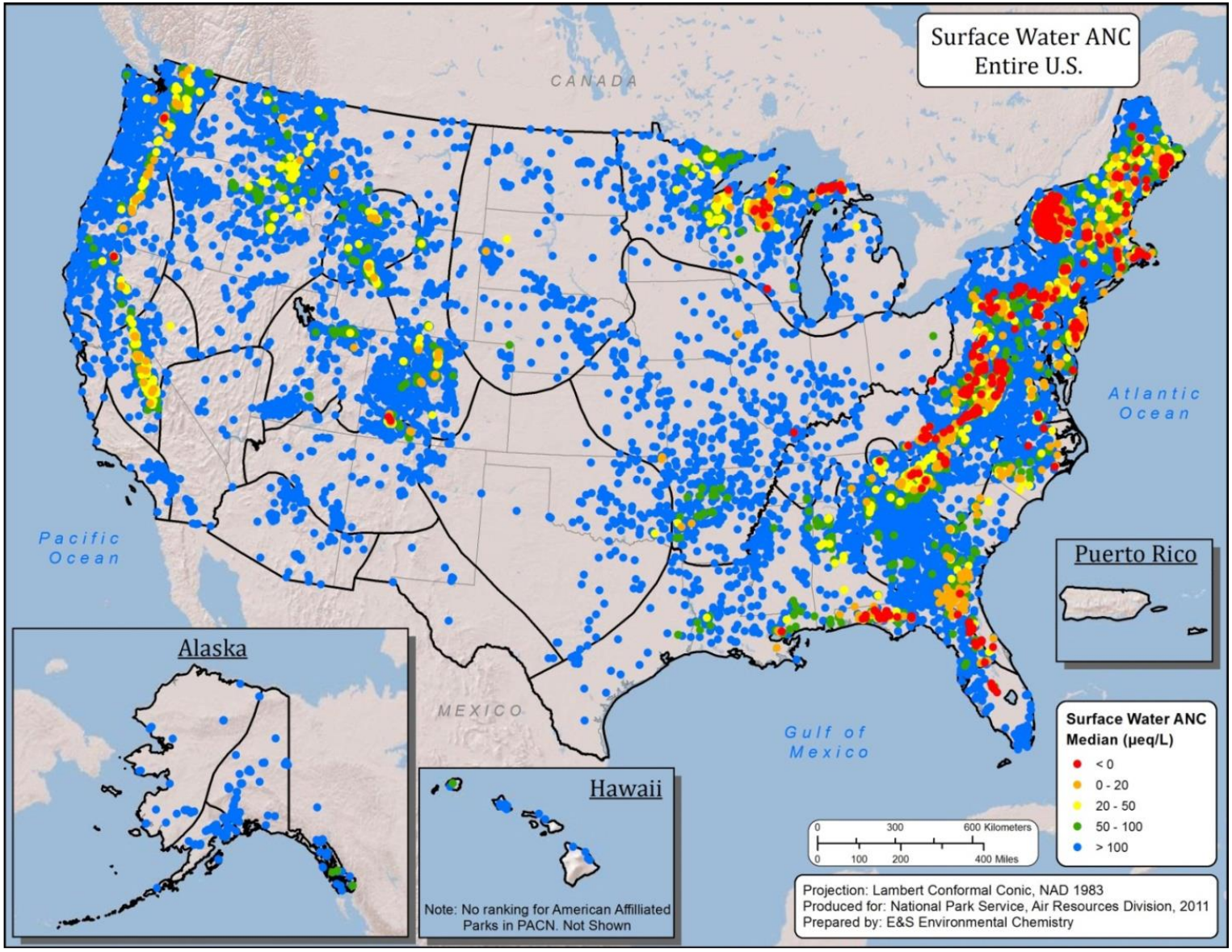

Figure 4-1. Surface water ANC map, based on data compiled by Sullivan (2017) (ISA, Appendix 8, Figure 8-11).

4.2.1.3 Key Uncertainties

In the longstanding evidence base for acidification effects of deposited S and N in aquatic ecosystems, uncertainties remain. Key uncertainties include those associated with inputs to models that simulate watershed chemistry and are employed to estimate waterbody buffering capacity, such as base cation weathering rates and leaching of S and N compounds from watershed soils. Uncertainties are associated with estimates of the response of waterbodies to different deposition levels in areas for which site-specific data are not available because of the high spatial variability of the factors that influence watershed sensitivity (ISA, Appendix 8, section 8.5.1; McNulty et al., 2007). For example, there are uncertainties related to limitations in water quality measurements, data on surface runoff characteristics, and other factors important to characterizing watershed supplies of base cations related to weathering of bedrock and soils. There are also uncertainties associated with our understanding of relationships between ANC and risk to native biota, particularly in waterbodies in geologic regions prone to waterbody acidity. These relate to the varying influences of site-specific factors other than ANC.

4.2.2 Terrestrial Ecosystems

There is longstanding evidence that changes in soil biogeochemical processes caused by acidifying deposition of N and S to terrestrial systems are linked to changes in terrestrial biota, with associated impacts on ecosystem characteristics. The currently available evidence, including that newly available in this review, supports and strengthens this understanding (ISA, Appendix 5, section 5.1). Consistent with conclusions in the last review the current body of evidence is sufficient to infer a causal relationship between acidifying deposition and alterations of biogeochemistry in terrestrial ecosystems. Additionally, and consistent with conclusions in the last review, the current body of evidence is sufficient to infer a causal relationship between acidifying N and S deposition and the alteration of the physiology and growth of terrestrial organisms and the productivity of terrestrial ecosystems. The current body of evidence is also sufficient to conclude that a causal relationship exists between acidifying N and S deposition and alterations of species richness, community composition, and biodiversity in terrestrial ecosystems (2008 ISA, Appendix 4, sections 4.2.1.1 and 4.2.1.2; 2020 ISA, Appendix 4, section 4.1 and Appendix 5, sections 5.7.1 and 5.7.2).

4.2.2.1 Nature of Effects and New Evidence

Deposition of acidifying compounds to acid-sensitive soils can cause soil acidification, increased mobilization of Al from soil to drainage water, and deplete the pool of exchangeable base cations in the soil (ISA, Appendix 5, section 5.2 and Appendix 4, sections 4.3.4 and 4.3.5). The physiological effects of acidification on terrestrial biota include slower growth and increased mortality among sensitive plant species, which are generally attributable to physiological

impairment caused by Al toxicity (related to increased availability of inorganic Al in soil water) and a reduced ability of plant roots to take up base cations (ISA, Appendix 4, section 4.3 and Appendix 5, section 5.2). The U.S. tree species most studied with regard to effects of acid deposition are red spruce and sugar maple, although there is also evidence for other tree species such as flowering dogwood (ISA, Appendix 5, section 5.2.1). The recently available evidence includes Ca addition experiments in which Ca is added to acidic soils and physiological and growth responses of red spruce and sugar maple are assessed to help understand the response of these species to the soil changes induced by acid deposition (ISA, Appendix 5, Table 5-2). Other recent studies have included addition or gradient studies evaluating relationships between soil chemistry indicators of acidification (e.g., soil pH, Bc:Al ratio, base saturation, and Al) and ecosystem biological endpoints, including physiological and community responses of trees and other vegetation, lichens, soil biota, and fauna (ISA, Appendix 5, Table 5-6).

Since the last review of the NAAQS for oxides of S and N, and as described in detail in Chapter 5 (and Appendix 5B), several observational studies have reported on statistical associations between tree growth or survival, as assessed at monitoring sites across the U.S. and estimates of average deposition of S or N compounds at those sites over time periods on the order of 10 years (section 5.3.2.3 and Appendix 5B, section 5B.2.2 below; ISA, Appendix 5, section 5.5.2 and Appendix 6, section 6.2.3.1; Dietze and Moorcroft, 2011; Thomas et al., 2010; Horn et al., 2018). Negative associations were observed for survival and growth in several species or species groups with S deposition metrics; positive and negative associations were reported with N deposition (see section 5.3.2.3 and 5.3.4 below and Appendix 5B).

The physiological effects of acidifying deposition on terrestrial biota can also result in changes in species composition whereby sensitive species are replaced by more tolerant species, or the sensitive species that were dominant in the community become a minority. For example, increasing soil cation availability (as in Ca addition or gradient experiments) was associated with greater growth and seedling colonization for sugar maple while American beech was more prevalent on soils with lower levels of base cations where sugar maple is less often found (ISA, Appendix 5, section 5.2.1.3.1; Duchesne and Ouimet, 2009). In a study of understory species composition, soil acid-base chemistry was found to be a predictor of understory species composition (ISA, Appendix 5, section 5.2.2.1). Additionally, limited evidence, including a recent S addition study and agricultural soil gradient study, indicated that soil acid-base chemistry predicted and was correlated with diversity and composition of soil bacteria, fungi, and nematodes (ISA, Appendix 5, section 5.2.4.1).

4.2.2.2 Terrestrial Ecosystem Sensitivity

Underlying geology is the principal factor governing the sensitivity of both terrestrial and aquatic ecosystems to acidification from S and N deposition. Geologic formations with low base cation supply (e.g., sandstone, quartzite), due mainly to low weathering rates, generally underlie these acid sensitive watersheds. Other factors also contribute to the overall sensitivity of an area to acidifying nitrogen and sulfur deposition including topography, soil chemistry, land use, and hydrology (ISA, Appendix 5, section 5.3). As observed in the ISA, "[a]cid-sensitive ecosystems are mostly located in upland mountainous terrain in the eastern and western U.S. and are underlain by bedrock that is resistant to weathering, such as granite or quartzite sandstone" (ISA, Appendix 7, p. 7-45). Further, as well documented in the evidence, biogeochemical sensitivity to deposition-driven acidification (and eutrophication [see section 4.3 below]) is the result of historical loading, geologic/soil conditions (e.g., mineral weathering and S adsorption), and nonanthropogenic sources of N and S loading to the system (ISA, Appendix 7, section 7.1.5).

Several different indicators are commonly used to identify areas at increased risk of acidification processes (ISA, Appendix 5, Table 4-1). They include the ratio of the molar sum of base cations to the molar amount of Al (BC:Al) in soil solution. The BC:Al ratio is commonly used, particularly in mass balance modeling approaches, such as the simple mass balance equation (SMBE; also referred to as the simple mass balance, SMB, model), that are intended to assess the vulnerability of different areas to acidification as a result of atmospheric deposition of N and S compounds. Higher values of this ratio indicate a lower potential for acidification-related biological effects (ISA, Table IS-2). The ratio value can be reduced by release of base cations from the soil (e.g., through the process of neutralizing drainage water acidity) which, in turn, reduces the base saturation of the soil. Soil base saturation[4] and changes to it can also be an indicator of acidification risk (ISA, Appendix 4, section 4.3.4). The accelerated loss of base cations through leaching, decrease in base saturation, and decreases in the BC:Al ratio all serve as indicators of soil acidification. This is because the input of base cations to soil solution, e.g., via soil weathering or base cation exchange, can neutralize inorganic and organic acids (ISA, Appendix 4, section 4.3).

Although there has been no systematic national survey of U.S. terrestrial ecosystem soils, several forest ecosystems are considered the most sensitive to terrestrial acidification from atmospheric deposition. These include forests of the Adirondack Mountains of New York, Green Mountains of Vermont, White Mountains of New Hampshire, the Allegheny Plateau of Pennsylvania, and mountain top and ridge forest ecosystems in the southern Appalachians (2008

[4] Soil base saturation expresses the concentration of exchangeable bases (Ca, Mg, potassium [K], sodium [Na]) as a percentage of the total cation exchange capacity (which includes exchangeable H^+ and inorganic Al).

ISA, Appendix 3, section 3.2.4.2; ISA, Appendix 5, section 5.3). A number of modeling analyses, including a national-scale analysis, have been performed to identify acid-sensitive areas, generally through estimates of indicators such as BC:Al (ISA, Appendix 5, sections 5.3, 5.4 and 5.5). In some cases, more recent analyses augment estimates from the previously available national-scale analysis (McNulty et al., 2007), potentially providing updated estimates. For example, a recent modeling analysis by Phelan et al. (2014) employed the PROFILE model to estimate BC_W in support of SMB modeling, a difference from the empirical approach (clay correlation-substrate method) used by McNulty et al. (2007). This more recent analysis suggested that Pennsylvania hardwood sites may not be as sensitive to acidifying deposition as previously estimated (ISA, Appendix 5, section 5.4; Phelan et al., 2014). Another commonly used indicator of acidification is soil base saturation (ISA, Appendix 4, Table 4-1). Values below 10% have been associated with areas experiencing acidification such as the eastern forests recognized above (ISA, Appendix 4, section 4.3.4).

Recently available evidence includes some studies describing early stages of recovery from soil acidification in some eastern forests. For example, studies at the Hubbard Brook Experimental Forest in New Hampshire reported indications of acidification recovery in soil solution measurements across the period from 1984 to 2011 (ISA, Appendix 4, section 4.6.1; Fuss et al., 2015). Another study of 27 sites in eastern Canada and the northeastern U.S. reported reductions in wet SO_4^{2-} deposition to be positively correlated with changes in base saturation and negatively correlated with changes in exchangeable Al between initial samplings in the mid-1980s to early 1990s and a resampling in the period 2003-2014. That is, reductions in wet deposition SO_4^{2-} were associated with increases in soil base saturation and decreases in exchangeable Al (ISA, Appendix 4, section 4.6.1; Lawrence et al., 2015). Modeling analyses indicate extended timeframes for recovery are likely, as well as delays or lags related to accumulated pools of S in forest soils (ISA, Appendix 4, section 4.6.1).

4.2.2.3 Key Uncertainties

Although the evidence clearly demonstrates that N and S deposition causes acidification related effects in terrestrial ecosystems, uncertainties remain that are important to our consideration of the evidence in this review. For example, there are uncertainties associated with the various approaches for estimating sensitive ecosystems and for understanding and characterizing long-term risks and processes against the backdrop of deposition reductions occurring over the past several decades. As summarized in section 4.2.2.1 above, modeling analyses are commonly employed, with several inputs recognized as contributing to overall uncertainty.

As noted in the ISA, the rate of base cation weathering "is one of the most influential yet difficult to estimate parameters" in modeling (such as the SMB) that estimate indicators of acidification as a function of deposition inputs (ISA, Appendix 4, section 4.5.1.1). Estimating this parameter continues to be a major source of uncertainty in such modeling. For example, in an analysis of uncertainties associated with simulating ANC in waterbodies of interest in response to acid deposition over a broad spatial scale, the primary source of uncertainty was identified to be from factors affecting base cation weathering and ANC, including BCw rates, soil depth and soil temperature (ISA, p. IS-114; Li and McNulty, 2007). The authors concluded that improvements in estimates of these factors are crucial to reducing uncertainty and successful model application for broader scales (e.g., where site-specific information is limited), including national scale (ISA, Appendix 4, section 4.6). Another analysis of major sources of uncertainty related to estimating soil acidification also found the greatest uncertainty to be associated with the BCw estimates, particularly citing the particle size class-based method commonly used to estimate the total specific surface area upon which weathering reactions can take place (Whitfield et al., 2018).

There are also more general sources of uncertainty associated with observational or gradient studies that relate variation in biological/ecological indices to variation in deposition metrics. For example, such studies may fail to account for influences such as variation in biological and biogeochemical processes imposed by climate, geology, biota, and other environmental factors. Further, observed variation in current or recent biological metrics may be affected by the lags reported in the evidence, both in ecosystem response to acid deposition and to ecosystem recovery from historic deposition. Additionally, biological measures in areas for which recent values of deposition metrics are relatively low, may be influenced by impacts from past deposition.

4.3 NITROGEN ENRICHMENT AND ASSOCIATED EFFECTS

The numerous ecosystem types that occur across the U.S. have a broad range of sensitivity to N enrichment. Organisms in their natural environments are commonly adapted to the nutrient availability in those environments. Historically, N has been the primary limiting nutrient in many ecosystems. In such ecosystems, when the limiting nutrient, N, becomes more available, whether from atmospheric deposition, runoff, or episodic events, the subset of species able to most effectively utilize the higher nitrogen levels may out-compete other species leading to a shift in the community composition that may be dominated by a smaller number of species (i.e., a community with lower diversity) (ISA, sections IS.6.1.1.2, IS.6.2.1.1 and IS.7.1.1, Appendix 6, section 6.2.4 and Appendix 7, section 7.2.6.6). Thus, change in the availability of nitrogen in nitrogen-limited systems can affect growth and productivity, with ramifications on

relative abundance of different species, and potentially further and broader ramifications on ecosystem processes, structure, and function. The term, eutrophication, refers to such processes that occur in response to enrichment of a system with nutrients. A common example of eutrophication in aquatic ecosystems is when increased loading of the limiting nutrient (usually N or phosphorus) results in rapid and appreciable algal growth. Decomposition of the plant biomass from the subsequent algal die-off contributes to reduced waterbody oxygen which in turn contributes to fish mortality (ISA, p. ES-18).

Both N oxides and reduced forms of nitrogen (NH_X) can contribute to N enrichment. In addition to atmospheric deposition, other sources of S and N can play relatively greater or lesser roles in contributing to S and N inputs, depending on location. For example, many waterbodies receive appreciable amounts of N from agricultural runoff and municipal or industrial wastewater discharges. For many terrestrial and freshwater ecosystems, sources of N other than atmospheric deposition, including fertilizer and waste treatment, contribute to ecosystem total N with contributions that can be larger than that from atmospheric deposition (ISA Appendix 7, sections 7.1 and 7.2). Additionally, the impacts of historic deposition in both aquatic and terrestrial ecosystems pose complications to discerning the potential effects of more recent lower deposition rates.

4.3.1 Aquatic and Wetland Ecosystems

Nitrogen additions, including from atmospheric deposition, to freshwater, estuarine and near-coastal ecosystems can contribute to eutrophication which typically begins with nutrient-stimulated rapid algal growth developing into an algal bloom that can, depending on various site-specific factors, be followed by anoxic conditions associated with the algal die-off (ISA, section ES.5.2). This reduction in dissolved oxygen can affect higher-trophic-level species (ISA, section ES.5.2). The extensive body of evidence in this area is sufficient to infer causal relationships between N deposition and the alteration of biogeochemistry in freshwater, estuarine and near-coastal marine systems (ISA, Appendix 7, sections 7.1 and 7.2). Further, consistent with findings in the last review, the current body of evidence is sufficient to infer a causal relationship between N deposition and changes in biota, including altered growth and productivity, species richness, community composition, and biodiversity due to N enrichment in freshwater ecosystems (ISA, Appendix 9, section 9.1). The body of evidence is sufficient to infer a causal relationship between N deposition and changes in biota, including altered growth, total primary production, total algal community biomass, species richness, community composition, and biodiversity due to N enrichment in estuarine environments (ISA, Appendix 10, section 10.1).

The impact of N additions on wetlands depends on the type of wetland and other factors. More specifically, the type of wetland, as well as hydrological conditions and season, influence

whether a wetland serves as a source, sink, or transformer of atmospherically deposited N (ISA, section IS.8.1 and Appendix 11, section 11.1). One of the transformations that may occur in wetlands is denitrification which leads to the production of N_2O, a greenhouse gas. This is a normal process in anaerobic soils but can be increased with the introduction of additional N, especially when in reduced forms such as NH_4^+ (ISA, Appendix 4, section 4.3.6). Whether wetlands are a source or a sink of N is extremely variable and depends on vegetation type, physiography, and local hydrology, as well as climate. Studies generally show N enrichment to decrease the ability of wetlands to retain and store N, which may diminish the wetland ecosystem service of improving water quality (ISA, section IS.8.1). Consistent with the evidence available in the last review, the current body of evidence is sufficient to infer a causal relationship between N deposition and the alteration of biogeochemical cycling in wetlands. Newly available evidence regarding N inputs and plant physiology, expands the evidence base related to species diversity. The currently available evidence, including that newly available, is sufficient to infer a causal relationship between N deposition and the alteration of growth and productivity, species physiology, species richness, community composition, and biodiversity in wetlands (ISA, Appendix 11, section 11.10).

4.3.1.1 Nature of Effects and New Evidence

As summarized above, N inputs and other factors contribute to nutrient enrichment which contribute to eutrophication, the process of enriching a water body with nutrients resulting in increased growth and change in the composition of primary producers (algae and/or aquatic plants) which can also lead to low oxygen levels in the water body when these primary producers decompose. Such nitrogen driven eutrophication alters freshwater biogeochemistry and can impact physiology, survival, and biodiversity of sensitive aquatic biota (Figure 4-2).

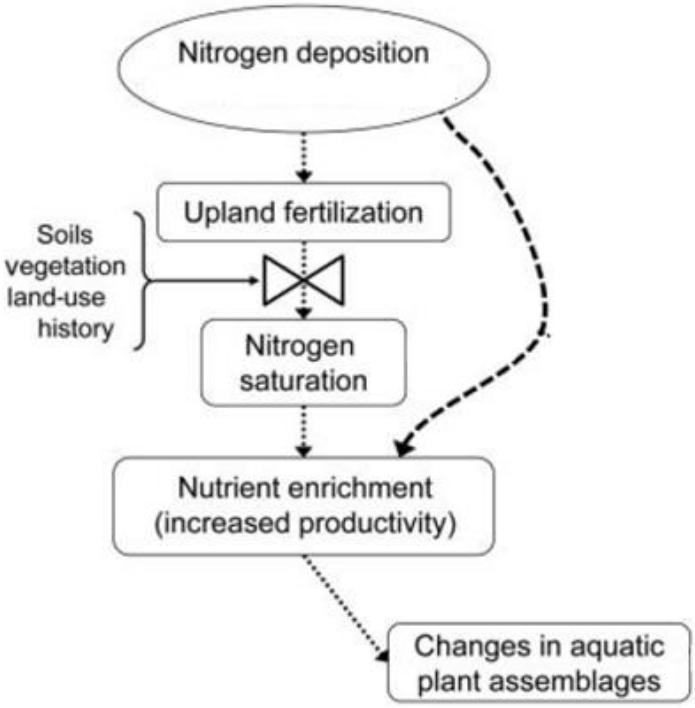

Figure 4-2. Conceptual model of the influence of atmospheric N deposition on freshwater nutrient enrichment (ISA, Appendix 9, Figure 9-1).

Evidence newly available in this review provides insights regarding N enrichment and its impacts in several types of aquatic systems, including freshwater streams and lakes, estuarine and near-coastal systems, and wetlands. For example, studies published since the 2008 ISA augment the evidence base for high-elevation waterbodies where the main source of N is atmospheric deposition, including a finding that N deposition is correlated with a shift from N to P limitation in certain water bodies (ISA, Appendix 9, section 9.1.1.3). The newly available evidence, including that from paleolimnological surveys, fertilization experiments, and gradient studies continues to show effects of N loading to sensitive freshwater systems, including an influence on the occurrence of harmful algal blooms (ISA, Appendix 9).

More specifically, the availability and form of N has been found to influence freshwater algal bloom composition and toxicity (ISA, Appendix 9, section 9.2.6.1). Information available in this review indicates that growth of some harmful algal species, including those that produce microcystin, are favored by increased availability of N and its availability in dissolved inorganic form (ISA, Appendix 9, p. 9-28). For example, studies in Lake Erie have indicated Microcystis bloom growth and microcystin concentration were stimulated more frequently to N additions than phosphorus additions (Davis et al., 2015). Further, inorganic N was also associated with peak surface water concentrations of microcystin, a cyanobacteria produced toxin that is enriched in N (Gobler et al., 2016).

Evidence of the influence of availability and form of N on algal blooms is also available in estuarine systems. For example, specific phytoplankton functional groups prefer reduced forms of N (such as NH_4^+) over oxidized forms (such as NO_3^-), and in many parts of the U.S., including the Southeast and Mid-Atlantic, reduced N deposition has increased relative to oxidized N deposition (ISA, Appendix 10, section 10.3.3). Very limited evidence suggests a role for atmospheric N deposition in taxonomic shifts and declines in some invertebrates, although "the effects attributed to N are difficult to separate from other stressors such as climate change and invasive species" (ISA, Appendix 9, section 9.6).

Evidence in coastal waters has recognized a role of nutrient enrichment in acidification of some coastal waters (ISA, Appendix 10, section 10.5). More specifically, nutrient-driven algal blooms may contribute to ocean acidification, possibly through increased decomposition which lowers dissolved oxygen levels in the water column and contributes to lower pH. Such nutrient-enhanced acidification can also be exacerbated by warming (associated with increased microbial respiration) and changes in buffering capacity (alkalinity) of freshwater inputs (ISA, Appendix 10, section 10.5).

4.3.1.2 Aquatic Ecosystem Sensitivity

Current evidence continues to support the conclusions of the previous review regarding ecosystem sensitivity to nutrient enrichment.

4.3.1.2.1 Freshwater Ecosystems

Freshwater systems that are likely to be most impacted by nutrient enrichment due to atmospheric deposition of N are remote, oligotrophic, high-elevation water bodies with limited local nutrient sources and with low N retention capacity. Freshwater systems sensitive to N nutrient enrichment include those in the Snowy Range in Wyoming, the Sierra Nevada Mountains, and the Colorado Front Range. A portion of these lakes and streams where effects are observed are in Class I wilderness areas (Williams et al., 2017a; Clow et al., 2015; Nanus et al., 2012).

Recent research also supports the 2008 ISA findings that N limitation is common in oligotrophic waters in the western U.S. (Elser et al., 2009b; Elser et al., 2009a). Shifts in nutrient limitation, from N limitation, to between N and P limitation, or to P limitation, were reported in some alpine lake studies reviewed in the 2008 ISA and in this review. Since the 2008 ISA, several meta-analyses have reported an increase in P deposition to water bodies (Stoddard et al., 2016; Brahney et al., 2015; Tipping et al., 2014) and highlight the need to account for how sustained P deposition can modify the effects of anthropogenically emitted N deposition on productivity. Even small inputs of N in these water bodies can increase nutrient availability or

alter the balance of N and P, which can stimulate growth of primary producers and lead to changes in species richness, community composition, and diversity.

The relative contribution of N deposition to total N loading varies among waterbodies. For example, atmospheric deposition is generally considered to be the main source of new N inputs to most headwater stream, high-elevation lake, and low-order stream watersheds that are far from the influence of other N sources like agricultural runoff and wastewater effluent (ISA, section ES5.2). In other fresh waterbodies, however, agricultural practices and point source discharges have been estimated to be larger contributors (ISA, Appendix 7, section 7.1.1.1).

Since the 2008 ISA, several long-term monitoring studies in the Appalachian Mountains, the Adirondacks, and the Rocky Mountains have reported temporal patterns of declines in surface water NO_3^- concentration corresponding to declines in atmospheric N deposition (ISA, Appendix 9, section 9.1.1.2). Declines in basin wide NO_3^- concentrations have also been reported for the nontidal Potomac River watershed and attributed to declines in atmospheric N deposition (ISA, Appendix 7, section 7.1.5.1). A study of water quality monitoring in a watershed in Rocky Mountain National Park has also reported reductions in stream water NO_3^- concentrations of more than 40% from peak concentrations in the mid-2000s, which corresponded to decreases in NO_X emissions and estimated N deposition (ISA, Appendix 7, section 7.1.5.1).

4.3.1.2.2 Estuarine and Coastal Ecosystems

Nutrient inputs to coastal and estuarine waters are important influences on the health of these waterbodies. As long recognized, "N enrichment of marine and estuarine waters can alter the ratios among nutrients such as P and Si and affect overall nutrient limitation" (ISA, Appendix 10, p. 10-6). Continued inputs of N, the most common limiting nutrient in estuarine and coastal systems, have resulted in N over enrichment and subsequent alterations to the nutrient balance in these systems (ISA, Appendix 10, p. 10-6). For example, the limiting nutrient may change (e.g., from phosphorus to N) as water moves from freshwater through the transition zones into estuaries and marine waters (ISA, Appendix 10, section 10.1.3). Further, "[l]evels of N limitations are also affected by seasonal patterns in estuaries, with N limited conditions likely occurring during the peak of annual productivity in the summer" (ISA, Appendix 10, p. 10-6). Moreover, the rate of N delivery to coastal waters is strongly correlated to changes in primary production and phytoplankton biomass (ISA, Appendix 10, section 10.1.3; Paerl and Piehler, 2008).

In estuarine and near coastal systems, the prevalence and health of submerged aquatic vegetation (SAV) has been identified as a biological indicator for estuarine condition (ISA, Appendix 10, section 10.2.5). Previously available evidence indicated the role of N loading in SAV declines in multiple U.S. estuaries through increased production of macroalgae or other

algae which reduce sunlight penetration into shallow waters where SAVs are found (ISA, Appendix 10, section 10.2.3). Newly available studies have reported findings of increased SAV populations in two tributaries of the Chesapeake Bay corresponding to reduction in total N loading from all sources since 1990 (ISA, Appendix 10, section 10.2.5). The newly available studies also identify other factors threatening SAV, including increasing temperature related to climate change (ISA, Appendix 10, section 10.2.5).

Algal blooms and associated die-offs can contribute to hypoxic conditions (most common during summer months), which can contribute to fish kills and associate reductions in marine populations. In the U.S., the documented incidence of hypoxia increased almost 30-fold from 1960 to 2008, at which time it was reported in more than 300 coastal areas (ISA, Appendix 10, section 10.2.4; Jewett et al., 2010). Areas of eutrophication-related hypoxia are found along the East coast, Gulf of Mexico coast and some areas of the Pacific coast (ISA, Appendix 10, Figure 10-5). In such low oxygen conditions, only tolerant organisms are present (Diaz et al., 2013; Jewett et al., 2010).

Increased N loading to coastal areas (regardless of source) can affect dissolved oxygen levels and lead to shifts in community composition, reduced biodiversity, and increased mortality of biota (ISA, Appendix 10, section 10.3). Studies of these categories of effects describe shifts in diatom communities over times of extremely low oxygen levels (ISA, Appendix10, section 10.3.1), altered phytoplankton community composition with higher N inputs (ISA, Appendix10, section 10.3.2), as well as correlation of waterbody levels of nitrogen compounds with changes to bacteria/archaea diversity (ISA, Appendix10, section 10.3.4), benthic diversity (ISA, Appendix10, section 10.3.5), and fish diversity (ISA, Appendix10, section 10.3.6). Further, the form of available N (e.g., NH_4^+ or NO_3^-) can influence phytoplankton community composition in estuarine and marine environments (ISA, Appendix 10, section 10.3.3). In hypoxic areas, mortality of stationary organisms and avoidance of hypoxic conditions by mobile organisms lead to changes in biodiversity and loss of biomass (ISA, Appendix 10, section 10.3.3; Diaz and Rosenberg, 2008) which can in turn affect energy transfer through the food web. The degree to which these impacts are driven by atmospheric N deposition vary greatly and are largely unique to the specific ecosystem.

Estimates of the relative contribution of atmospheric deposition to total N loading vary among estuaries. Analyses based on data across two to three decades extending from the 1990s through about 2010 estimate that most of the analyzed estuaries receive 15-40% of their N inputs from atmospheric sources (ISA, section ES5.2; ISA, Appendix 7, section 7.2.1) though for specific estuaries contributions can vary more widely. In areas along the West Coast, N sources may include coastal upwelling from oceanic waters, as well as transport from watersheds. Common N inputs to estuaries include those associated with freshwater inflows transporting N

from agriculture, urban, wastewater sources, in addition to atmospheric deposition across the watershed (ISA, IS2.2.2; ISA, Appendix 7, section 7.2.1).

Estimates of N loading to estuaries from atmospheric deposition has been estimated in several recent modeling studies (ISA, Table 7-9). One analysis of estuaries along the Atlantic Coast and the Gulf of Mexico, which estimated that 62−81% of N delivered to the eastern U.S coastal zone is anthropogenic in source, also reported that atmospheric N deposition to freshwater that is subsequently transported to estuaries represents 17−21% of the total N loading into the coastal zone (McCrackin et al., 2013; Moore et al., 2011). In the Gulf of Mexico, 26% of the N transported to the Gulf in the Mississippi/Atchafalaya River basin was estimated to be contributed from atmospheric deposition (which may include volatilized losses from natural, urban, and agricultural sources) (Robertson and Saad, 2013). Another modeling analysis identified atmospheric deposition to watersheds as the dominant source of N to the estuaries of the Connecticut, Kennebec, and Penobscot rivers. For the entire Northeast and mid-Atlantic coastal region, it dropped to third largest source (20%), following agriculture (37%) and sewage and population-related sources (28%) (ISA, Appendix 7, section 7.2.1). Estimates for West Coast estuaries indicate much smaller contribution from atmospheric deposition. For example, analyses for Yaquina Bay, Oregon, estimated direct deposition to contribute only 0.03% of N inputs; estimated N input to the watershed from N fixing red alder (Alnus rubra) trees was a much larger (8%) source (ISA, Appendix 7, section 7.2.1; Brown and Ozretich, 2009).

4.3.1.2.3 Wetlands

With regard to wetland sensitivity to N deposition, in general, those wetlands receiving a larger fraction of their total water budget in the form of precipitation are more sensitive to the effects of N deposition. The relative contribution of atmospheric deposition to total wetland N loading varies with wetland type, with bogs receiving the greatest contribution and accordingly being most vulnerable to nutrient enrichment effects of N deposition (ISA, Appendix 11, section 11.1). For example, bogs (70−100% of hydrological input from rainfall) are more sensitive to N deposition than fens (55−83% as rainfall), which are more sensitive than coastal wetlands (10−20% as rainfall) (ISA, Appendix 11, section 11.10). Nearly all N loading to ombrotrophic bogs[5] comes from atmospheric deposition because precipitation is the only source of water to these wetlands. For freshwater fens, marshes, and swamps, inputs from ground and surface water are often of similar order of magnitude as that from precipitation. Similarly, estuarine and coastal wetlands receive water from multiple sources that include precipitation, ground and/or surface water, and marine and/or estuarine waters (ISA, Appendix 11, section 11.1).

[5] Ombrotrophic bogs develop in areas where drainage is impeded and precipitation exceeds evapotranspiration (ISA, Appendix 11, section 11.1).

4.3.1.3 Key Uncertainties

Models are used extensively to simulate the movement of N to sensitive receptors in aquatic ecosystems, and to estimate indicators of eutrophication risk. In the case of estuarine and near-coastal systems, the models are hydrodynamically complex and due to the need for inputs particular to the waterbody to which they are applied, tend to be site specific (NRC, 2000; ISA, Appendix 7, section 7.2.8.2). Other model uncertainties may arise from the difficulties in disentangling N input sources and apportioning the source of N in the ecosystem correctly. This leads to uncertainty in the role of atmospheric deposition in the N driven effects that are observed.

Several uncertainties contribute to estimates of N deposition associated with certain water body responses. These include a difficulty in estimating dry deposition of gaseous and particulate N to complex surfaces; extremely limited data, particularly for arid, mountainous terrain; and difficulties estimating deposition in areas with high snowfall, cloud water or fog (ISA, Appendix 9, section 9.5; Pardo et al., 2011). For example, "N deposition estimates at high-elevation sites such as those in the Rocky and Sierra Nevada mountains are associated with considerable uncertainty, especially uncertainty for estimates of dry deposition" (ISA, Appendix 9, p. 9-44; Williams et al., 2017b). For estimates of N deposition associated with other sensitive responses, such as shifts in phytoplankton communities in high-elevation lakes, "N deposition model bias may be close to, or exceed, predicted [critical load] values" (ISA, Appendix 9, p. 9-44; Williams et al., 2017b).

4.3.2 Terrestrial Ecosystems

It is long established that N enrichment of terrestrial ecosystems increases plant productivity (ISA, Appendix 6, section 6.1). Building on this, the currently available evidence, including evidence that is longstanding, is sufficient to infer a causal relationship between N deposition and the alteration of the physiology and growth of terrestrial organisms and the productivity of terrestrial ecosystems (ISA, Appendix 5, section 5.2 and Appendix 6, section 6.2). Responsive ecosystems include those that are N limited and/or contain species that have evolved in nutrient-poor environments. In these ecosystems the N-enrichment changes in plant physiology and growth rates vary among species, with species that are adapted to low N supply being readily outcompeted by species that have higher N demand. In this manner, the relative representation of different species may be altered, and some species may be eliminated altogether, such that community composition is changed and species diversity declines (ISA, Appendix 6, sections 6.3.2 and 6.3.8). The currently available evidence in this area is sufficient to infer a causal relationship between N deposition and the alteration of species richness,

community composition, and biodiversity in terrestrial ecosystems (ISA, section IS.5.3 and Appendix 6, section 6.3).

4.3.2.1 Nature of Effects and New Evidence

Previously available evidence described the role of N deposition in changing soil carbon and N pools and fluxes, as well as altering plant and microbial growth and physiology in an array of terrestrial ecosystems. This evidence supported our understanding in the last review of how N deposition influences plant physiology, growth, and terrestrial ecosystem productivity. The newly available evidence confirms these conclusions and improves our understanding of the mechanisms that link N deposition and biogeochemistry in terrestrial ecosystems. The new evidence supports a more detailed understanding of how N influences terrestrial ecosystem growth and productivity; community composition and biodiversity in sensitive ecosystems (ISA, Appendix 6, section 6.2.1).

A supply of N is essential for plant growth and, as was clear in the last review, N availability is broadly limiting for productivity in many terrestrial ecosystems (ISA, Appendix 6, section 6.2.1). Accordingly, N additions contribute to increased productivity and can alter biodiversity. Eutrophication, one of the mechanisms by which this can occur, comprises multiple effects that include changes to the physiology of individual organisms, alteration of the relative growth and abundance of various species, transformation of relationships between species, and indirect effects on availability of essential resources other than N, such as light, water, and nutrients (ISA, Appendix 6, section 6.2.1).

The currently available evidence for the terrestrial ecosystem effects of N enrichment, including eutrophication, includes studies in a wide array of systems, including forests (tropical, temperate, and boreal), grasslands, arid and semi-arid scrublands, and tundra (ISA, Appendix 6). The organisms affected include trees, herbs and shrubs, and lichen, as well as fungal, microbial, and arthropod communities. As recognized in section 4.1 above, lichen communities, which have important roles in hydrologic cycling, nutrient cycling, and as sources of food and habitat for other species, are also affected by atmospheric N (ISA, Appendix 6). The recently available studies on the biological effects of added N in terrestrial ecosystems include investigations of plant and microbial physiology, long-term ecosystem-scale N addition experiments, regional and continental-scale monitoring studies, and syntheses.

The previously available evidence included N addition studies in the U.S. and N deposition gradient studies in Europe that reported associations of N deposition with reduced species richness and altered community composition for grassland plants, forest understory plants, and mycorrhizal fungi (soil fungi that have a symbiotic relationship with plant roots) (ISA, Appendix 6, section 6.3). Since 2008, new research techniques have been developed to

understand community composition, additional communities have been surveyed, and new studies have made it possible to isolate the influence of N deposition from other environmental factors. In addition, new evidence has been developed for forest communities indicating that N deposition alters the physiology and growth of overstory trees, and that N deposition has the potential to change the community composition of forests (ISA, Appendix 6, section 6.6). Recent studies on forest trees include analyses of long-term forest inventory data collected from across the U.S. and Europe (ISA, Appendix 6, section 6.2.3.1). New research also expands the understanding that N deposition can alter the physiology, growth, and community composition of understory plants, lichens, mycorrhizal fungi, soil microorganisms, and arthropods (ISA, Appendix 6, section 6.2.3 and 6.3.3).

The recent evidence includes findings of variation in forest understory and non-forest plant communities with atmospheric N deposition gradients in the U.S. and in Europe. For example, gradient studies in Europe have found higher N deposition to be associated with forest understory plant communities with more nutrient-demanding and shade-tolerant plant species (ISA, Appendix 6, section 6.3.3.2). A recent gradient study in the U.S. found forest understory species richness to be highly dependent on soil pH, with negative associations of species richness with N deposition rates above 11.6 kg N/ha-yr at sites with low soil pH but not at the sites with basic soils (ISA, Appendix 6, section 6.3.3.2).

Among the new studies are investigations of effects of N on mycorrhizal fungi and lichens. Studies indicate that increased N in forest systems can result in changes in mycorrhizal community composition (ISA, Appendix 6, section 6.2). Forest microbial biomass and community composition can also be affected, which can contribute to impacts on arthropod communities (ISA, Appendix 6, section 6.3.3.4). Recent evidence includes associations of variation in lichen community composition with N deposition gradients in the U.S. and Europe, (ISA, Appendix 6, section 6.2.6; Table 6-23). Differences in lichen community composition have been attributed to atmospheric N pollution in forests throughout the West Coast, in the Rocky Mountains, and in southeastern Alaska. Differences in epiphytic lichen growth or physiology have been observed along atmospheric N deposition gradients in the highly impacted area of southern California, and also in more remote locations such as Wyoming and southeastern Alaska (ISA, Appendix 6, section 6.3.7). Historical deposition may play a role in observational studies of N deposition effects, complicating the disentangling of responses that may be related to more recent N loading.

Newly available findings from N addition experiments expand on the understanding of mechanisms linking changes in plant and microbial community composition to increased N availability. Such experiments in arid and semi-arid environments indicate that competition for resources such as water may exacerbate the effects of N addition on diversity (ISA, Appendix 6,

section 6.2.6). A 25-year experiment with N additions ranging from 10 to 95 kg N/ha-yr (and background wet deposition of N estimated at 6 kg N/ha-yr) observed grassland composition to change from a high-diversity, native-dominated state to a low-diversity, non-native dominated state (ISA, Appendix 6, section 6.3.5). The newly available evidence also includes studies in arid and semiarid ecosystems, particularly in southern California, that have reported changes in plant community composition, in the context of a long history of significant N deposition, with fewer observations of plant species loss or changes in plant diversity (ISA, Appendix 6, section 6.3.6).

4.3.2.2 Terrestrial Ecosystem Sensitivity

In general, most terrestrial ecosystems are N limited and, consequently, sensitive to effects related to N enrichment (ISA, Appendix 6, section 6.3.8). Factors identified as governing the sensitivity of terrestrial ecosystems to nutrient enrichment from N deposition include "the rates of N deposition, degree of N limitation, ecosystem productivity, elevation, species composition, length of growing season, and soil N retention capacity" (ISA, Appendix 6, p. 6-162). One example is that of alpine tundra ecosystems, which: (1) are typically strongly N limited, and contain vegetation adapted to low N availability; (2) often have thin soils with limited N retention capacity; and (3) have short growing seasons (ISA, Appendix 6, section 6.3.8). Given the evidence regarding sensitivity of lichens and ectomycorrhizal fungi to N enrichment effects, it may be that ecosystems containing a large number and/or diversity of these organisms, such as temperate and boreal forests and alpine tundra, could be considered particularly sensitive to N deposition (ISA, Appendix 6, sections 6.2.3.2, 6.2.3.3, 6.2.4, and 6.3.8).

In the currently available evidence, studies conducted in grassland and coastal sage shrub communities, and in arid ecosystems, such as the Mojave Desert, indicate sensitivity of those communities. For example, N addition studies in Joshua Tree National Park have reported losses in forb species richness (which make up most of the grassland biodiversity), greater growth of grass species (which make up the majority of grassland biomass), and changes in reproductive rates. Accordingly, the N limitation in grasslands and the dominance by fast-growing species that can shift in abundance rapidly (in contrast to forest trees) contribute to an increased sensitivity of grassland ecosystems to N inputs (ISA, Appendix 6, section 6.3.6). Studies in southern California coastal sage scrub communities, including studies of the long-term history of N deposition, which was appreciably greater in the past than recent rates, indicate impacts on community composition and species richness in these ecosystems (ISA, Appendix 6, sections 6.2.6 and 6.3.6). In summary, the ability of atmospheric N deposition to override the natural spatial heterogeneity in N availability in arid ecosystems, such as the Mojave Desert and

California coastal sage scrub ecosystems in southern California, makes these ecosystems sensitive to N deposition (ISA, Appendix 6, section 6.3.8).

The current evidence includes relatively few studies of N enrichment recovery in terrestrial ecosystems. Among N addition studies assessing responses after cessation of additions, it has been observed that soil nitrate and ammonium concentrations recovered to levels observed in untreated controls within 1 to 3 years of the cessation of additions, but soil processes such as N mineralization and litter decomposition were slower to recover (ISA, Appendix 6, section 6.3.2; Stevens, 2016). A range of recovery times have been reported for mycorrhizal community composition and abundance from a few years in some systems to as long as 28 or 48 years in others (ISA, Appendix 6, section 6.3.2; Stevens, 2016; Emmett et al., 1998; Strengbom et al., 2001). An N addition study in the midwestern U.S. observed that plant physiological processes recovered in less than 2 years, although grassland communities were slower to recover and still differed from controls 20 years after the cessation of N additions (ISA, Appendix 6, section 6.3.2; Isbell et al., 2013).

4.3.2.3 Key Uncertainties

Just as there are uncertainties associated with estimating N deposition associated with ecological responses in aquatic systems (as summarized in section 4.3.1.3 above), such uncertainties exist with terrestrial ecosystem analyses. For example, regarding wet deposition measurements, there are uncertainties associated with monitoring instrumentation and measurement protocols, as well as limitations in the spatial extent of existing monitoring networks, especially in remote areas. Given limitations in our ability to estimate dry deposition, estimates are often based on model predictions, for which there are various sources of uncertainty, including model formulation and inputs for the simulation of chemistry and transport processes. Other uncertainties are associated with an incomplete understanding of the underlying scientific processes influencing atmospheric deposition that are not possible to quantify. For example, uncertainties associated with deposition estimates (that may be utilized in observational studies) include those associated with simulating effects of the tree canopy on N oxides (including both bidirectional gas exchange and canopy reactions), bidirectional exchange of NH_3 with biota and soils, and processes determining transference ratios that relate average concentration to deposition. (ISA, section IS.14.1.3).

There is also uncertainty with regard to the relative importance of different N species in effects of N enrichment on terrestrial ecosystem [ISA, Appendix 6, section 6.3.2]. Although there are few direct analyses comparing the impacts of oxidized and reduced forms of N deposition on biodiversity, it is plausible that NO_3^- may be less likely to accumulate in soil, with associated effects, due to its greater tendency to be more readily lost to both leaching and

denitrification than NH_4^+ (ISA, Appendix 6, section 6.3.2). Further, while multiple meta-analyses have generally not reported differences in the relationship of different N forms with ecological and biogeochemical endpoints, such as plant productivity or microbial biomass, several individual studies have observed differential effects on diversity of NH_4^+ versus NO_3^- additions. For example, an experiment involving a nutrient-poor, Mediterranean site found that while an NH_4^+ addition (40 kg N/ha-yr) increased plant richness, addition of the same amount of N comprised of half NH_4^+ and half NO_3^- did not (ISA, Appendix 6, section 6.3.2).

With regard to ecological responses and impacts of concern, there are several key areas of uncertainty. In observational studies, in addition to uncertainty regarding the role of historical deposition, other confounding factors such as drought and ozone may also contribute to impacts of concern. Further, there is wide variability in the response of plants to nitrogen inputs and the impacts of spatially variable factors such as climate, geology and past deposition on that response is generally unknown. Spatially, variation in biological and biogeochemical processes imposed by climate, geology, biota, and other environmental factors may affect observed associations of ecological metrics with deposition metrics.

Uncertainties also relate to time scales and lags. For example, while atmospheric deposition responds dynamically to shifts in emissions and weather patterns, ecological processes react to environmental stress at a variety of timescales, which due to intervening ecosystem processes usually lag changes in deposition. There are also uncertainties related to the role of historic patterns of deposition in ecosystem effects initially attributed to recent gradients in deposition. These may loom larger for geographic regions, such as the northeastern U.S. or southern California that have long and geographically extensive histories of elevated N deposition.

4.4 OTHER DEPOSITION-RELATED EFFECTS

Additional categories of effects for which the current evidence is sufficient to infer causal relationships include changes in mercury methylation processes in freshwater ecosystems, changes in aquatic biota due to sulfide phytotoxicity, and ecological effects from PM deposition (ISA, Table IS-1).

4.4.1 Mercury Methylation

The current evidence, including that newly available in this review, is sufficient to infer a causal relationship between S deposition and the alteration of mercury methylation in surface water, sediment, and soils in wetland and freshwater ecosystems. The process of mercury methylation is influenced in part by surface water SO_4^{2-} concentrations, as well as the presence of mercury. Accordingly, in waterbodies where mercury is present, S deposition, particularly that

associated with SO_X has a role in production of methylmercury, which contributes to methylmercury accumulation in fish (ISA, Appendix 12, section 12.8).

Newly available evidence has improved our scientific understanding of the types of organisms involved in the methylation process, as well as the environments in which they are found. Studies have also identified additional areas within the U.S. containing habitats with conditions suitable for methylation, and species that accumulate methylmercury (ISA, Appendix 12, section 12.3). The evidence also contributes to our understanding of factors that can influence the relationship between atmospheric S deposition and methylmercury in aquatic systems; such factors include oxygen content, temperature, pH, and carbon supply, which themselves vary temporally, seasonally, and geographically (ISA, Appendix 12, section 12.3).

4.4.2 Sulfide Toxicity

The evidence newly available in this review regarding non-acidifying sulfur effects on biota expands upon that available for the 2008 ISA. The currently available evidence is sufficient to infer a new causal relationship between S deposition and changes in biota due to sulfide phytotoxicity, including alteration of growth and productivity, species physiology, species richness, community composition, and biodiversity in wetland and freshwater ecosystems (ISA, section IS.9). The currently available evidence indicates that the presence of sulfide, produced through microbial transformation, interferes with nutrient uptake in roots of plants in wetlands and other fresh waterbodies. Studies also report that elevated sulfide can result in decreased seed mass, seed viability, seedling emergence rates, decreased seedling height, decreased seedling survival rates, and reductions in total plant cover, all which have the potential to contribute to shifts in plant community composition (ISA, Appendix 12, section 12.2.3). Sulfur deposition can contribute to sulfide and associated phytotoxicity in freshwater wetlands and lakes. Recently available studies indicate that sulfide toxicity can occur in wetland habitats and suggests that sulfide toxicity can determine plant community composition in freshwater wetlands. These studies indicate sulfide toxicity to have occurred in multiple wetland ecosystems in North America (ISA, Appendix 12, sections12.2.3 and 12.7.3).

4.4.3 Ecological Effects of PM Other Than N and S Deposition

Particulate matter includes a heterogeneous mixture of particles differing in origin, size, and chemical composition. In addition to N and S and their transformation products, other PM components, such as trace metals and organic compounds are also deposited to ecosystems and may affect biota. Material deposited onto leaf surfaces can alter leaf processes and PM components deposited to soils and waterbodies may be taken up into biota, with the potential for effects on biological and ecosystem processes. The currently available evidence is sufficient to

infer a likely causal relationship between deposition of PM and a variety of effects on individual organisms and ecosystems (ISA, Appendix 15, section 15.1).

The effects of PM on ecological receptors can be both chemical and physical, and particles that elicit effects on ecological receptors vary by size, origin, and chemical composition. Studies involving ambient air PM have generally involved conditions that would not be expected to meet the current secondary standards for PM, e.g., polluted locations in India or Argentina (ISA, Appendix 15, sections 15.4.3 and 15.4.4). Similarly, reduced photosynthesis has been reported for rice plants experiencing fly ash particle deposition of 0.5 to 1.5 grams per square meter per day (g/m^2-day), a loading which corresponds to greater than 1000 kg/ha-yr (ISA, Appendix 15, sections 15.4.3 and 15.4.6). Further, studies of the direct effects of PM in ambient air on plant reproduction in near roadway locations in the U.S. have not reported a relationship between PM concentrations and pollen germination (ISA, Appendix 15, section 15.4.6). Rather, the evidence related to PM is that associated with deposition of its components, as summarized in section 4.4.3 below.

Although in some limited cases, effects have been attributed to particle size (e.g., soiling of leaves by large coarse particles near industrial facilities or unpaved roads), ecological effects of PM have been largely attributed more to particle composition (Grantz et al., 2003; ISA, Appendix 15, section 15.2). For example, exposure to a given mass-per-volume or -mass concentration may result in quite different ecological effects depending on the PM components. Depending on concentration, trace metals, some of which are biologically essential, can be toxic in large amounts (ISA, Appendix 15, section 15.3.1). Depending on conditions, deposited PM has been associated with effects on vegetation including effects on plant surfaces, foliar uptake processes, gas exchange, physiology, growth, and reproduction. The evidence largely comes from studies involving elevated concentrations such as near industrial areas or historically polluted cities (ISA, Appendix 15, section 15.4). Recent assays have supported previously available evidence that toxicity relates more to chemical components than total mass. Additionally recent experiments have suggested that PM deposition can influence responses in microbial communities (ISA, Appendix 15, section 15.8). Quantifying relationships between ambient air concentrations of PM and ecosystem response are difficult and uncertain.

4.5 PUBLIC WELFARE IMPLICATIONS

The public welfare implications of the evidence regarding S and N related welfare effects are dependent on the type and severity of the effects, as well as the extent of the effect at a particular biological or ecological level of organization or spatial scale. We discuss such factors here in light of judgments and conclusions made in NAAQS reviews regarding effects on the public welfare.

As provided in section 109(b)(2) of the CAA, the secondary standard is to "specify a level of air quality the attainment and maintenance of which in the judgment of the Administrator … is requisite to protect the public welfare from any known or anticipated adverse effects associated with the presence of such air pollutant in the ambient air." The secondary standard is not meant to protect against all known or anticipated welfare effects related to oxides of N and S, and particulate matter, but rather those that are judged to be adverse to the public welfare, and a bright-line determination of adversity is not required in judging what is requisite (78 FR 3212, January 15, 2013; 80 FR 65376, October 26, 2015; see also 73 FR 16496, March 27, 2008). Thus, the level of protection from known or anticipated adverse effects to public welfare that is requisite for the secondary standard is a public welfare policy judgment made by the Administrator. The Administrator's judgment regarding the available information and adequacy of protection provided by an existing standard is generally informed by considerations in prior reviews and associated conclusions.

- **What does the available information indicate regarding the public welfare implications of S and N deposition-related welfare effects?**

The categories of effects identified in the CAA to be included among welfare effects are quite diverse,[6] and among these categories, any single category includes many different types of effects that are of broadly varying specificity and level of resolution. For example, effects on vegetation and effects on animals are categories identified in CAA section 302(h), and the ISA recognizes effects of N and S deposition at the organism, population, community, and ecosystem level, as summarized in sections 4.1 and 4.2 above (ISA, sections IS.5 to IS.9). As noted in the last review of the secondary NAAQS for N oxides and SO_X, while the CAA section 302(h) lists a number of welfare effects, "these effects do not define public welfare in and of themselves" (77 FR 20232, April 3, 2012).

The significance of each type of effect with regard to potential effects on the public welfare depends on the type and severity of effects, as well as the extent of such effects on the affected environmental entity, and on the societal use of the affected entity and the entity's significance to the public welfare. Such factors have been considered in the context of judgments and conclusions made in some prior reviews regarding public welfare effects. For example, in the context of secondary NAAQS decisions for ozone, judgments regarding public welfare significance have given particular attention to effects in areas with special federal protections

[6] Section 302(h) of the CAA states that language referring to "effects on welfare" in the CAA "includes, but is not limited to, effects on soils, water, crops, vegetation, manmade materials, animals, wildlife, weather, visibility, and climate, damage to and deterioration of property, and hazards to transportation, as well as effects on economic values and on personal comfort and well-being" (CAA section 302(h)).

(such as Class I areas),[7] and lands set aside by states, tribes and public interest groups to provide similar benefits to the public welfare (73 FR 16496, March 27, 2008; 80 FR 65292, October 26, 2015).[8] In the 2015 O_3 NAAQS review, the EPA recognized the "clear public interest in and value of maintaining these areas in a condition that does not impair their intended use and the fact that many of these lands contain O_3-sensitive species" (73 FR 16496, March 27, 2008).

Judgments regarding effects on the public welfare can depend on the intended use for, or service (and value) of, the affected vegetation, ecological receptors, ecosystems and resources and the significance of that use to the public welfare (73 FR 16496, March 27, 2008: 80 FR 65377, October 26, 2015). Uses or services provided by areas that have been afforded special protection can flow in part or entirely from the vegetation that grows there or other natural resources. Ecosystem services range from those directly related to the natural functioning of the ecosystem to ecosystem uses for human recreation or profit, such as through the production of lumber or fuel (Costanza et al., 2017; ISA, section IS.5.1). The spatial, temporal, and social dimensions of public welfare impacts are also influenced by the type of service affected. For example, a national park can provide direct recreational services to the thousands of visitors that come each year, but also provide an indirect value to the millions who may not visit but receive satisfaction from knowing it exists and is preserved for the future (80 FR 65377, October 26, 2015).

In the last review of the secondary NAAQS for N oxides and SO_X, ecosystem services were discussed as a method of assessing the magnitude and significance to the public of resources affected by ambient air concentrations of oxides of nitrogen and sulfur and associated deposition in sensitive ecosystems (77 FR 20232, April 3, 2012). That review recognized that although there is no specific definition of adversity to public welfare, one paradigm might involve ascribing public welfare significance to disruptions in ecosystem structure and function. The concept of considering the extent to which a pollutant effect will contribute to such

[7] Areas designated as Class I include all international parks, national wilderness areas which exceed 5,000 acres in size, national memorial parks which exceed 5,000 acres in size, and national parks which exceed 6,000 acres in size, provided the park or wilderness area was in existence on August 7, 1977. Other areas may also be Class I if designated as Class I consistent with the CAA.

[8] For example, the fundamental purpose of parks in the National Park System "is to conserve the scenery, natural and historic objects, and wild life in the System units and to provide for the enjoyment of the scenery, natural and historic objects, and wild life in such manner and by such means as will leave them unimpaired for the enjoyment of future generations" (54 U.S.C. 100101). Additionally, the Wilderness Act of 1964 defines designated "wilderness areas" in part as areas "protected and managed so as to preserve [their] natural conditions" and requires that these areas "shall be administered for the use and enjoyment of the American people in such manner as will leave them unimpaired for future use and enjoyment as wilderness, and so as to provide for the protection of these areas, [and] the preservation of their wilderness character …" (16 U.S.C. 1131 (a) and (c)). Other lands that benefit the public welfare include national forests which are managed for multiple uses including sustained yield management in accordance with land management plans (see 16 U.S.C. 1600(1)-(3); 16 U.S.C. 1601(d)(1)).

disruptions has been used broadly by the EPA in considering effects. An evaluation of adversity to public welfare might also consider the likelihood, type, magnitude, and spatial scale of the effect, as well as the potential for recovery and any uncertainties relating to these considerations (77 FR 20218, April 3, 2012).

The types of effects on aquatic and terrestrial ecosystems discussed in sections 4.1 through 4.4 above differ with regard to aspects important to judging their public welfare significance. For example, in the case of effects on timber harvest, such judgments may consider aspects such as the heavy management of silviculture in the U.S., while judgments for other categories of effects may generally relate to considerations regarding natural areas, including specifically those areas that are not managed for harvest. For example, effects on tree growth and survival have the potential to be significant to the public welfare through impacts in Class I and other areas given special protection in their natural/existing state, although they differ in how they might be significant.

In this context, it may be important to consider that S and N deposition-related effects, such as changes in growth and survival of plant and animal species, could, depending on severity, extent, and other factors, lead to effects on a larger scale including changes in overall productivity and altered community composition (ISA, section IS.2.2.1 and Appendices 5, 6, 8, 9, and 10). Further, effects on individual species could contribute to impacts on community composition through effects on growth and reproductive success of sensitive species in the community, with varying impacts to the system through many factors including changes to competitive interactions (ISA, section IS.5.2 and Appendix 6, section 6.3.2).

With respect to aquatic acidification effects, because acidification primarily affects the diversity and abundance of aquatic biota, it also affects the ecosystem services that are derived from the fish and other aquatic life found in these surface waters (2011 PA, section 4.4.5). Fresh surface waters support several cultural services, such as aesthetic and educational services; the type of service that is likely to be most widely and significantly affected by aquatic acidification is recreational fishing. Multiple studies have documented the economic benefits of recreational fishing. While the freshwater rivers and lakes of the northeastern United States, surface waters that have been most affected by acidification, are not a major source of commercially raised or caught fish, they are a source of food for some recreational and subsistence fishers and for other consumers (2009 REA, section 4.2.1.3). It is not known, however, if and how consumption patterns of these fishers may have been affected by the historical impacts of surface water acidification in the affected systems. Non-use services, which include existence (protection and preservation with no expectation of direct use) and bequest values, are arguably a significant source of benefits from reduced acidification (Banzhaf et al., 2006).

Nitrogen loading in aquatic ecosystems, particularly large estuarine and coastal water bodies, has and continues to pose risks to the services provided by those ecosystems, with clear implications to the public welfare (2011 PA, section 4.4.2; ISA, Appendix 14, section 14.3.2). For example, the large estuaries of the eastern U.S. are an important source of fish and shellfish production, capable of supporting large stocks of resident commercial species and serving as breeding grounds and interim habitat for several migratory species (2009 REA, section 5.2.1.3). These estuaries also provide an important and substantial variety of cultural ecosystem services, including water-based recreational and aesthetic services. And as noted for fresh waters above, these systems have non-use benefits to the public (2011 PA, section 4.4.5). As discussed in section 4.3.1.2.2 above, the relative contribution of atmospheric deposition to total N loading varies widely among estuaries and has declined in more recent years.

A complication to consideration of public welfare implications that is specific to N deposition in terrestrial systems is its potential to increase growth and yield of agricultural and forest crops, which may be judged and valued differentially than changes in growth of some species in natural ecosystems. As discussed further in section 4.3.2 above, N enrichment in natural ecosystems can, by increasing growth of N limited plant species, change competitive advantages of species in a community, with associated impacts on the composition of the ecosystem's plant community. The public welfare implications of such effects may vary depending on their severity, prevalence or magnitude, such as with only those rising to a particular severity (e.g., with associated significant impact on key ecosystem functions or other services), magnitude or prevalence considered of public welfare significance. Impacts on some of these characteristics (e.g., forest or forest community composition) may be considered of greater public welfare significance when occurring in Class I or other protected areas, due to the value that the public places on such areas.[9] Other ecosystem services that can be affected are summarized below in Figure 4-3[10] (ISA, Appendix 14). In considering such services in past reviews for secondary standards for other pollutants (e.g., O_3), the Agency has given particular attention to effects in natural ecosystems, indicating that a protective standard, based on consideration of effects in natural ecosystems in areas afforded special protection, would also "provide a level of protection for other vegetation that is used by the public and potentially affected by O_3 including timber, produce grown for consumption and horticultural plants used for landscaping" (80 FR 65403, October 26, 2015).

[9] Locations of the Class I areas identified under the Clean Air Act, section 162(1) are shown in Figure 4-4 (https://www.epa.gov/visibility/regional-haze-program).

[10] The articulation of welfare effects in Figure 4-3 is intended to reflect the ISA causal determinations in an easier to comprehend manner that also illustrates connections among effects.

Although more sensitive effects are described with increasingly greater frequency in the evidence base of effects related to ecosystem deposition of N and S compounds, the available information does not yet provide a framework that can specifically tie various magnitudes or prevalences of changes in a biological or ecological indicator (e.g., lichen abundance or community composition) to broader effects on the public welfare. This gap creates uncertainties when considering the public welfare implications of some biological or geochemical responses to ecosystem acidification or N enrichment, and accordingly judgments on the potential for public welfare significance. That notwithstanding, while shifts in species abundance or composition of various ecological communities may not be easily judged with regard to public welfare significance, at some level, such changes, especially if occurring broadly in specially protected areas, where the public can be expected to place high value, might reasonably be concluded to impact the public welfare. An additional complexity in the current review is the current much-improved air quality and associated reduced deposition within the context of a longer history that included appreciably greater deposition in the middle of the last century, the environmental impacts of which may remain.

In summary, several considerations are recognized as important to judgments on the public welfare significance of the array of welfare effects at different exposure conditions. These include uncertainties and limitations that must be taken into account regarding the magnitude of key effects that might be concluded to be adverse to ecosystem health and associated services. Additionally, there are numerous locations vulnerable to public welfare impacts from S or N deposition-related effects on terrestrial and aquatic ecosystems and their associated services. Other important considerations include the exposure circumstances that may elicit effects and the potential for the significance of the effects to vary in specific situations due to differences in sensitivity of the exposed species, the severity and associated significance of the observed or predicted effect, the role that the species plays in the ecosystem, the intended use of the affected species and its associated ecosystem and services, the presence of other co-occurring predisposing or mitigating factors, and associated uncertainties and limitations.

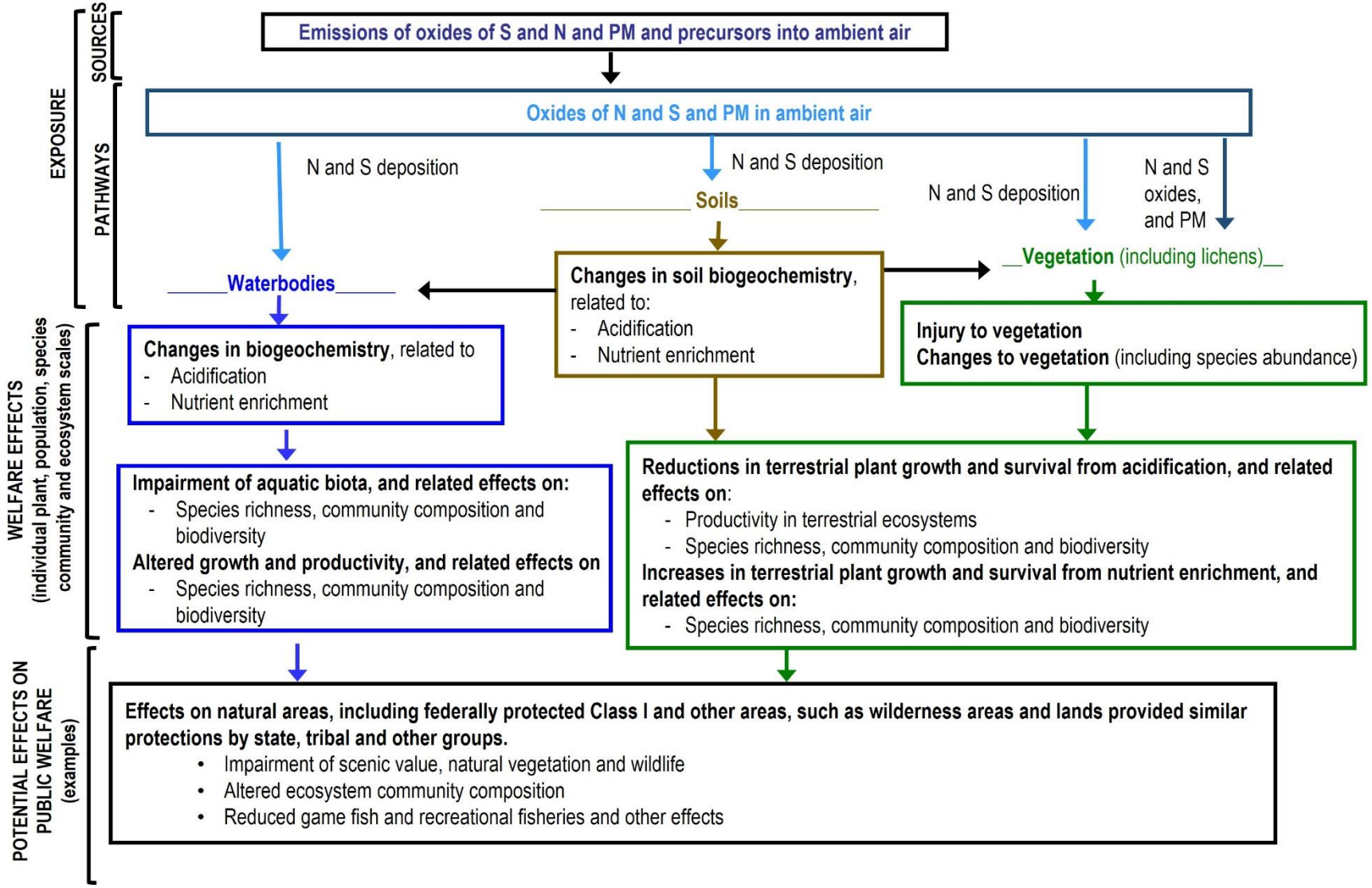

Figure 4-3. Potential effects on the public welfare of ecological effects of N Oxides, SOx, and PM.

Figure 4-4. Locations of areas designated Class I under section 162(a) of the Clean Air Act.

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5 EXPOSURE CONDITIONS ASSOCIATED WITH EFFECTS

In this review, we consider two categories of exposure conditions associated with welfare effects. The first is the less complex consideration of the direct exposures to pollutants in ambient air, which were the focus in the establishment of the standards. The second is the more complex consideration of exposures related to atmospheric deposition associated with the pollutants in ambient air. In our consideration in this chapter of exposure conditions associated with effects, we have generally addressed the two categories in separate sections beginning with the second category. This is done in the context of the following overarching question:

- **To what extent does the available evidence include quantitative exposure and response information that can inform judgments on air exposures and deposition levels of concern and accordingly, the likelihood of occurrence of such effects in response to air quality that meets the current standards?**

With regard to the more complex consideration of deposition-related effects such as ecosystem acidification and N enrichment, there is wide variation in the extent and level of detail of the evidence available to describe the ecosystem characteristics (e.g., physical, chemical, and geological characteristics, as well as atmospheric deposition history) that influence the degree to which deposition of N and S associated with the oxides of S and N and PM in ambient air elicit ecological effects. One reason for this relates to the contribution of many decades of uncontrolled atmospheric deposition before the establishment of NAAQS for PM, oxides of S and oxides of N, followed by the subsequent decades of continued deposition as standards were implemented and updated. The impacts of this deposition history remain in soils of many parts of the U.S. today (e.g., in the Northeast and portions of the Appalachian Mountains in both hardwood and coniferous forests, as well as areas in and near the Los Angeles Basin), with recent signs of recovery in some areas (ISA, Appendix 4, section 4.6.1; 2008 ISA, section 3.2.1.1). This backdrop and associated site-specific characteristics are among the challenges we consider in our task of identifying deposition targets to provide protection going forward against the array of effects for which we have evidence of occurrence in sensitive ecosystems as a result of the deposition of the past.

With regard to aquatic systems, prior to the peak of S deposition levels that occurred in the 1970s and early 1980s, surface water sulfate concentrations increased in response to S deposition. Subsequently, and especially more recently, surface water sulfate concentrations have generally decreased, particularly in the Northeast. Some waterbodies, however, continue to exhibit little reduction in acidic ions, such as in the Blue Ridge Mountains region in Virginia,

where surface water SO_4^{2-} has remained relatively stable even as emissions declined. This is an example of the competing role of changes in S adsorption on soils and the release of historically deposited S from soils into surface water, which some modeling has suggested will delay chemical recovery in those water bodies (ISA, Appendix 7, section 7.1.2.2).

In this chapter, we first consider aquatic acidification, a category of effects for which quantitative assessment approaches for atmospheric deposition are well established. In the 2012 review of the oxides of N and S, quantitative analyses relating deposition in recent times (e.g., since 2000) to ecosystem acidification, and particularly aquatic acidification, were generally considered to be less uncertain and the ability of those analyses to inform NAAQS policy judgments more robust than analyses related to deposition and ecosystem nutrient enrichment, or eutrophication (2011 PA). While quantitative assessment approaches for aquatic eutrophication as a result of total N loading are also well established, and the evidence base regarding atmospheric deposition and nutrient enrichment has expanded since the 2012 review (as summarized in section 4.3 above), the significance of non-air N loading to rivers, estuaries and coastal waters continues to complicate the assessment of nutrient enrichment-related risks specifically related to atmospheric N deposition. Accordingly, the quantitative REA developed in this review focused on aquatic acidification. This chapter, however, addresses both the quantitative information available for aquatic acidification (section 5.1 summarizes the REA that is described in Appendix 5A in detail) and aquatic nutrient enrichment (section 5.2), as well as terrestrial and other effects of S and N deposition.

Critical loads are frequently used in studies that investigate associations between various chemical, biological, ecological and ecosystem characteristics and a variety of N or S deposition-related metrics.[1] These studies vary widely with regard to the specific ecosystem characteristics being evaluated, as well as the benchmarks selected for judging them, such as the deposition-related metrics, their scope, method of estimation and time period. The specific details of these various factors influence the strengths and limitations for different uses and have associated uncertainties. Given the role of the PA both in focusing on the most policy-relevant aspects of the currently available information (reviewed in the 2020 and 2008 ISAs and past AQCDs) and in clearly describing key aspects, including limitations and associated uncertainties, this

[1] The term, critical load, which in general terms refers to an amount (or a rate of addition) of a pollutant to an ecosystem that is estimated to be at, or just below, that which would have an effect of interest, has multiple interpretations or applications (ISA, p. IS-14). This multiplicity or variety in meanings stems, at least in part, from differing judgments and associated identifications regarding the effect of interest, and judgment of its harm. There is additionally the complication of the dynamic nature of ecosystem pollutant processing and the broad array of factors that influence it. As a result, time is an important dimension, sometimes unstated, as in empirical or observational analyses, sometimes explicit, as in steady-state or dynamic modeling analyses (ISA, section IS.2.2.4).

document is intended to reach beyond individual critical loads developed over a variety of studies and ecosystems and consider the underlying study findings with regard to key aspects of the environmental conditions and ecological characteristics studied. A more quantitative variation of this is the methodology developed for the analyses of aquatic systems and acidification, summarized in section 5.1.2 below. In these analyses, the concept of a critical load is employed with steady-state modeling that relates deposition to waterbody acid neutralizing capacity. This specific use of critical loads in the REA analyses in this review is explicitly described in section 5.1.2.

While recognizing the inherent connections between watersheds and waterbodies, such as lakes and streams, the organization of this chapter recognizes the more established state of the information, tools and data for aquatic ecosystems for characterizing relationships between atmospheric deposition and acidification and/or nutrient enrichment effects under air quality associated with the current standards. Further, we recognize the relatively greater role of atmospheric deposition in aquatic acidification compared to aquatic eutrophication, to which surface water discharges in populated watersheds have long contributed. Therefore, with regard to deposition-related effects, we focus first on the quantitative information for aquatic ecosystem effects in sections 5.1 and 5.2. Section 5.3 discusses the available evidence regarding relationships between deposition-related exposures and the occurrence and severity of effects on trees and understory communities in terrestrial ecosystems. Section 5.4 discusses the currently available information related to consideration of exposure concentrations associated with other welfare effects of nitrogen and sulfur oxides and PM in ambient air.

5.1 AQUATIC ECOSYSTEM ACIDIFICATION

Changes in biogeochemical processes and water chemistry caused by deposition of nitrogen and sulfur compounds to surface waters and their watersheds have been well characterized for decades and have ramifications for biological functioning of freshwater ecosystems, as summarized in section 4.2.1 above (ISA, Appendix 8, section 8.1). These deposited acidic compounds infiltrate both terrestrial and aquatic systems and may contribute to changes to soils and water that are harmful to biota (ISA, section IS.5.3). These changes are dependent on a number of factors that influence the sensitivity of a system to acidification including weathering rates, bedrock composition, topography, vegetation and microbial processes, physical and chemical characteristics of soils and hydrology (ISA, Appendix 4, section 4.3).

The quantitative assessment of aquatic acidification risk performed for this review (described in detail in Appendix 5A) is based on established modeling approaches, extensive databases of site-specific water quality measurements and a commonly recognized indicator of

acidification risk, ANC. The extensive evidence, history of quantitative modeling and site-specific model evaluation supports this assessment. The ability to characterize the role of atmospheric deposition of the pollutants under review is also a factor in the decision to focus quantitative analysis on acid deposition into aquatic ecosystems.

Key aspects of this REA and its results are summarized in the following subsections, with details provided in Appendix 5A. Section 5.1.1 provides background information on the evidence supporting the use of ANC as an indicator of acidification risk in the assessment. The conceptual model and the analysis approach are summarized in section 5.1.2. Results for analyses at three scales are presented in section 5.1.3, and a characterization of the analysis uncertainty is summarized in section 5.1.4. Overall findings are summarized in section 5.1.5.

5.1.1 Role of ANC as Acidification Indicator

Several measures of surface water chemistry are commonly used in assessments of aquatic acidification. These include surface water base cations, pH, inorganic Al and ANC (ISA, Table IS-3). Accordingly, risk to aquatic systems from acidifying deposition can be assessed as a change in specific water quality metrics as a result of nitrogen and/or sulfur deposition. Changes in surface water chemistry reflect the influence of acidic inputs from precipitation, gases, and particles, as well as local geology and soil conditions. As described in section 4.2.1 above, surface water chemical factors such as pH, Ca^{2+}, ANC, ionic metals concentrations, and dissolved organic carbon (DOC) are affected by acid deposition and, accordingly, are commonly used indicators of acidification. Although ANC does not directly cause effects on biota, it relates to pH and aluminum levels, and biological effects are primarily attributable to low pH and high inorganic aluminum concentration (ISA, section ES.5.1). The most widely used measure of surface water acidification, and subsequent recovery under scenarios with lower acidifying deposition, is ANC (ISA, Appendix 7, section 7.1.2.6). This is for several reasons: (1) ANC is associated with the surface water constituents that directly cause or reduce acidity-related stress, in particular pH, Ca^{2+}, and inorganic Al concentrations; (2) ANC is generally a more stable (less variable) measurement than pH; and (3) ANC reflects sensitivity and effects of acidification in a linear fashion across the full range of ANC values (ISA, Appendix 7, section 7.1.2.6).

As summarized in section 4.2.1.2 above, the evidence of effects on biota from aquatic acidification indicates a range of severity with varying pH and ANC levels. The evidence relates to biota ranging from phytoplankton and invertebrates to fish communities. For example, a review by Lacoul et al. (2011) of aquatic acidification effects on aquatic organisms in Atlantic Canada observed that the greatest differences in phytoplankton species richness occurred across a pH range of 4.7 to 5.5 (ANC range of 0 to 20 μeq/L), just below the range (pH 5.5 to 6.5) where bicarbonate becomes rapidly depleted in the water (ISA, Appendix 8, section 8.3.1.1). Under

acidifying conditions, these phytoplankton communities shifted from dominance by chrysophytes, other flagellates, and diatoms to dominance by larger dinoflagellates. In benthic invertebrates residing in sediments of acidic streams, Al concentration is a key influence on the presence of sensitive species. Studies of macroinvertebrate species have reported reduced species richness at lower pH, with the most sensitive group, mayflies, absent at the lowest levels. Values of pH below 5 (which may correspond to ANC levels below 0 μeq/L)[2] were associated with the virtual elimination of all acid-sensitive mayfly and stonefly species over the period from 1937-42 to 1984-85 in two streams in Ontario (Baker and Christensen, 1991). In a more recent study, Baldigo et al. (2009) showed species richness of macroinvertebrate assemblages in the southwestern Adirondack Mountains were severely impacted at median stream pH values below 5.1, moderately impacted at pH values from 5.1 to 5.7, slightly impacted at pH from 5.7 to 6.4 and usually unaffected above pH 6.4 (Figure 5-1). In Atlantic Canada, Lacoul et al. (2011) found the median pH for sensitive invertebrate species occurrence was between 5.2 and 6.1 (ANC of 10 and 80 μeq/L), below which such species tended to be absent. For example, some benthic macroinvertebrates, including several species of mayfly and some gastropods, are intolerant of acid conditions and only occur at pH ≥5.5 (ANC 20 μeq/L) and ≥6, (ANC 50 μeq/L) respectively (ISA, Appendix 8, section 8.3.3).

[2] The citing of ANC values from studies that reported only pH, depended on relating pH and ANC to one another using a generalized relationship based on the assumption of equilibrium with atmospheric CO_2 concentration (Cole and Prairie, 2010).

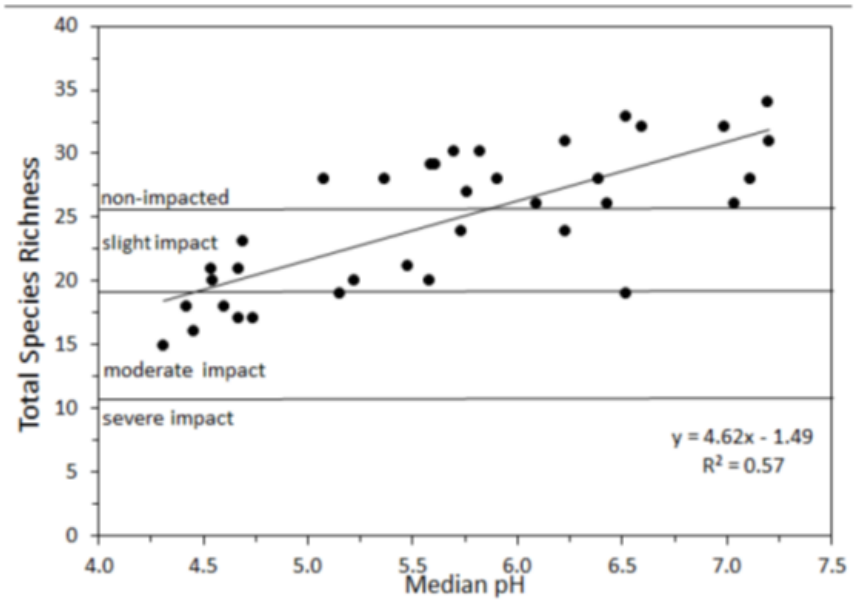

Figure 5-1. Total macroinvertebrate species richness as a function of pH in 36 streams in western Adirondack Mountains of New York, 2003-2005. From Baldigo et al. (2009); see ISA, Appendix 8, section 8.3.3 and p. 8-12.

Responses among fish species and life stages to changes in ANC, pH and Al in surface waters are variable (ISA, Appendix 8, section 8.3.6). Early life stages such as larvae and smolts are more sensitive to acidic conditions than the young-of-the-year, yearlings, and adults (Baker, et al., 1990; Johnson et al., 1987; Baker and Schofield, 1982). Some species and life stages experienced significant mortality in bioassays at relatively high pH (e.g., pH 6.0−6.5; ANC 50-100 μeq/L for eggs and fry of striped bass and fathead minnow [McCormick et al., 1989; Buckler et al., 1987]), whereas other species were able to survive at quite low pH without adverse effects. Many minnows and dace (Cyprinidae) are highly sensitive to acidity, but some common game species such as brook trout, largemouth bass, and smallmouth bass are less sensitive (threshold effects at pH <5.0 to near 5.5; ANC 20 and 50 μeq/L). A study by Neff et al. (2008) investigated the effects of two acid runoff episodes in the Great Smoky Mountains National Park on native brook trout using an in-situ bioassay. The whole-body sodium concentrations differed before and after the episodes. More specifically, the reduction in whole-body sodium when stream pH dropped below 5.1 (ANC 0 μeq/L) indicated that the trout had lost the ability to ionoregulate (ISA, Appendix 8, section 8.3.6.1). Field and laboratory bioassay studies indicate a wide variation in optimal pH range among fish species (Figure 5-2).

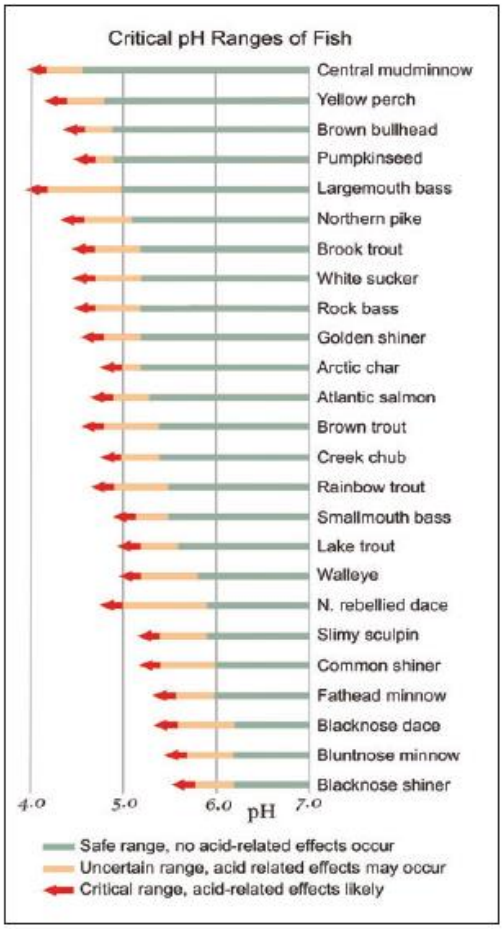

Figure 5-2. Critical aquatic pH range for fish species. Notes: Baker and Christensen (1991) generally defined bioassay thresholds as statistically significant increases in mortality or by survival rates less than 50% of survival rates in control waters. For field surveys, values reported represent pH levels consistently associated with population absence or loss. Source: Fenn et al. (2011) based on Baker and Christensen (1991). (ISA, Appendix 8, Figure 8-3)

As noted in the ISA, "[a]cross the eastern U.S., brook trout are often selected as a biological indicator of aquatic acidification because they are native to many eastern surface waters and because residents place substantial recreational and aesthetic value on this species," although compared to other fish species in Appalachian streams this species is relatively acid tolerant (ISA, Appendix 8, p. 8-26). For example, "[in many Appalachian mountain streams that have been acidified by acidic deposition, brook trout is the last fish species to disappear; it is generally lost at pH near 5.0 (MacAvoy and Bulger, 1995), which usually corresponds in these streams with ANC near 0 μeq/L (Sullivan et al., 2003)" (ISA, Appendix 8, p. 8-21).

As described in section 4.2.1 above episodic acidification during storm events can pose risks in low ANC streams. For example, streams with ANC around 20 μeq/L or less at base flow may be considered vulnerable to episodic acidification events that could reduce pH and ANC to

levels potentially harmful to brook trout and other species. Streams with suitable habitat and annual average ANC greater than about 50 μeq/L are often considered suitable for brook trout in southeastern U.S. streams, and reproducing brook trout populations are expected (Bulger et al., 2000). Streams of this type "provide sufficient buffering capacity to prevent acidification from eliminating this species and there is reduced likelihood of lethal storm-induced acidic episodes" (ISA, Appendix 8, p. 8-26). Results of a study by Andrén and Rydin (2012) suggested a threshold less than 20 ug/L Al and pH higher than 5.0 for healthy brown trout populations by exposing yearling trout to a pH and inorganic Al gradient in humic streams in Scandinavia (ISA, Appendix 8, section 8.3.6.2). Another recently available study that investigated the effects of episodic pH shifts fluctuations in waterbodies of eastern Maine reported that episodes resulting in pH dropping below 5.9 (ANC of ~50 μeq/L) have the potential for harmful physiological effects to Atlantic salmon smolts if coinciding with the smolt migration in eastern Maine rivers (Liebich et al., 2011; ISA, Appendix 8, section 8.3.6.2).

Investigations of waterbody recovery from historic deposition have reported on episodic acidification associated with the high SO_4^{2-} remaining in watershed soils. For example, monitoring data in the Great Smoky Mountains National Park indicated that while the majority of SO_4^{2-} entering the study watershed was retained, SO_4^{2-} in wet deposition moved more directly and rapidly to streams during large precipitation events, contributing to episodic acidification of receiving streams and posing increased risk to biota (ISA, Appendix 7, section 7.1.5.1.4). High flow episodes in historically impacted watersheds of the Appalachians have been reported to appreciably reduce stream ANC (Lawrence et al., 2015).

There is often a positive relationship between pH or ANC and number of fish species, at least for pH values between about 5.0 and 6.5, or ANC values between about 0 and 50 to 100 μeq/L (Cosby et al., 2006; Sullivan et al., 2006; Bulger et al., 1999). This is because energy cost in maintaining physiological homeostasis, growth, and reproduction is high at low ANC levels (Schreck, 1982; Wedemeyer et al., 1990). As noted in section 4.2.1.2 above, surveys in the heavily impacted Adirondack mountains found that lakes and streams having an annual average ANC < 0 μeq/L and pH near or below 5.0 generally support few or no fish species to no fish at all, as illustrated in Figure 5-3 below (Sullivan et al., 2006; ISA, Appendix 8, section 8.3.6.3.

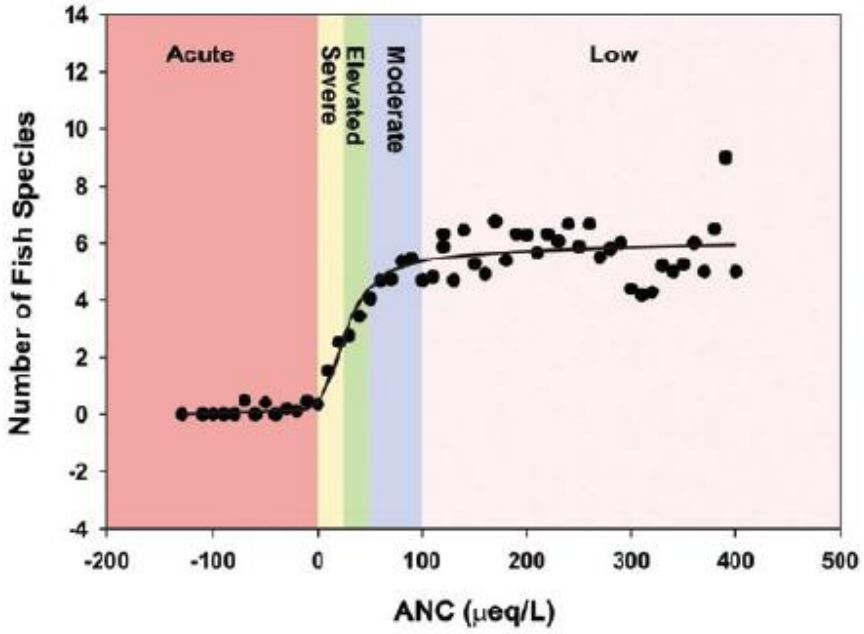

Figure 5-3. Number of fish species per lake *versus* acidity status, expressed as ANC, for Adirondack lakes. Notes: The data are presented as the mean (filled circles) of species richness within 10 μeq/L ANC categories, based on data collected by the Adirondacks Lakes Survey Corporation. Source: Modified from Sullivan et al. (2006) (ISA, Appendix 8, Figure 8-4).

The data presented in Figure 5-3 above suggest that there could be a loss of fish species in these lakes with decreases in ANC below approximately 50 to 100 μeq/L (Sullivan et al., 2006). For streams in Shenandoah National Park, a statistically robust relationship between ANC and fish species richness was also documented by Bulger et al. (2000). However, interpretation of species richness relationship with ANC can be difficult and misleading, because more species tend to occur in larger lakes and streams as compared with smaller ones, irrespective of acidity (Sullivan et al., 2006) because of increased aquatic habitat complexity in larger lakes and streams (Sullivan et al., 2003; ISA, Appendix 8, section 8.3.6.3).

Observations of effects in watersheds impacted by historic acidification can also reflect the influence of episodic high flow events that lower pH and ANC appreciably below the baseflow ANC (as described above). Studies described above are summarized below in the context of ANC ranges: <0, 0-20, 20-50, 50-80, and >80 μeq/L:

- At ANC levels <0 μeq/L, aquatic ecosystems have exhibited low to a near loss of aquatic diversity and small population sizes. For example, planktonic and macroinvertebrates communities shift to the most acid tolerant species (Lacoul et al., 2011), and mayflies can be eliminated (Baker and Christensen, 1991). A near to complete loss of fish populations can occur, including non-acid-sensitive native species such as brook trout (*Salvelinus fontinalis*), northern pike (*Esox lucius*), and others (Sullivan et al., 2003, 2006; Bulger et al., 2000), which is in most cases attributed to elevated inorganic monomeric Al

concentration (Baldigo and Murdoch, 1997). At this level, aquatic diversity is at its lowest (Bulger et al., 2000; Baldigo et al., 2009; Sullivan et al. 2006) with only acidophilic species being present.

- In waterbodies with ANC levels between 0 and 20 μeq/L, acidophilic species dominate other species (Matuszek and Beggs, 1988; Driscoll et al., 2001) and diversity is low (Bulger et al., 2000; Baldigo et al., 2009; Sullivan et al., 2006). Plankton and macroinvertebrate populations have been observed to decline, and acid-tolerant species have outnumbered non-acid-sensitive species (Liebich et al., 2011). Sensitive species are often absent (e.g., brown trout, common shiner) while non-sensitive fish species populations may be reduced (Bulger et al., 2000). Episodic acidification events (e.g., inflow with ANC <0 μeq/L and pH< 5), may have lethal impacts on sensitive lifestages of some biota, including brook trout and other fish species (Matuszek and Beggs, 1988; Driscoll et al., 2001).
- Levels of ANC between 20 and 50 μeq/L have been associated with the loss and/or reduction in fitness of aquatic biota that are sensitive to acidification in some waterbodies of the Adirondacks and Appalachians. Such effects included reduced aquatic diversity (Kretser et al., 1989; Lawrence et al., 2015; Dennis and Bulger, 1995) with some sensitive species missing (Bulger et al., 2000; Sullivan et al., 2006). In historically impacted watersheds, waterbodies with ANC below 50 μeq/L are more vulnerable to increased potential for harm associated with episodic acidification (ISA, Appendix 8, section 8.2). Comparatively, acid tolerant species, such as brook trout may have moderate to healthy populations (Kretser et al., 1989, Lawrence et al., 2015; Dennis and Bulger, 1995).
- At an ANC between 50 and 80 μeq L, the fitness and population size of some sensitive species have been affected in some historically impacted watersheds. Levels of ANC above 50 μeq/L are considered suitable for brook trout and most fish species because buffering capacity is sufficient to prevent the likelihood of lethal episodic acidification events (Driscoll et al., 2001; Baker and Christensen 1991). However, depending on other factors, the most sensitive species have been reported to experience a reduction in fitness and/or population size in some waterbodies (e.g., blacknose shiner [Baldigo et al., 2009; Kretser et al., 1989; Lawrence et al., 2015; Dennis and Bulger, 1995]). Fish species richness has also been reported to be affected in some Adirondack streams at ANC 50 (Sullivan et al., 2006).
- Values of ANC >80 μeq/L have generally not been associated with harmful effects on biota (Bulger et al., 1999; Driscoll et al., 2001; Kretser et al., 1989; Sullivan et al., 2006).

5.1.2 Conceptual Model and Analysis Approach

The impact of acidifying deposition on aquatic ecosystems across the U.S. was evaluated in this review by developing analyses using a CL approach with ANC as the acidification indicator. This approach provides a means of assessing risk to a group of lakes, streams, and rivers (i.e., waterbodies) in a given area from various levels of N and/or S deposition. ANC was used as the water quality metric where ANC targets (see description of the 5 categories above) were used to correspond to different levels of acidification risk. This approach was used to

characterize the risk of acidifying deposition on aquatic acidification across the contiguous U.S. (CONUS) with a focus on acid-sensitive areas.

These linkages between acidifying deposition of nitrogen and sulfur; water chemistry changes (reflected by changes in ANC and pH); and waterbody health and biodiversity are the basis for the quantitative assessments that were performed in this review and provide the foundation for describing the potential impacts from acidification across the U.S. The following schematic (Figure 5-4) represents the conceptual model used in the analyses to link these factors.

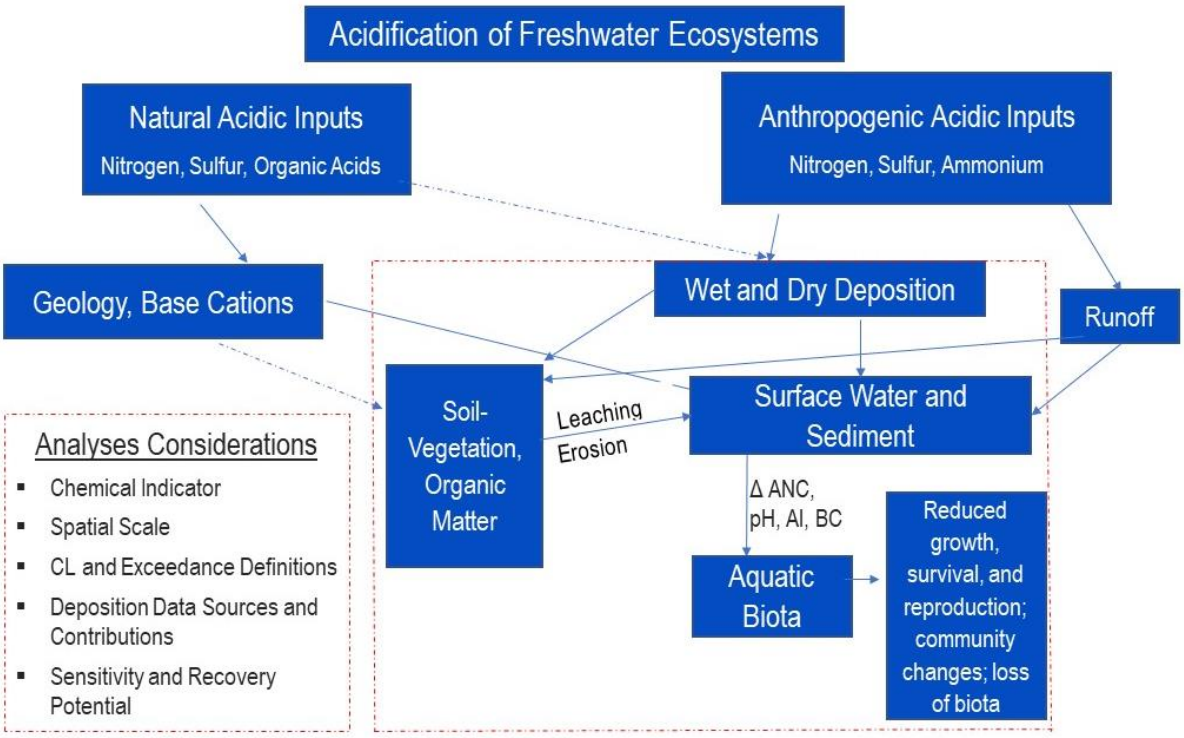

Figure 5-4. Conceptual model for aquatic acidification analyses.

In the analyses described below, waterbody estimates of deposition were compared to atmospheric loading (CLs) estimated to support ANC levels equal to each of several targets (described in section 5.1.2.2 below). Depending on the ANC target, low CL values may indicate that the watershed has a limited ability to neutralize the addition of acidic anions and, hence, may be susceptible to acidification as a result of acidic inputs. In general, the higher the CL value, the greater the ability of a given watershed to neutralize additional acidic anions. Similarly, for any specific ANC target, lower CL estimates are associated with more acid-sensitive waterbodies. Further, given the negative relationship between acidic loading and ANC, the CL estimates for any one waterbody are lower for the higher ANC targets.

Key aspects of the assessments described in the subsections below include the spatial scales of assessment (section 5.1.2.1), the chemical indicator (section 5.1.2.2), identification of CL estimates for this assessment (section 5.1.2.3) and determining exceedances (section 5.1.2.4), as well as sources of waterbody deposition estimates (section 5.1.2.5). Also discussed is the approach for interpreting results, including regarding ecosystems with sensitivity to acidic deposition, ecosystems for which factors other than deposition are critical influences on waterbody ANC, and waterbodies for which CL estimates above zero cannot be derived for ANC levels of interest. Results of the assessments are presented in section 5.1.3. The characterization of uncertainty is described in section 5.1.4, and key findings are summarized in section 5.1.5.

5.1.2.1 Spatial Scale

For this assessment, we developed a multi-scale analysis to assess aquatic acidification at three levels of spatial extent: national, ecoregion, and case study. The national assessment included the CONUS only since there are insufficient data available for Hawaii, Alaska, and the territories. The Omernik ecoregion classifications were used for the regional assessments, and case study locations were areas likely to be most impacted and for which sufficient data were available. Further discussion of these spatial scales can be found below. Since acidification of waterbodies is controlled by local factors such as geology, hydrology, etc. the aquatic CLs for acidification are unique to the waterbody itself, and information about the waterbody, like water quality, is needed to determine its CL. For these reasons, CLs were determined at the waterbody level and then summarized at the national, ecoregion, and case study level. The national assessment is a combined summary of aquatic CLs across the CONUS.

It is important to note that aquatic ecosystems across the CONUS exhibit a wide range of sensitivity to acidification because of multiple landscape factors, such as geology, hydrology, soils, catchment scale, and vegetation characteristics, that control whether a waterbody will be acidified by atmospheric deposition. Consequently, variations in ecosystem sensitivity must be taken into account in order to characterize sensitive populations of waterbodies and relevant regions across the CONUS. The EPA's Omernik Ecoregions classifications were used to define ecologically relevant, spatially aggregated, acid-sensitive regions across the CONUS in order to better characterize the regional difference in the impact of deposition-driven aquatic acidification.

Ecoregions are areas of similarity regarding patterns in vegetation, aquatic, and terrestrial ecosystem components. The Omernik ecoregion categorization scheme categorizes ecoregions using a holistic, "weight-of-evidence" approach in which the relative importance of factors may vary from region to region (Omernik, 1987). The method used to map ecoregions is described in Omernik (1987) and classifies regions through the analysis of the patterns and the composition of

biotic and abiotic phenomena that affect or reflect differences in ecosystem quality and integrity. Factors include geology, physiography, vegetation, climate, soils, land use, wildlife, and hydrology. Four hierarchical levels of ecoregions distinguish coarser (more general) and finer (more detailed) categorization (Omernik and Griffith, 2014). Level I is the coarsest level, dividing North America into 12 ecoregions. At level II, the continent is subdivided into 25 ecoregions and the contiguous U.S. (CONUS) into 20 ecoregions (Figure 5-5). Level III is a further subdivision of level II and divides CONUS into 84 ecoregions. Level IV is a subdivision of level III and divides CONUS into 967 ecoregions. For the analyses in this review, we used the level III categorization to give the greatest sensitivity for variation in ecoregion response while allowing us to aggregate available water quality data while maintaining its representativeness.

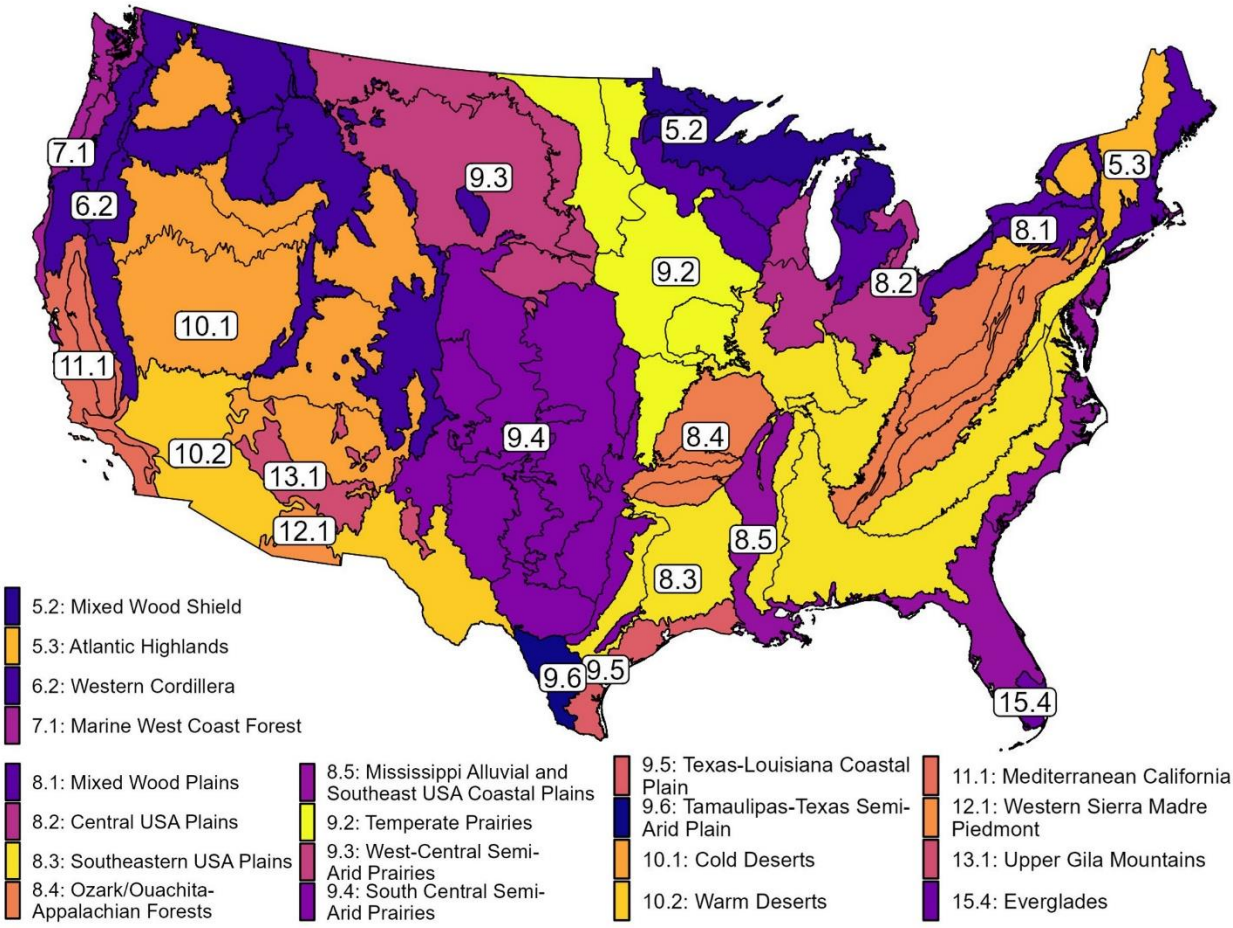

Figure 5-5. Level II ecoregions of the contiguous U.S.

In order to focus our analyses on those areas that were likely to be affected by acidification and that were also driven primarily by deposition of N and S from ambient air, we looked more closely at the ecoregions and their underlying characteristics. We also identified

those ecoregions where, for various reasons, target ANC values could not be achieved. These factors are discussed fully in the REA presented in Appendix 5A, and summarized below.

Based on this analysis, 30 ecoregions were identified as sensitive to acidification (Appendix 5A, Table 5A-5). Of these 30 ecoregions, three were identified as having natural acidity, based on DOC as an indicator of natural acidity (ISA, Appendix 7, section 7.1.2.5; 2008 ISA, section 3.2.4.2 and Annex B, p. B-35). The acid-sensitive ecoregions are most generally areas with mountains, high elevation terrain or waterbodies in northern latitudes (northern areas of Minnesota, Wisconsin, and Michigan; and New England). The northern, non-mountainous regions that are sensitive to acidity share attributes (e.g., growing season, vegetation, soils, and geology) similar to mountainous regions and typically are located in rural areas, often in tracts of designated wilderness, park and recreation areas. The three naturally acidic ecoregions, located on eastern coastal plain, were excluded from the analyses because of their natural acidity indicated by high DOC values: (1) Middle Atlantic Coastal Plain (8.5.1), (2) Southern Coastal Plains (8.5.3), and (3) Atlantic Coastal Pine Barrens (8.5.4). These ecoregions generally lie along the Atlantic coast from New Jersey south to northern Florida (Figure 5-6). A more complete discussion of ecoregion sensitivity can be found in Appendix 5A, section 5A.1.7.

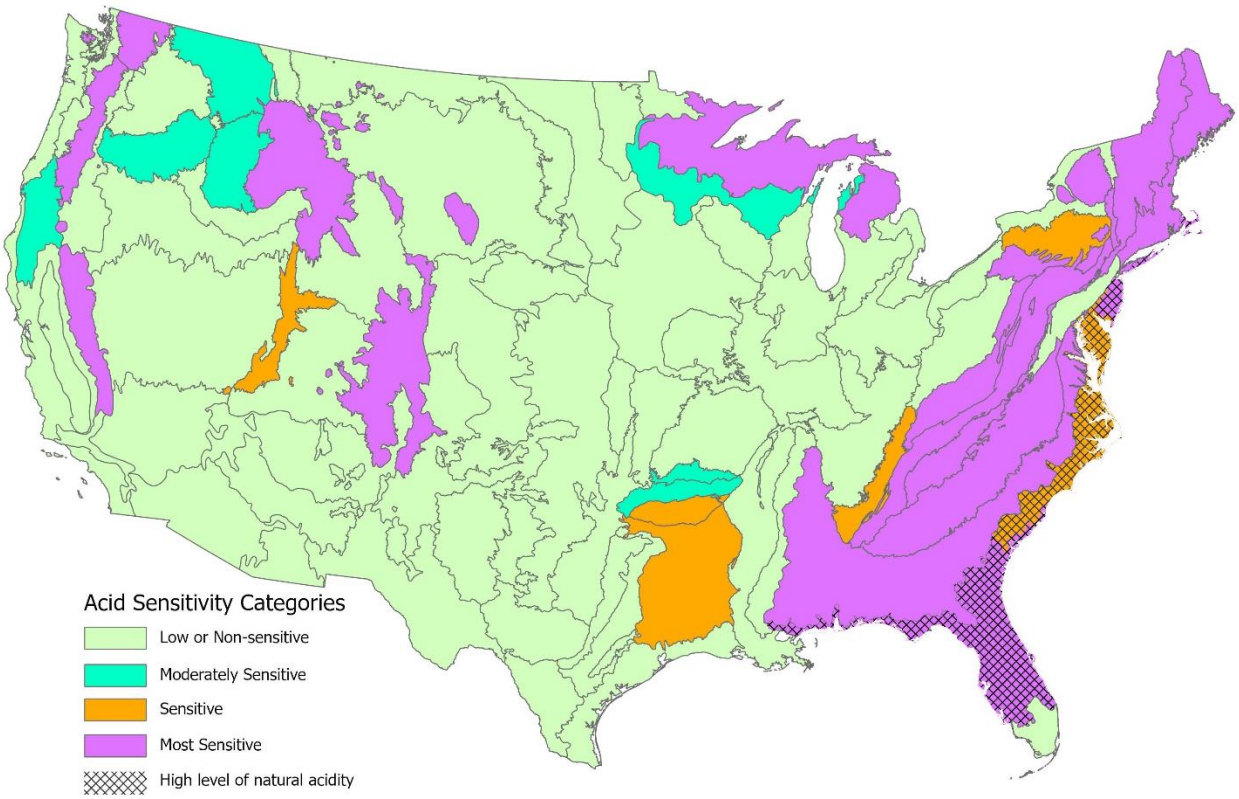

Figure 5-6. Level III ecoregions grouped into acid sensitivity categories.

The case study scale represents the smallest scale at which we performed our analyses and is intended to give some insight into the local impact of aquatic acidification. Five case study areas across the U.S. were examined: Shenandoah Valley, White Mountain National Forest, Northern Minnesota, Sierra Nevada Mountains, and Rocky Mountain National Park. These areas include several parks and national forests that vary in their sensitivity to acidification but represent high value or protected ecosystems, such as Class 1 areas, wilderness, and national forests (as further described in Appendix 5A, section 5A.2.3.2).

5.1.2.2 Chemical Indicator

The chemical indicator of acidification risk used in this assessment is ANC, as calculated in model simulations (described in Appendix 5A, section 5A.1.5). Although biological effects are primarily attributable to low pH and high inorganic aluminum concentration, ANC is more commonly used for estimating CLs for N and S in the U.S. as it is a more stable and more easily modelled, as described in Appendix 5A, section 5A.1 (ISA, section ES.5.1 and Appendix 7, section 7.1.2.5). Additionally, CL estimates generally are linearly associated with ANC levels. In our use of ANC, we have also looked most closely at those waterbodies for which deposition was the main source of acidifying input and eliminated from consideration those waterbodies for which either other sources of acidifying input were significant (for example, runoff) or for which natural conditions were such that those waterbodies would be unable to reach specific ANC thresholds.

For the analyses described below, we evaluated CLs for three different ANC targets: 20 μeq/L, 30 μeq/L and 50 μeq/L. Selection of these target ANC values reflects several considerations. For example, most aquatic CL studies conducted in the U.S. since 2010 use an ANC of 20 and/or 50 μeq/L, because 20 μeq/L has been suggested to provide protection for "natural" or "historical" range of ANC and 50 μeq/L provides greater protection (Dupont et al., 2005; McDonnell et al., 2012, 2014; Sullivan et al., 2012a, 2012b; Lynch et al., 2022; Fakhraei et al., 2014; Lawrence et al., 2015). In the western U.S., lakes and streams vulnerable to deposition-driven aquatic acidification are often found in the mountains where surface water ANC levels are naturally low and typically vary between 0 and 30 μeq/L (Williams and Labou, 2017; Shaw et al., 2014). For these reasons, previous studies and the National Critical Load Database (NCLD) uses an ANC threshold of 50 μeq/L for the eastern CONUS and 20 μeq/L for the western CONUS (denoted as "50/20" μeq/L). With regard to higher ANC levels, such as 80 μeq/L, it was also recognized that many waterbodies, particularly in acid-sensitive regions of CONUS never had an ANC that high and would never reach an ANC that high naturally (Williams and Labou 2017; Shaw et al., 2014). Additionally, in conveying its advice in the 2012 review, the CASAC expressed its view that "[l]evels of 50 μeq/L and higher would provide

additional protection, but the Panel has less confidence in the significance of the incremental benefits as the level increases above 50 μeq/L" (Russell and Samet, 2010; pp. 15-16).

For the analyses included below, ANC target values of 20, 30 and 50 μeq/L were selected for the following reasons:

ANC of 20 μeq/L :

- In western high elevation sites, ANC is typically below 50 μeq/L (e.g., median around 30 μeq/L in Sierra Nevada) even though acidifying deposition is low at those sites (Shaw et al., 2014). Accordingly, a target of 20 μeq/L is commonly considered an appropriate target for western sites.
- ANC levels below 20 μeq/L in sensitive Shenandoah/Adirondack waterbodies are associated with significant/appreciable reduction in fish species (Bulger et al., 2000; Sullivan et al., 2006). Thus, ANC of 20 μeq/L is considered a minimum/lower bound target for such eastern mountain sites.

ANC of 30 μeq/L:

- While ecological effects occur at ANC levels at 30 μeq/L in some sensitive ecosystems (based primarily on studies in Shenandoah/Adirondack waterbodies), the degree and nature of those effects are less significant than at levels below 20 μeq/L.
- Research in New England, the Adirondacks and Northern Appalachian Plateau indicates ANC of 30-40 μeq/L may protect from spring episodic acidification in those watersheds (Driscoll et al., 2001; Baker and Christensen, 1991).

ANC of 50 μeq/L

- ANC of 50 μeq/L is is commonly cited as a target for eastern sites (Dupont et al., 2005; McDonnell et al., 2012; McDonnell et al., 2014; Sullivan et al., 2012a; Sullivan et al., 2012b; Lynch et al., 2022; Fakhraei et al., 2014; Lawrence et al., 2015).
- In the 2012 review, ANC values at/above 50 μeq/L were concluded to provide additional protection although with increasingly greater uncertainty for values at/above 75 μeq/L (2011 PA, pp. 7-47 to 7-48).

5.1.2.3 Critical Load Estimates Based on ANC

Considerable new research on critical loads for acidification is available since the 2008 ISA and both steady-state and dynamic models have been used to generate ANC-based critical loads for much of the U.S. (ISA, Appendix 8, section 8.5.4.1.2). Steady-state CLs are calculated from mass-balance models under assumed or modeled equilibrium conditions based in part on water quality measurements. While the models used to derive steady-state CLs vary in complexity, fundamentally they rely on the calculation of elemental mass balances. Dynamic models have also been used to develop CLs. These models simulate soil or water chemistry or biological response to calculate a target within a specified time period, such as by the year 2100, and they can also be used to calculate a CL comparable to a long-term steady-state CL by

applying the model to a date in the distant future. Since the 2008 ISA, studies utilizing dynamic modeling of CLs have generally been focused on the Adirondacks, the Appalachians, and the Rocky Mountains or Sierra Nevada (ISA, Appendix 8, section 8.5.4.1.2.2).

Aquatic CLs used in this assessment came from the NCLD version 3.2.1 (Lynch et al., 2022), and studies identified in the ISA (e.g., Shaw et al., 2014; McDonnell et al., 2014; Sullivan et al., 2012a). The NCLD is comprised of CLs calculated from several common models, including the steady-state mass-balance model (SMBE), Steady State Water Chemistry (SSWC) model, and dynamic models such as the Model of Acidification of Groundwater In Catchment (MAGIC) run out to year 2100 or 3000. The overwhelming majority of CLs (more than 90%) are based on application of the SSWC model (as described in Appendix 5A, section 5A.1.5). Data in the NCLD are focused on waterbodies that are typically impacted by deposition driven acidification. A waterbody[3] is represented as a single CL value. In many cases where more than one CL value has been estimated for a waterbody (e.g., via different studies) the CL from the most recent study was selected or, when the CL estimates are from publications of the same timeframe, they were averaged for our analysis (see Appendix 5A, section 5A.1.5). The unique locations for the 13,824 CLs used in this assessment are indicated in Figure 5-12 below.

There are several newly available studies using steady-state modeling. Sullivan et al. (2012b) and McDonnell et al. (2012) developed an approach for deriving regional estimates of base cation weathering to support steady-state CL estimates for the protection of southern Appalachian Mountain streams against acidification. Calculated CL values were low at many locations, suggesting high acidification sensitivity. In the Blue Ridge ecoregion, calculated CL values to maintain stream ANC at 50 μeq/L were less than 500 equivalents per hectare per year (eq/ha-yr) at one third of the study sites. In another model simulation for Appalachian Mountain streams, McDonnell et al. (2014) calculated critical values, including steady-state aquatic CLs to protect streams against acidification. They based the CLs on ANC thresholds of 50 μeq/L, and nearly one-third of the stream length assessed in the study region had a CL for S deposition below 500 eq/ha-yr (ISA, Appendix 8, section 8.6.8).

Critical loads have most frequently been developed for waterbodies concentrated in areas that are acid sensitive, primarily, the eastern U.S. and the Rocky Mountain and Pacific Northwest regions of the West. Not all waterbodies are sensitive to acidification. As noted in the ISA, "acid-sensitive ecosystems are mostly located in upland mountainous terrain in the eastern and western U.S. and are underlain by bedrock that is resistant to weathering, such as granite or quartzite sandstone" (ISA, Appendix 7, p. 7-45). Small to median size lakes (>200 Ha) and lower

[3] A waterbody for the purposes of our analyses is a unique stream or lake represented in the critical loads database.

order-streams tend to be the waterbodies that are impacted by deposition driven acidification, while rivers are not typically impacted (ISA, Appendix 7, section 7.1.2).

5.1.2.4 Critical Load-Based Analysis

In this analysis, we compared waterbody deposition estimates to critical loads derived for the three ANC targets. As well documented in the evidence, deposition of both S and N contributes to acid deposition and associated acidification risk of a waterbody. However, as not all N deposition to a watershed will contribute to acidification, evaluating acidic deposition for N and S together is complex. Nitrogen deposition inputs below what is removed by long-term N processes in the soil and waterbody (e.g., N uptake and immobilization) do not contribute to acidification, but the amount above this minimum will likely contribute to acidification. Therefore, if N removal is greater than N deposition, only S deposition will contribute to the acidification and thereby to any potential for exceedance of the acidification CL (ISA, Appendix 7, section 7.1.2.1). The analyses performed for this PA first investigated the contribution to acidification from N deposition and, based on the finding of little appreciable contribution, then focused on S only deposition (Appendix 5A, section 5A.2.1).

This analysis focused on the S component of acidic deposition due to the finding of little appreciable contribution of N deposition to acidification beyond that associated with S deposition. For 2014-2016 and 2018-2020 deposition estimates, very few CL exceedances were driven by N. Thus, adding N from leaching to the critical load exceedances with S was not found to substantially change the percent of waterbodies that exceed their CL. This was found for national-scale analyses that compared the percentage of CL exceedances in waterbodies with both N and S exceedance versus only S exceedances (see Appendix 5A, section 5A.2.1). The results of these national-scale analyses support the assumption that most of the N deposition entering the watersheds during the analyses' time periods was retained within the watershed and/or converted to gaseous N (e.g., N_2O, N_2, etc.). Different methods have been developed to determine the amount of N deposition that acidifies related to aquatic CL exceedances. There are two common approaches in the studies that derived CLs used in this assessment: the first approach is based on the amount of "N leaching" to the waterbody determined by the amount of dissolved N in the water measured as the concentration of nitrite and runoff as presented in Henriksen and Posch, (2001).[4] The second approach is the use of a "set value" based on long-term estimate of N immobilization and denitrification as described by McNulty et al. (2007). Those methods and the details for calculating CL exceedance are also discussed in Appendix 5A, section 5A.1.6.2.

[4] Analyses in the Appendix 5A, section 5A.3.2 evaluate uncertainty associated with the input data for this approach.

However, it is important to take into account the uncertainty associated with the CL estimates in the calculation of CL exceedances. Specifically, in the analyses for this REA, CLs are exceeded when the S deposition estimates are greater than the CLs by at least a margin of 3.125 milliequivalents per square meter per year (meq S/m^2-yr) or 0.5 kg S/ha-yr. An exceedance was not concluded when the S deposition estimate was below the CL by less than 3.125 meq S/m^2-yr or 0.5 kg S/ha-yr. Estimates of S deposition that are within 3.125 meq S/m^2-yr or 0.5 kg S/ha-yr of the CL are described for the purpose of our analyses as being "at" the CL. This factor is derived from the CL uncertainty analysis (see Appendix 5A, section 5A.3**)**.

Estimates of CL less than zero indicate that a target ANC value is not expected to be reached regardless of the level of acidifying deposition. Areas with negative CLs, by and large, are those that, due to either base cation loss from past deposition or natural conditions, would not be able to achieve the target ANC values of 20, 30 or 50 µeq/L under any deposition scenario. In our analyses, exceedances are reported separately for these areas from those areas with CL estimates greater than zero (see Appendix 5A, section 5A.2.1).

5.1.2.5 Waterbody Deposition Estimates

Estimates of waterbody deposition used in this assessment were based on the Total Deposition (TDep) model.[5] This model is discussed more fully in section 2.5. Both total N and S deposition were estimated at a resolution of a 4 km grid cell for each stream reach or lake location. For each waterbody, total N and S deposition were determined for each year from 2000 to 2020 and used to derive three-year averages for five periods: 2001-03, 2006-08, 2010-2012, 2014-16 and 2018-20. The extent of critical load exceedances across the waterbodies with CLs was then calculated for each of these five periods and summarized nationally and by ecoregion (sections 5.1.3.1 and 5.1.3.2).

5.1.3 Estimates for Achieving ANC Targets with Different Deposition Levels

The aquatic acidification assessments developed for this review are intended to estimate the ecological exposure and risk posed to aquatic ecosystems from the acidification effects of S and/or N deposition at varying levels to sensitive regions across the CONUS. They were performed at three spatial scales of differing levels of complexity. The results of these analyses are presented below. Section 5.1.3.1 presents the results of the national-scale analyses whereas sections 5.1.3.2 and 5.1.3.3 present the results of the ecoregion-scale and case study analyses respectively.

[5] The TDep modeling approach was developed by Schwede and Lear (2014) and the recent iterations are documented on the TDep website (https://nadp.slh.wisc.edu/committees/tdep/). Data were downloaded for exceedance calculations on September 26, 2022.

5.1.3.1 National-scale Analysis

A total of 13,824 unique waterbodies across the CONUS had calculated CLs available in NCLD. Most of those waterbodies had CLs that were less than 18 kg S/ha-yr across all the target ANC levels (Appendix 5A, Table 5A-6). Note that as discussed above, for the purpose of this analysis we focused here on CL estimates greater than zero (CL>0) and S only. The 50/20 values reflect a threshold ANC of 50 µeq/L in the eastern portion of the U.S. and one of 20 µeq/L in the west.[6] For the waterbody sites with CL values above zero,[7] Table 5-1 contains a summary of the percent of waterbodies with CL exceedances for S only for annual average deposition in the five 3-year periods for the ANC thresholds for an ANC of 20, 30, 50, and 50/20 µeq/L (additional detail in Appendix 5A, Table 5A-7).

Table 5-1. Percentage of waterbodies nationally for which annual average S deposition during the five time periods assessed exceed the waterbody CL (for CLs>0) for each of the ANC targets.

ANC (µeq/L)	2018-20	2014-16	2010-12	2006-08	2001-03
20	1%	3%	5%	16%	22%
30	2%	4%	7%	19%	25%
50	4%	6%	11%	24%	28%
50/20	4%	6%	10%	23%	28%

The geographic distribution of the waterbodies for which S deposition during the five time periods exceeded CLs for the target ANC values is shown in Figures 5-7 to 5-11. Most exceedances occurred in New England, the Adirondacks, the Appalachian Mountain range (New England to Georgia), the upper Midwest, Florida, and the Sierra Nevada mountains in California as expected. As discussed above, waterbodies in Florida and other coastal plain ecoregions that exceed the CL are likely not related to deposition of S, but instead are related to high levels of natural acidity in these drainage waters. These drainage waters tend to be naturally high in dissolved organic carbon, causing these systems to be acidic. Because these are waterbodies that are highly sensitive to acidification and likely naturally acidic, they exceed the calculated CL at any deposition amount. These three ecoregions (8.5.1, 8.5.3 and 8.5.4) are not included in the

[6] Consistent with regional definitions based on groups of states that were employed in the last review, in analyses in this PA, the West includes the states of ND, SD, CO, WY, MT, AZ, NM, UT, ID, CA, OR, WA (2009 REA, Appendix 1, p. 1-21). Accordingly, an ecoregion is designated western if it intersects or overlaps with these ten states, and eastern ecoregions are those not designated as western.

[7] For ANC threshold of 50 µeq/L, there are 13,184 sites with CL values above zero, 13,649 for ANC of 30 µeq/L and 13,771 for ANC of 20 µeq/L. For ANC of 50 (East) and 20 (West), 13,344 sites have CL values above zero.

ecoregion-scale analyses (see section 5.1.3.2). For more information on these areas see Appendix 5A, section 5A.2.1.

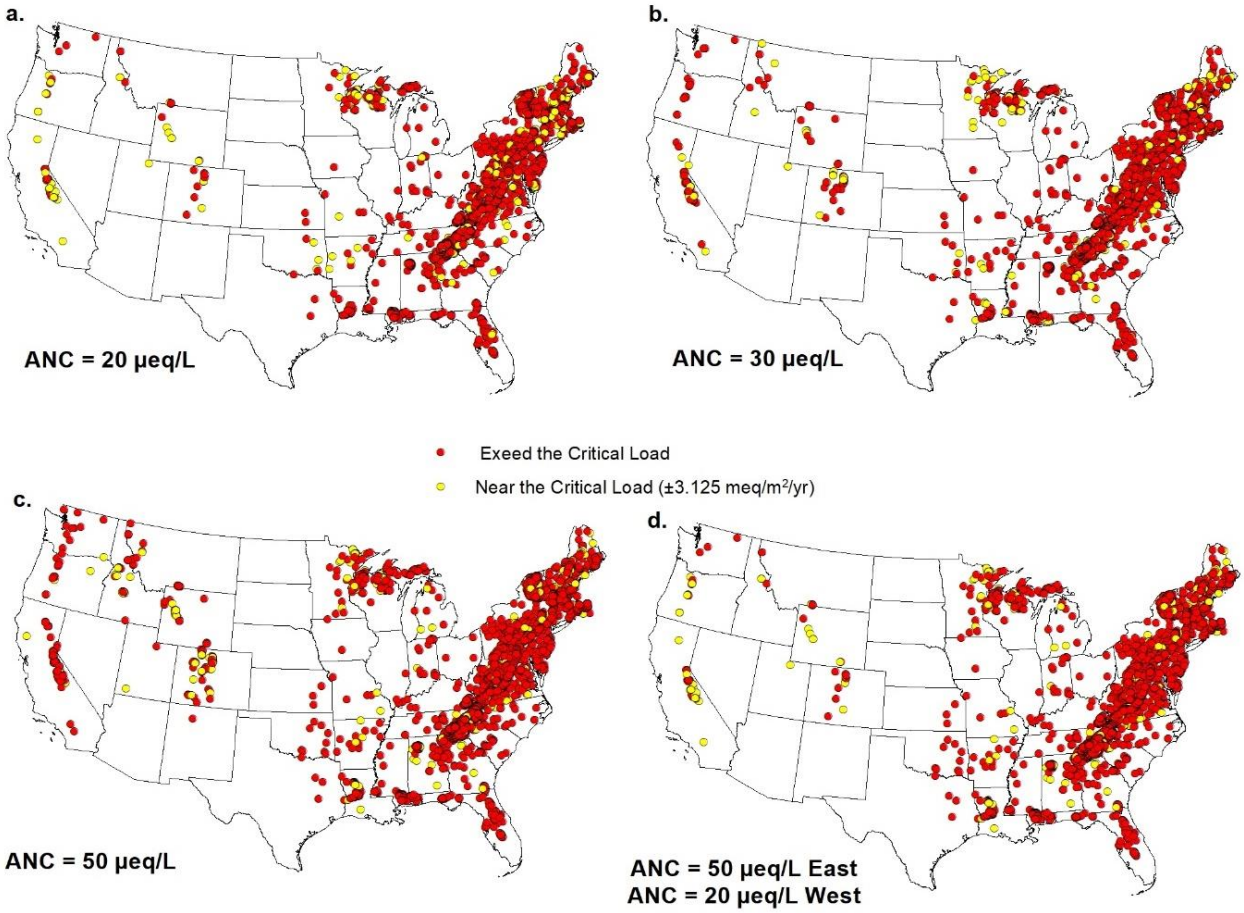

Figure 5-7. Waterbodies for which annual average S only deposition for 2001-03 exceed CLs for ANC thresholds: a. 20, b. 30, c. 50, d, 50/20 µeq/L.

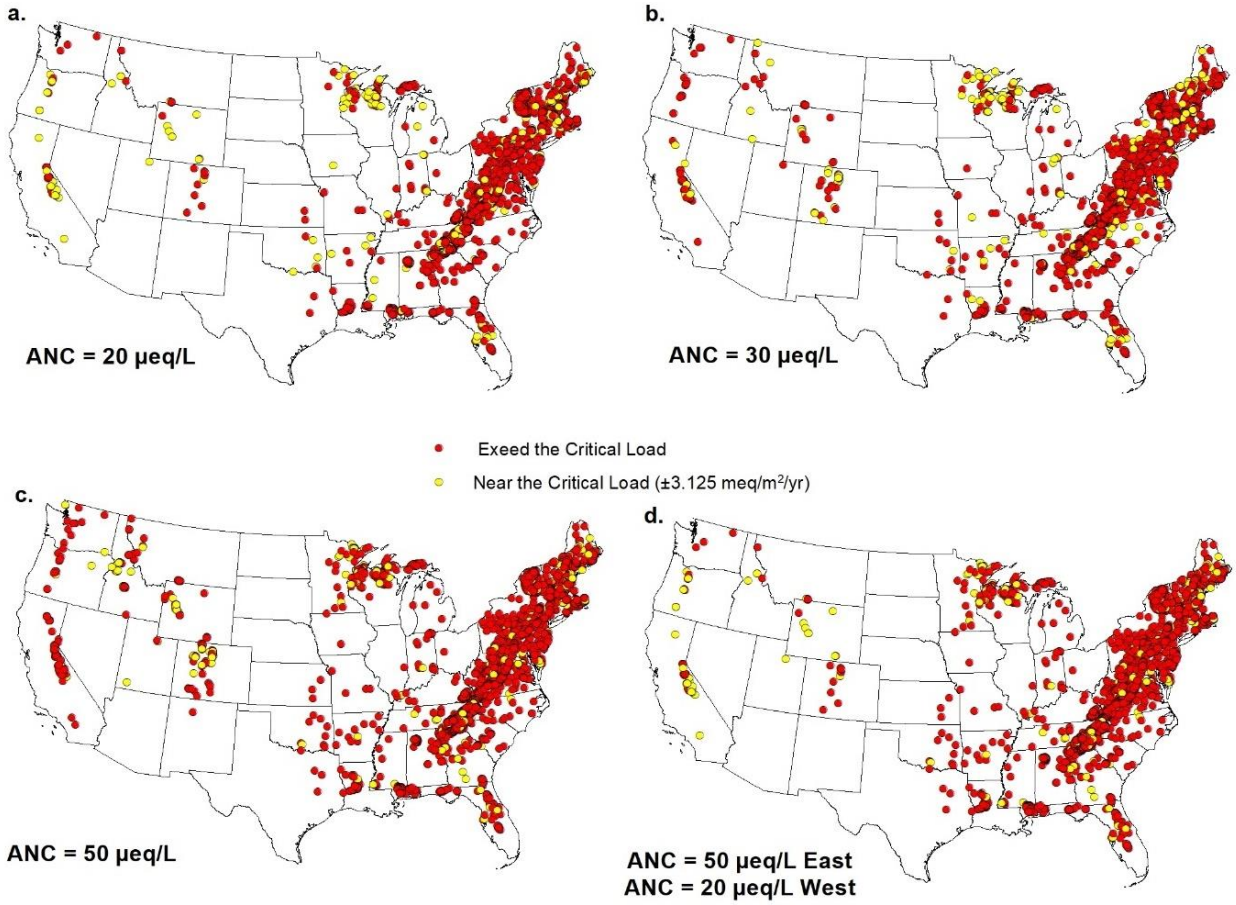

Figure 5-8. Waterbodies for which annual average S only deposition for 2006-08 exceed CLs for ANC thresholds: a. 20, b. 30, c. 50, d, 50/20 µeq/L.

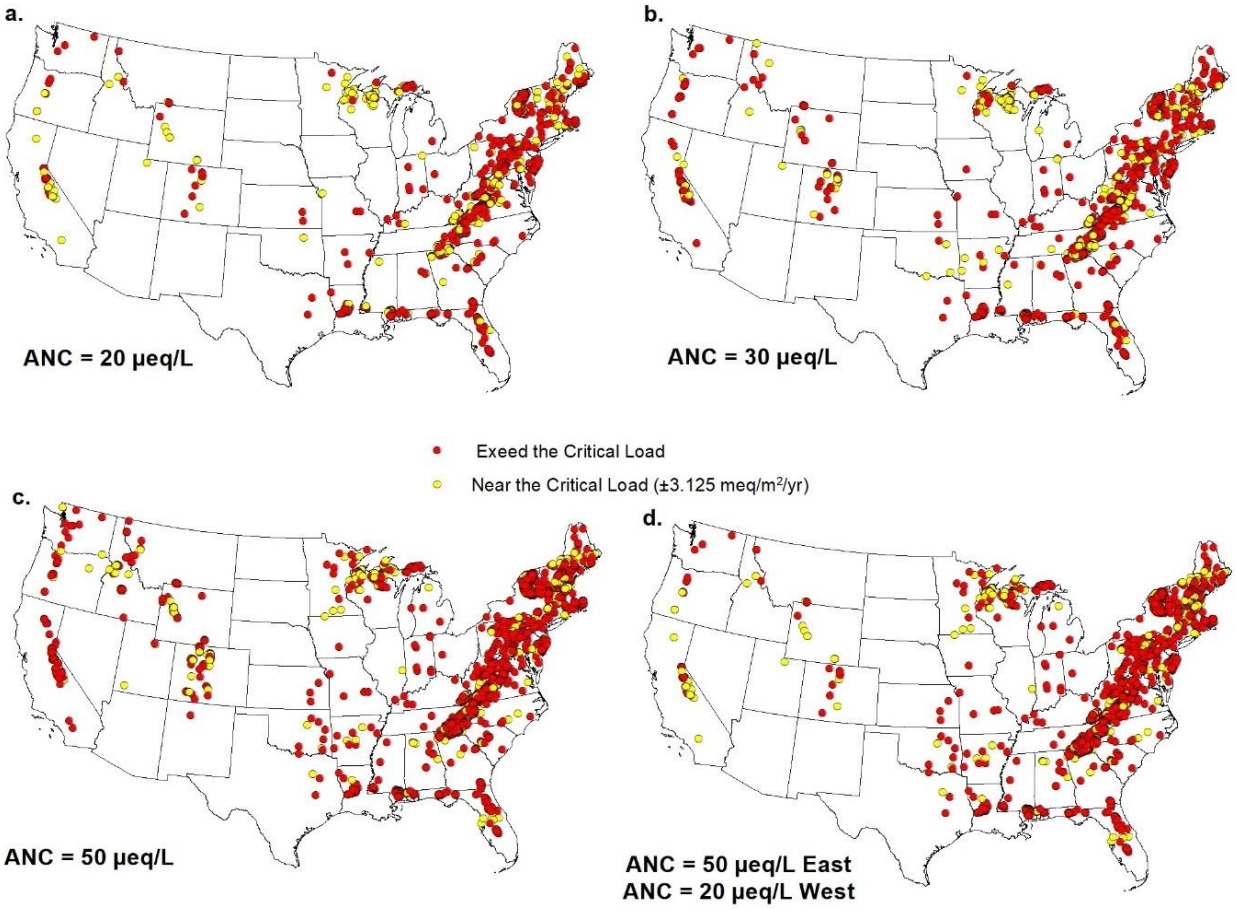

Figure 5-9. Waterbodies for which annual average S only deposition for 2010-12 exceed CLs for ANC thresholds: a. 20, b. 30, c. 50, d, 50/20 µeq/L.

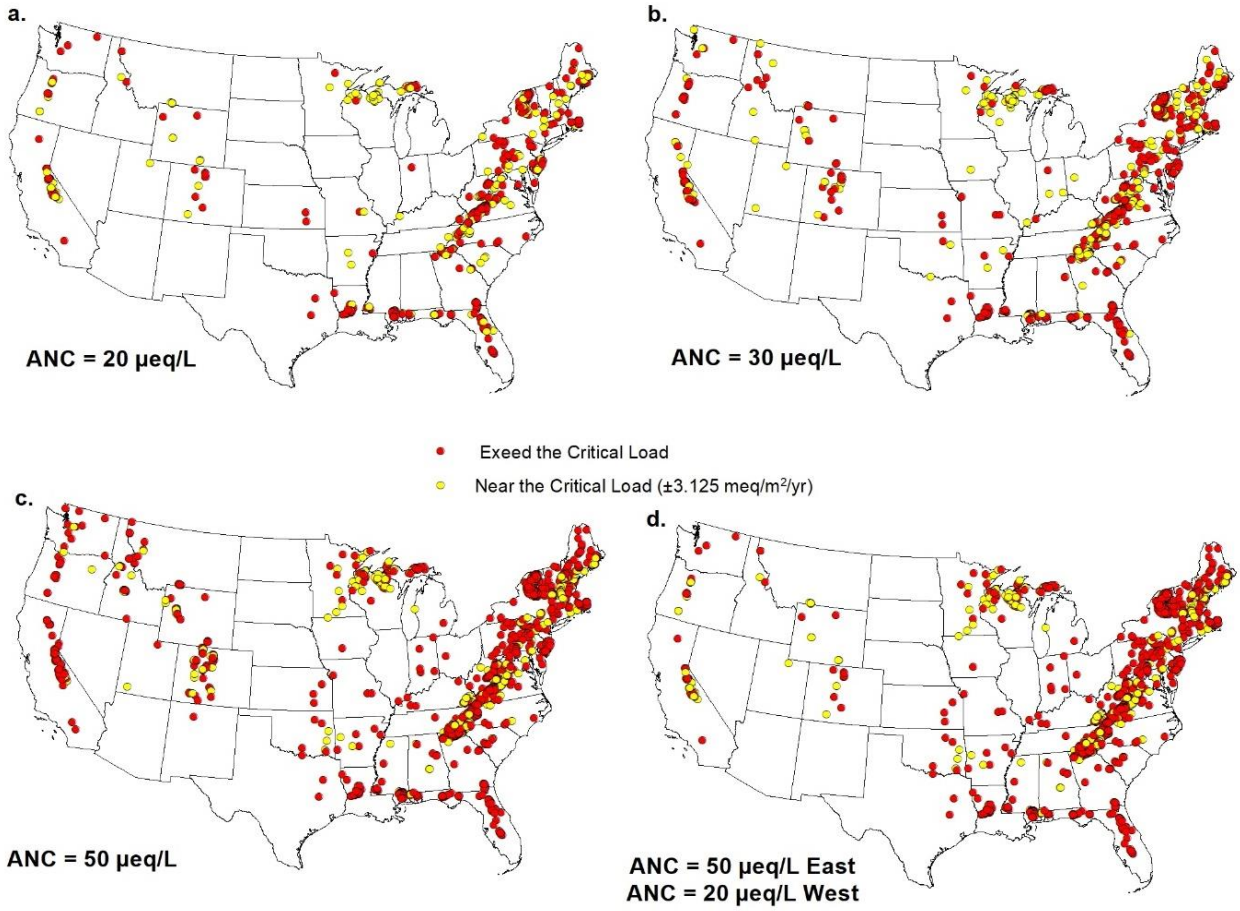

Figure 5-10. Waterbodies for which annual average S only deposition for 2014-16 exceed CLs for ANC thresholds: a. 20, b. 30, c. 50, d, 50/20 µeq/L.

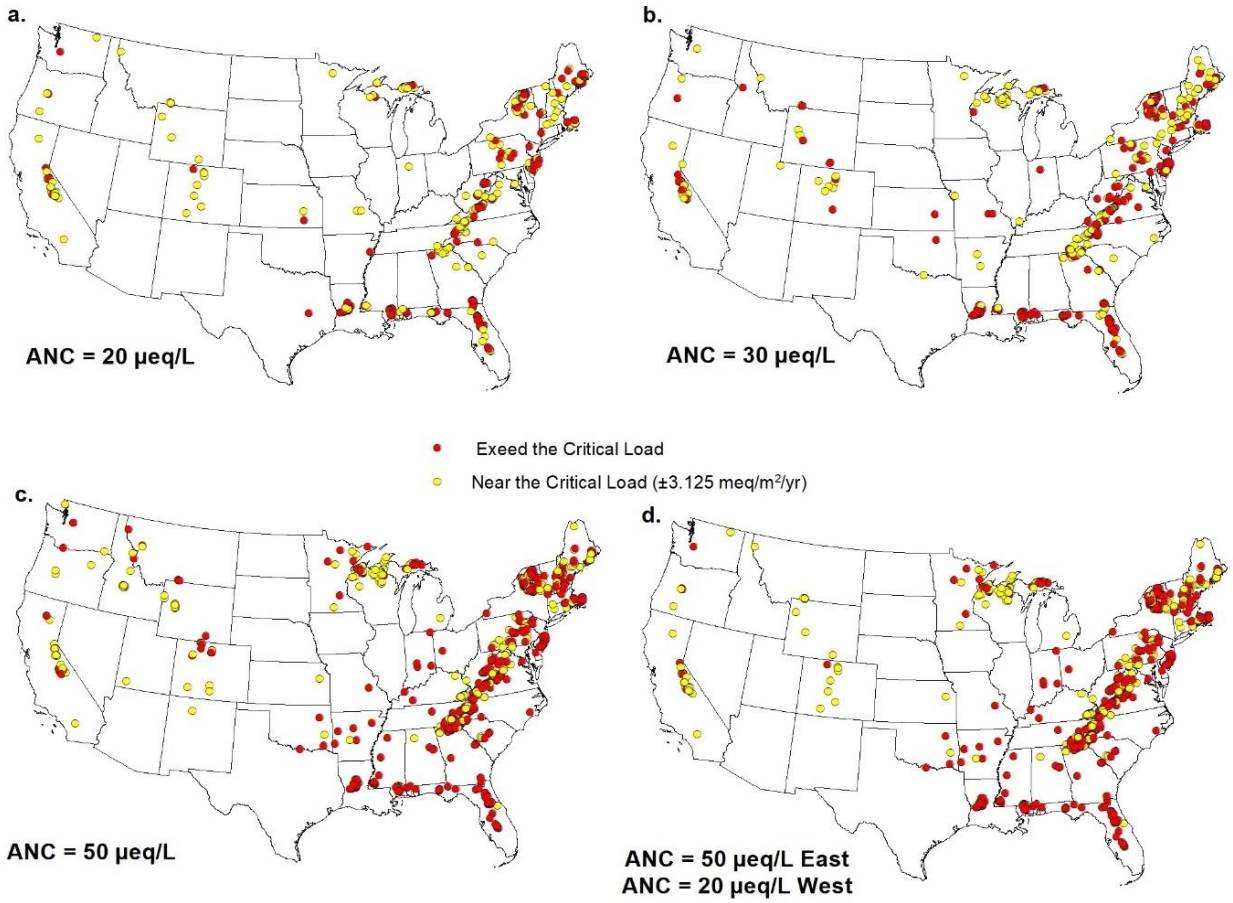

Figure 5-11. Waterbodies for which annual average S only deposition for 2018-20 exceed CLs for ANC thresholds: a. 20, b. 30, c. 50, d, 50/20 µeq/L.

The results of the national-scale analyses show a significant reduction in exceedances over time as sulfur deposition has decreased (see section 2.5.4 above for temporal trends in deposition across the U.S.). It also provides the foundation for the additional analyses below to look at what impacts might be expected under different geographic scales and deposition scenarios.

5.1.3.2 Ecoregion Analyses

The ecoregion-level analyses, summarized below, focused on level III ecoregion delineations (from this point on the term ecoregions refers to ecoregions delineated to level III). These analyses provide further characterization of both spatial variability of acid-sensitive waterbodies across the U.S. and the extent of deposition driven acidification impacts. Since the acidification of waterbodies is controlled by local factors such as geology and hydrology, aquatic CLs for acidification are unique to the waterbody itself and information about the waterbody, like water quality, is needed to determine its critical load. Unfortunately, not all waterbodies within an ecoregion have sufficient data to calculate a CL. This is the case for many ecoregions,

although generally ecoregions in historically recognized acid-sensitive areas have been heavily sampled, and, hence, include many waterbodies for which CLs have been estimated (see Figure 5-12). These waterbodies tend to be in the eastern CONUS in such ecoregions as Central Appalachian (8.4.2), the Northern Appalachian and Atlantic Maritime Highlands (5.3.1), and the Blue Ridge (8.4.2). Areas in the Rocky Mountains (6.2.10 and 6.2.14) and Sierra Nevada (6.2.12) also have been sampled extensively and include many waterbodies for which CLs have been estimated. The Northern Appalachian and Atlantic Maritime Highlands ecoregion (5.3.1) had the most waterbodies with a CL at 2,851 (see Appendix 5A, Table 5A-15).

Having more waterbodies with CLs in an ecoregion helps to capture the spatial variability of acid-sensitive areas across the landscape and provide a more accurate measurement of the impact of deposition driven acidification. In ecoregions with few waterbodies for which CLs have been developed, however, the spatial variability of acid-sensitive areas cannot be well described, which in turn limits our confidence in the representativeness of the estimated percent of exceedances for the ecoregion. For this reason, ecoregions with more than 50 CLs were the focus of this analysis.

Across the CONUS there are a total of 84 level III ecoregions, with a subset of 69 in which there is at least one waterbody with a CL estimated (Figure 5-12 and Appendix 5A, Table 5A-15). Ecoregions included in the analysis presented here are those for which there are at least 50 waterbodies with CLs and that (1) are not one of the three ecoregions identified as naturally acidic (see section 5.1.2.1 above) and (2) are not one of ecoregions that, for all of the five time periods, had no waterbodies with a CL exceedance for a CL greater than zero (based on ANC of 50 in the East and 20 in the West). There are 25 ecoregions that meet these criteria: 18 are in the east and 7 in the west.

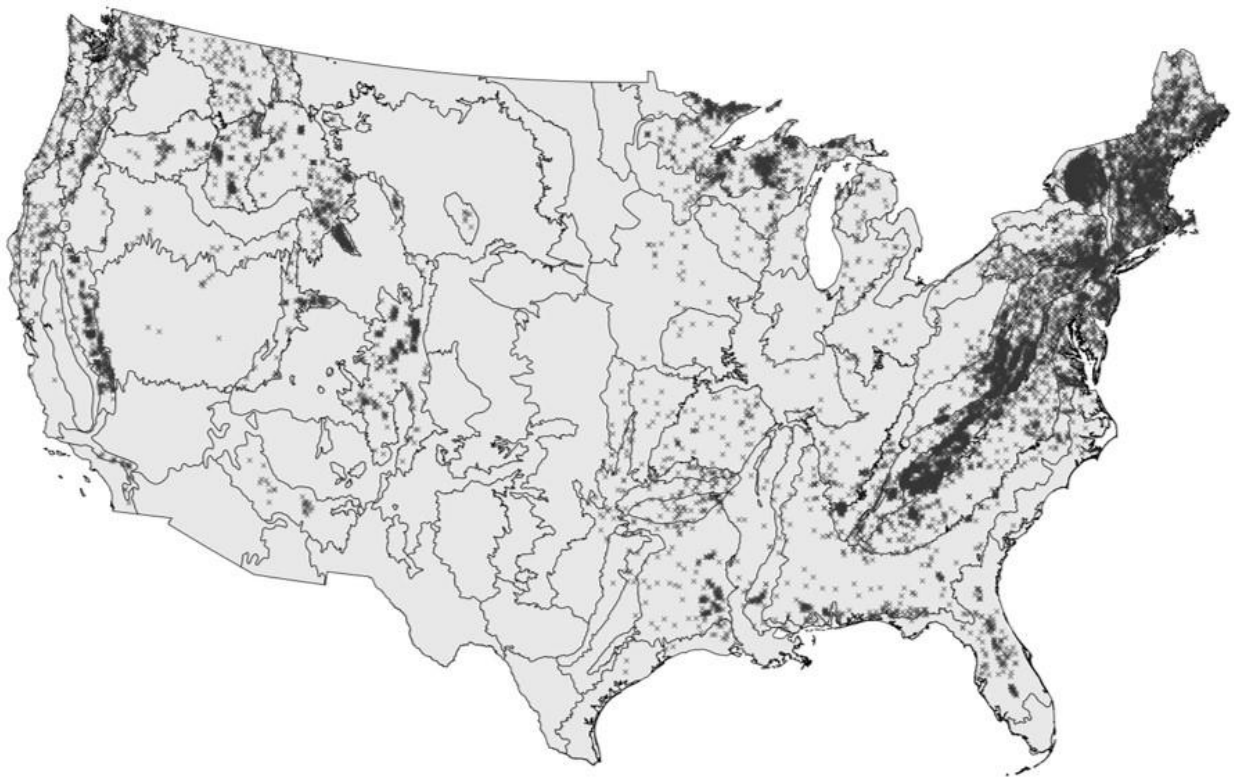

Figure 5-12. Locations of aquatic critical loads (x's) within level III ecoregion boundaries.

For each of the 25 ecoregions in this analysis, median annual average S deposition[8] declined across the five 3-year periods. The minimum to maximum range for median S deposition in these ecoregions was 0.90-18.08 kg S/ha-yr for 2001-2003 and 0.54-3.64 kg S/ha-yr for 2018 – 2020 (Table 5-2). Deposition for the 18 eastern ecoregions had a median value of 11.0 kg S/ha-yr in 2001-03 and 2.0 kg S/ha-yr in 2018-20 (Table 5-2). Deposition was lower for the seven western ecoregions, with the median of ecoregion medians ranging from 1.14 kg S/ha-yr in 2001-03 (highest median was 1.69 kg/ha-yr) to 0.71 kg S/ha-yr in 2080-20, when highest median was 1.24 kg/ha-yr. For the period 2001-2003, 17 of the 25 ecoregions had a median total S deposition over 10 kg S/ha-yr, while the highest ecoregion median in the period 2018-2020 was 3.64 kg S/ha-yr (South Central Plains ecoregion [8.3.7]) (Appendix 5A, Table 5A-11). Among the 25 ecoregions in the analysis, the ones with the highest median S deposition were the North Central Appalachians, Central Appalachians, Northern Piedmont, Southwestern Appalachians, and Ridge and Valley, all in the Mid-Atlantic region of the eastern U.S (see Appendix 5A, Table 5A-15).

[8] The ecoregion medians summarized here are spatial medians derived by GIS zonal statistic. The median was calculated across TDep grid cells, which are 4 km x 4 km, within each ecoregion.

Table 5-2. Ecoregion median S deposition estimates derived as medians of all ecoregion grid cell estimates (TDep).

	Ecoregion Median* Total Sulfur Deposition (kg S/ha-yr)				
	2001-03	**2006-08**	**2010-12**	**2014-16**	**2018-20**
	All 25 Ecoregions				
Minimum	0.90	0.98	0.83	0.79	0.54
Maximum	18.1	15.1	7.24	4.70	3.64
Median	7.34	6.78	4.04	2.61	1.68
	18 Eastern Ecoregions				
Minimum	4.29	3.24	2.38	1.65	1.22
Maximum	18.1	15.1	7.24	4.70	3.64
Median	11.0	9.04	4.53	2.99	2.04
	7 Western Ecoregions				
Minimum	0.90	0.98	0.83	0.79	0.52
Maximum	1.69	1.66	1.41	1.51	1.24
Median	1.14	1.16	1.10	0.93	0.71

* The ecoregion medians for which descriptive statistics are presented are the medians of the 4 x 4 km TDep grid cells within each ecoregion. The number of grid cells varies across ecoregions based on the size of the ecoregion.

Ecoregion median S deposition was also derived based on the TDep grid cells for locations with a CL estimate in each ecoregion. Descriptive statistics for these ecoregion medians are summarized in Table 5-3 below. For each of the 25 ecoregions, Figure 5-13 presents the temporal trend in percentage of waterbody sites at which the TDep grid cell S deposition estimates exceeded the CL estimates (Appendix 5A, section 5A.2.2.1).

Table 5-3. Summary of ecoregion medians derived as median of TDep S deposition estimates at CL sites within each ecoregion.

	Ecoregion Median* Total Sulfur Deposition (kg S/ha-yr)				
	2001-03	**2006-08**	**2010-12**	**2014-16**	**2018-20**
	All 25 Ecoregions				
Minimum	1.18	1.22	1.02	1.08	0.62
Maximum	17.27	14.44	7.25	4.58	3.88
Median	7.77	6.50	3.71	2.32	1.73
	18 Eastern Ecoregions				
Minimum	4.01	3.10	2.34	1.88	1.31
Maximum	17.27	14.44	7.25	4.58	3.88
Median	11.08	9.36	4.76	2.97	2.04
	7 Western Ecoregions				
Minimum	1.18	1.22	1.02	1.08	0.62
Maximum	1.94	1.83	1.47	1.56	1.19
Median	1.40	1.52	1.29	1.17	0.87

* The ecoregion medians for which descriptive statistics are presented here are medians of TDep estimates across each ecoregion's waterbody sites with CL estimates.

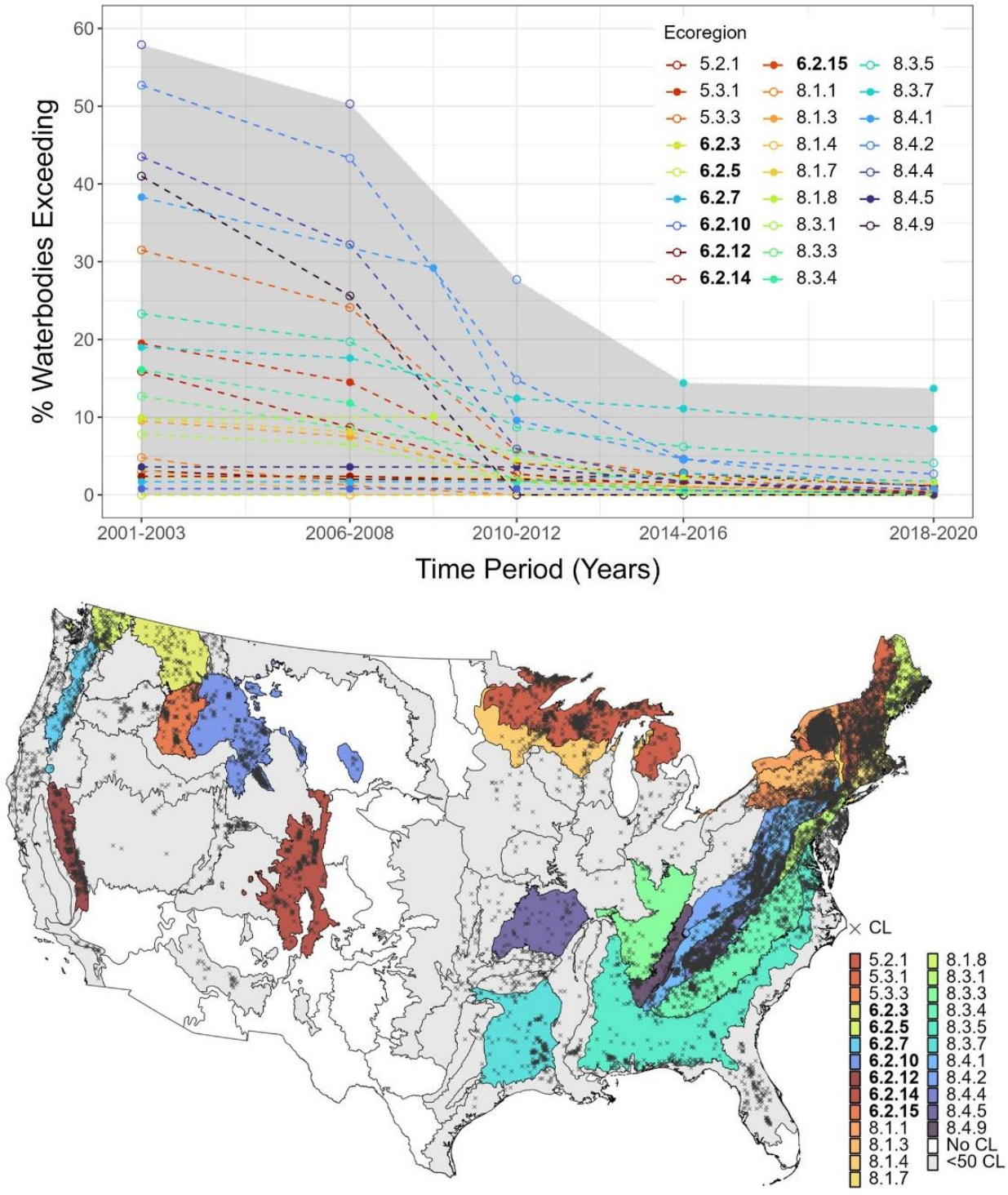

Figure 5-13. Percentage of waterbodies exceeding their CLs per ecoregion for ANC of 20 µeq/L, with shading indicating the maximum ecoregion percentage exceeding CLs for ANC of 50 µeq/L (upper panel). Symbols on the upper line of the grey shaded area indicate the ecoregion with this maximum. Ecoregion locations are shown on map (lower panel), with bold indicating those designated as "West" (N=7) and regular font indicating eastern ecoregions (N=18).

We summarize below the CL exceedance results for the 25 ecoregions analyzed, in terms of number and percentage of waterbodies per ecoregion with CL exceedances in every ecoregion-time period combination, using ecoregion deposition estimates (medians of deposition estimates at waterbodies with CLs in each ecoregion) as the organizing parameter. For example, Table 5-4 presents the CL exceedance results of the ecoregion level analyses for the three ANC target levels, summarized by bins for different magnitudes of ecoregion median annual average S deposition (regardless of the 3-year period in which it occurred). For each S deposition bin (e.g., S deposition at or below 5 kg S/ha-yr), Table 5-4 presents the number of ecoregion-time period combinations with more than 10, 15, 20, 25 and 30% of waterbodies exceeding their CL for the specified ANC target level.

For example, among the eastern and western ecoregion-time period combinations with S deposition at or below 2 kg S/ha-yr across ecoregions and deposition periods, there are no ecoregions that have more than 10% of their waterbodies exceeding their CLs for any of the three ANC targets (Table 5-4**)**. In contrast, for annual average S deposition at or below 10 kg S/ha-yr, there are 22 of the 90 eastern ecoregion-time period combinations with more than 10% of their waterbodies exceeding their CLs for an ANC of 50 μeq/L, one of which had more than 30% of its waterbodies exceeding their CLs. The lowest annual average S deposition level associated with any ecoregion-time period combinations having more than 30% of waterbodies exceeding their CLs was 10 kg S/ha-yr, for which one ecoregion in one time period had more than 30% of the waterbodies exceeding their CLs for all three ANC targets.

Table 5-4. Number of ecoregion-time period combinations with more than 10, 15, 20, 25, and 30% of waterbodies exceeding their CLs for three ANC targets as a function of ecoregion-level estimates of annual average S deposition.

S Deposition (kg/ha-yr):	No. of Eastern Ecoregion-Time Periods	Number of eastern ecoregion-time periods with more than specified percent of waterbodies exceeding their CLs															Number of western ecoregion-time periods with more than 10% of waterbodies exceeding their CLs for ANC target of 20, 30 or 50 µeq/L		
		10%	15%	20%	25%	30%	10%	15%	20%	25%	30%	10%	15%	20%	25%	30%	S Deposition (kg/ha-yr)	No. ecoregion -time periods	>10%
		ANC target of 20 µeq/L					*ANC target of 30 µeq/L*					*ANC target of 50 µeq/L*							
≤2	10	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	≤2	35	0
≤3	29	0	0	0	0	0	0	0	0	0	0	1	0	0	0	0			
<4	41	0	0	0	0	0	2	0	0	0	0	3	1	0	0	0			
≤5	51	2	1	0	0	0	4	1	0	0	0	9	3	2	1	0	None of the 35 western ecoregion-time periods (7 ecoregions and 5 time periods) in analysis had ecoregion S deposition estimates above 2 kg S/ha-yr		
≤6	59	4	1	0	0	0	7	1	0	0	0	13	4	2	1	0			
≤7	63	5	1	0	0	0	8	2	0	0	0	14	5	3	1	0			
≤8	67	9	4	0	0	0	12	6	1	0	0	18	9	5	3	0			
≤9	69	9	4	0	0	0	13	6	1	0	0	19	9	5	3	0			
≤10	73	11	6	1	1	1	16	8	2	1	1	22	11	6	4	1			
≤11	76	13	7	2	1	1	18	9	3	1	1	24	13	7	4	1			
≤12	79	15	9	4	3	2	21	11	5	3	3	27	15	9	6	3			
≤13	81	16	10	4	3	2	22	12	5	3	3	28	16	10	6	3			
≤14	84	19	12	6	4	3	25	14	7	5	4	31	18	12	8	5			
≤15	86	21	14	8	6	4	27	16	9	7	6	33	20	14	10	7			
≤16	88	22	15	9	7	5	28	17	10	8	7	34	21	15	11	8			
≤17	88	22	15	9	7	5	28	17	10	8	7	34	21	15	11	8			
≤18	90	24	17	11	9	7	30	19	12	10	9	36	23	17	13	10			

As none of the 7 western ecoregions had more than 10% of their waterbodies exceeding their CLs for any of the ANC thresholds in any of the five time periods, we focus the remaining presentations on the eastern ecoregions. We considered these ecoregion-scale results from the perspective of the extent to which waterbodies within the eastern ecoregions were estimated to achieve the various ANC targets across the S deposition levels for the 18 ecoregions and five time periods. This can be considered the inverse of the presentation in Table 5-4 above, using percentages instead of absolute counts in the presentation. For example, rather than the number of ecoregion-time periods, with a particular range of S deposition estimates, that have more than 10% of waterbodies exceeding their CLs for an ANC target of 20 μeq/L, Figure 5-14 presents the percentage of ecoregion-time periods that have less than or equal to 10% (or 15, 20, 25 or 30%) of waterbodies exceeding their CLs for each of the three ANC levels (20, 30 and 50 μeq/L). The same dataset is presented in Table 5-5, but in terms of percentage of waterbodies that are not exceeding their CLs (i.e., that are estimated to achieve the ANC target). Results also presented in Appendix 5A, section 5A.2.2.

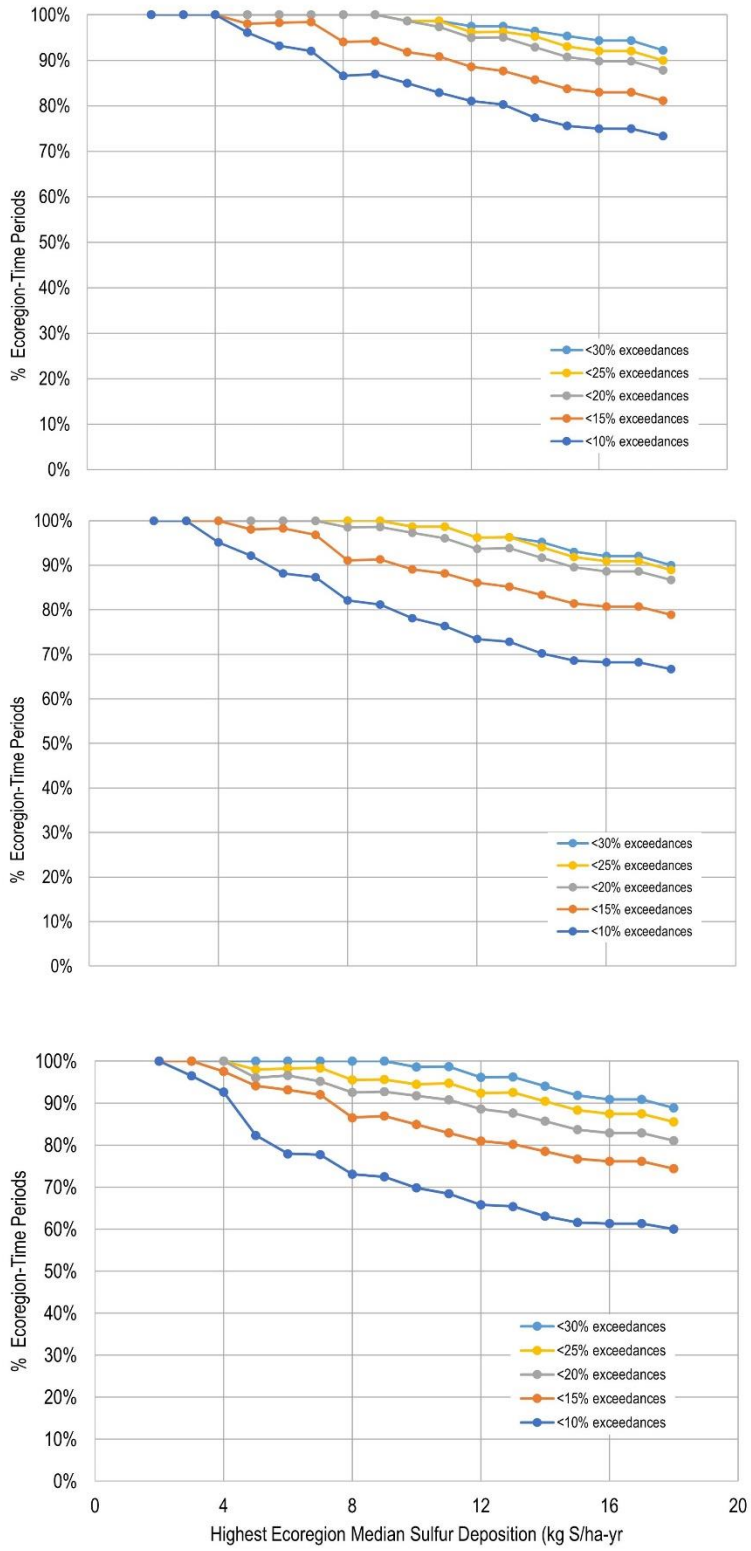

Figure 5-14. Percentage of ecoregion-time period combinations with less than or equal to 10, 15, 20, 25 and 30% of waterbodies exceeding their CLs for ANC of 20 (top), 30 (middle) and 50 µeq/L (bottom) for 18 eastern ecoregions.

Table 5-5. Percentage of ecoregion-time periods combinations with at least 90, 85, 80, 75 and 70% of waterbodies estimated to achieve an ANC at/above the ANC targets of 20, 30 and 50 µeq/L as a function of annual average S deposition for 18 eastern ecoregions (90 ecoregion-time period combinations).

Total Sulfur Deposition (kg S/ha-yr) at/below:	No. of Ecoregion-Time Periods	% Waterbodies per ecoregion-time period meeting specified ANC target														
		90%	85%	80%	75%	70%	90%	85%	80%	75%	70%	90%	85%	80%	75%	70%
		ANC target of 20 µeq/L					*ANC target of 30 µeq/L*					*ANC target of 50 µeq/L*				
2	10	100%	100%	100%	100%	100%	100%	100%	100%	100%	100%	100%	100%	100%	100%	100%
3	29	100%	100%	100%	100%	100%	100%	100%	100%	100%	100%	97%	100%	100%	100%	100%
4	41	100%	100%	100%	100%	100%	95%	100%	100%	100%	100%	93%	98%	100%	100%	100%
5	51	96%	98%	100%	100%	100%	92%	98%	100%	100%	100%	82%	94%	96%	98%	100%
6	59	93%	98%	100%	100%	100%	88%	98%	100%	100%	100%	78%	93%	97%	98%	100%
7	63	92%	98%	100%	100%	100%	87%	97%	100%	100%	100%	78%	92%	95%	98%	100%
8	67	87%	94%	100%	100%	100%	82%	91%	99%	100%	100%	73%	87%	93%	96%	100%
9	69	87%	94%	100%	100%	100%	81%	91%	99%	100%	100%	72%	87%	93%	96%	100%
10	73	85%	92%	99%	99%	99%	78%	89%	97%	99%	99%	70%	85%	92%	95%	99%
11	76	83%	91%	97%	99%	99%	76%	88%	96%	99%	99%	68%	83%	91%	95%	99%
12	79	81%	89%	95%	96%	97%	73%	86%	94%	96%	96%	66%	81%	89%	92%	96%
13	81	80%	88%	95%	96%	98%	73%	85%	94%	96%	96%	65%	80%	88%	93%	96%
14	84	77%	86%	93%	95%	96%	70%	83%	92%	94%	95%	63%	79%	86%	90%	94%
15	86	76%	84%	91%	93%	95%	69%	81%	90%	92%	93%	62%	77%	84%	88%	92%
16	88	75%	83%	90%	92%	94%	68%	81%	89%	91%	92%	61%	76%	83%	88%	91%
17	88	75%	83%	90%	92%	94%	68%	81%	89%	91%	92%	61%	76%	83%	88%	91%
18	90	73%	81%	88%	90%	92%	67%	79%	87%	89%	90%	60%	74%	81%	86%	89%

Overall, the S deposition levels in the 18 eastern ecoregions and five time periods analyzed include a range from below 2 up to nearly 18 kg/ha-yr. Across all 90 eastern ecoregion-time period combinations (including S deposition estimates up to near 18 kg/ha-yr), 73% of the combinations had at least 90% of waterbodies per ecoregion estimated to achieve ANC at or above 20 µeq/L, and 60% had at least 90% of the waterbodies estimated to achieve ANC at or above 50 µeq/L. Less than half of the eastern ecoregion-time period combinations (and all of the western combinations) had an S deposition estimate below 4 kg/ha-yr. Ninety percent of the eastern combinations were at or below 13 kg/ha-yr. For the 75 western-time period combinations, all of which had an S deposition estimate below 4 kg/ha-yr, at least 90% of waterbodies per ecoregion were estimated to achieve an ANC at or above 50 µg/L The results by annual average S deposition bin are summarized below for the bins from 13 kg/ha-yr down to 5 kg/ha-yr (the bin that includes at least half of this dataset):

- For S deposition estimates at or below 13 kg/ha-yr, at least 90% of waterbodies per ecoregion were estimated to achieve an ANC at or above 20, 30 and 50 µeq/L in 80%, 73% and 65% of all ecoregion-time period combinations, respectively.
- For S deposition at or below 11 kg/ha-yr, at least 90% of all waterbodies per ecoregion were estimated to achieve ANC at or above 20, 30 and 50 µeq/L in 83%, 77% and 68% of all ecoregion-time period combinations, respectively.
- For S deposition at or below 9 kg/ha-yr, at least 90% of all waterbodies per ecoregion were estimated to achieve ANC at or above 20, 30 and 50 µeq/L in 87%, 81% and 72% of combinations, respectively.
    - At least 80%, 75% and 70% of waterbodies per ecoregion were estimated to achieve ANC at or above 20, 30 and 50 µeq/L, respectively, in all ecoregion-time period combinations.
- For S deposition at or below 7 kg/ha-yr, at least 90% of waterbodies per ecoregion were estimated to achieve ANC at or above 20, 30 and 50 µeq/L in 92, 87 and 78% of combinations, respectively.
    - At least 80%, 80% and 70% of waterbodies per ecoregion were estimated to achieve ANC at or above 20, 30 and 50 µeq/L, respectively, in all ecoregion-time period combinations.
- For S deposition at or below 5 kg/ha-yr, at least 90% of all waterbodies per region were estimated to achieve ANC at or above 20, 30 and 50 µeq/L in 96%, 92% and 82% of combinations, respectively.
    - At least 80%, 80% and 70% of waterbodies per ecoregion were estimated to achieve ANC at or above 20, 30 and 50 µeq/L, respectively, in all ecoregion-time period combinations.
- For S deposition at or below 4 kg/ha-yr, at least 90% of all waterbodies per region were estimated to achieve ANC at or above 20 in all 41 ecoregion-time period combinations for that deposition bin, and to achieve ANC at or above 30 and 50 µeq/L in 95% and 97%

of those combinations, respectively. The number of ecoregion-time period combinations in this deposition bin is less than half the full dataset for the 18 eastern ecoregions.

To further describe the results for recent conditions, we looked at S deposition for the 25 ecoregions in the two most recent time periods, 2014-2016 and 2018-2020, and the critical load exceedances for the three ANC targets (Figures 5-15 and 5-16). Only one ecoregion had more than 10% of its waterbodies exceeding a CL for any target ANC values in either time period. This was the South-Central Plains ecoregion (8.3.7), which covers portions of eastern Texas, western Louisiana and southwestern Arkansas, an area dominated by pine forest (which tend to be in acidic soils). The median of the 18 eastern ecoregion median S deposition values for the 2014-2016 time period was 3.0 kg/ha-yr, dropping to 2.0 kg/ha-yr in the 2018 2020 time period.

Figure 5-17 through 5-19 show the eastern ecoregions with exceedances of target critical loads under the two most recent time periods. Figure 5-20 shows the ecoregions with exceedances for the entire U.S. for the most recent time periods using an ANC target of 50 µeq/L for the east and 20 µeq/L for the west.

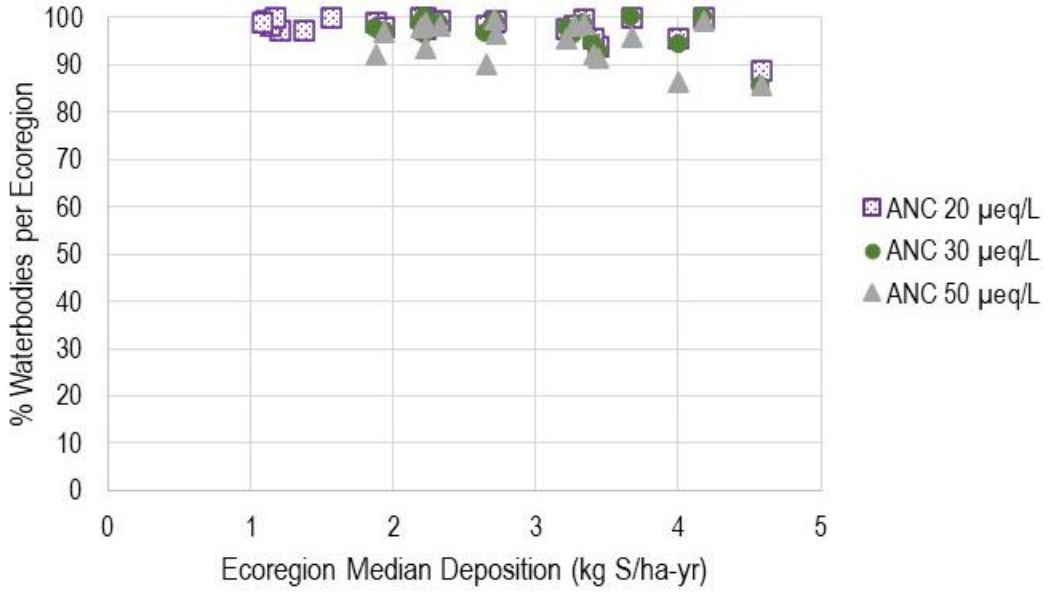

Figure 5-15. Percentage of waterbodies in each of the 25 ecoregions estimated to achieve ANC values of 20 (E&W), 30 (E only) and 50 (E only) µeq/L as a function of ecoregion annual average S deposition for 2014-2016 (median across CL sites).

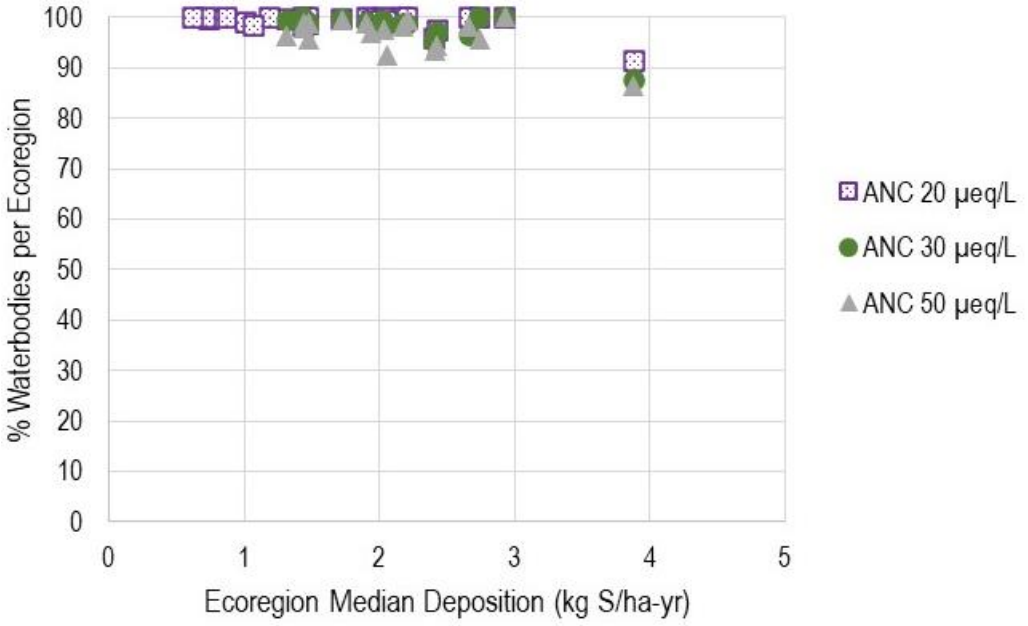

Figure 5-16. Percent of waterbodies in each of the 25 ecoregions estimated to achieve ANC values of 20 (E&W), 30 (E only) and 50 (E only) µeq/L as a function of ecoregion annual average S deposition for 2018-2020 (median across CL sites).

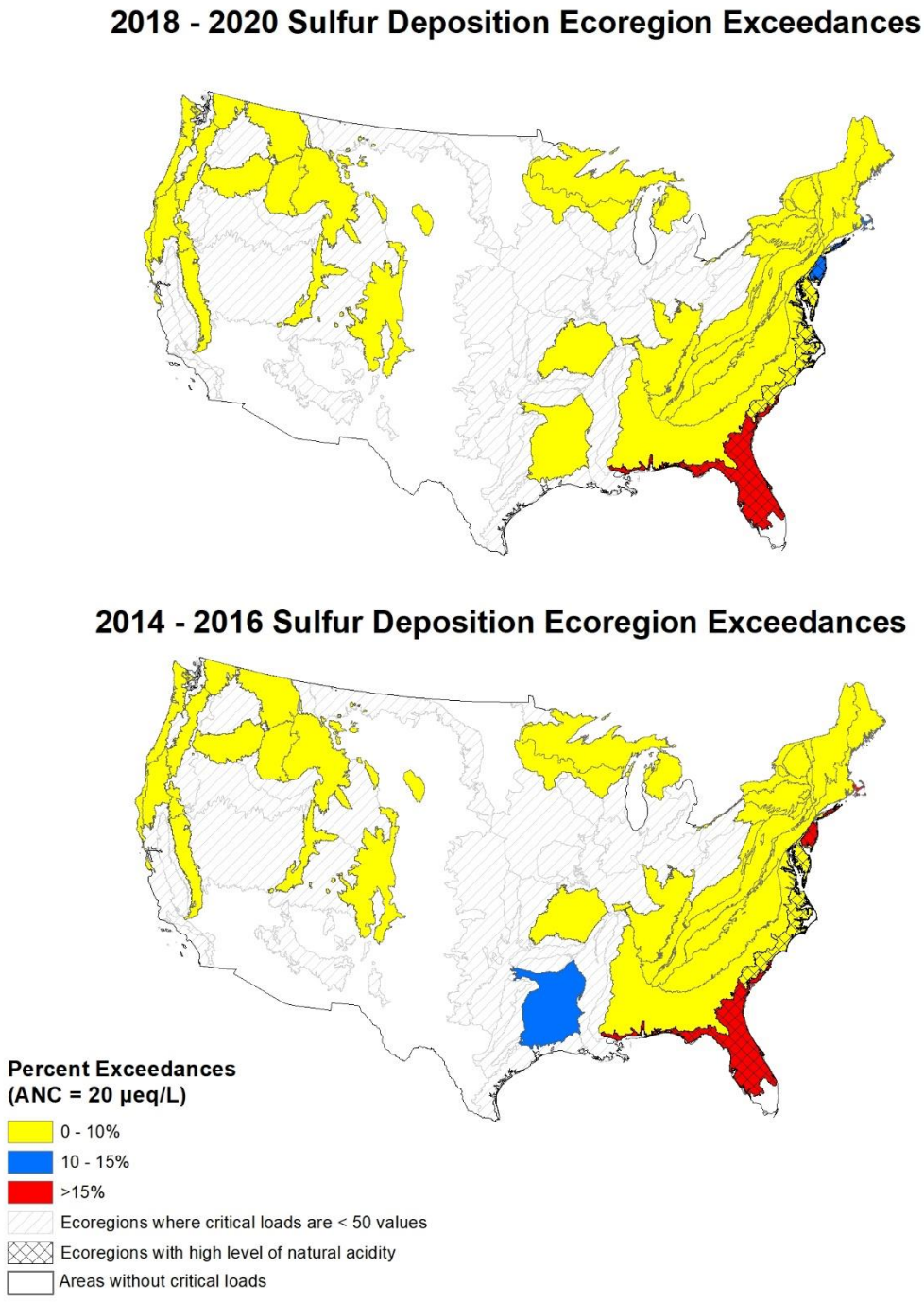

Figure 5-17. Map of critical load exceedances for S only deposition from 2018-20 (top) and 2014-16 (bottom) for ANC threshold of 20 µeq/L.

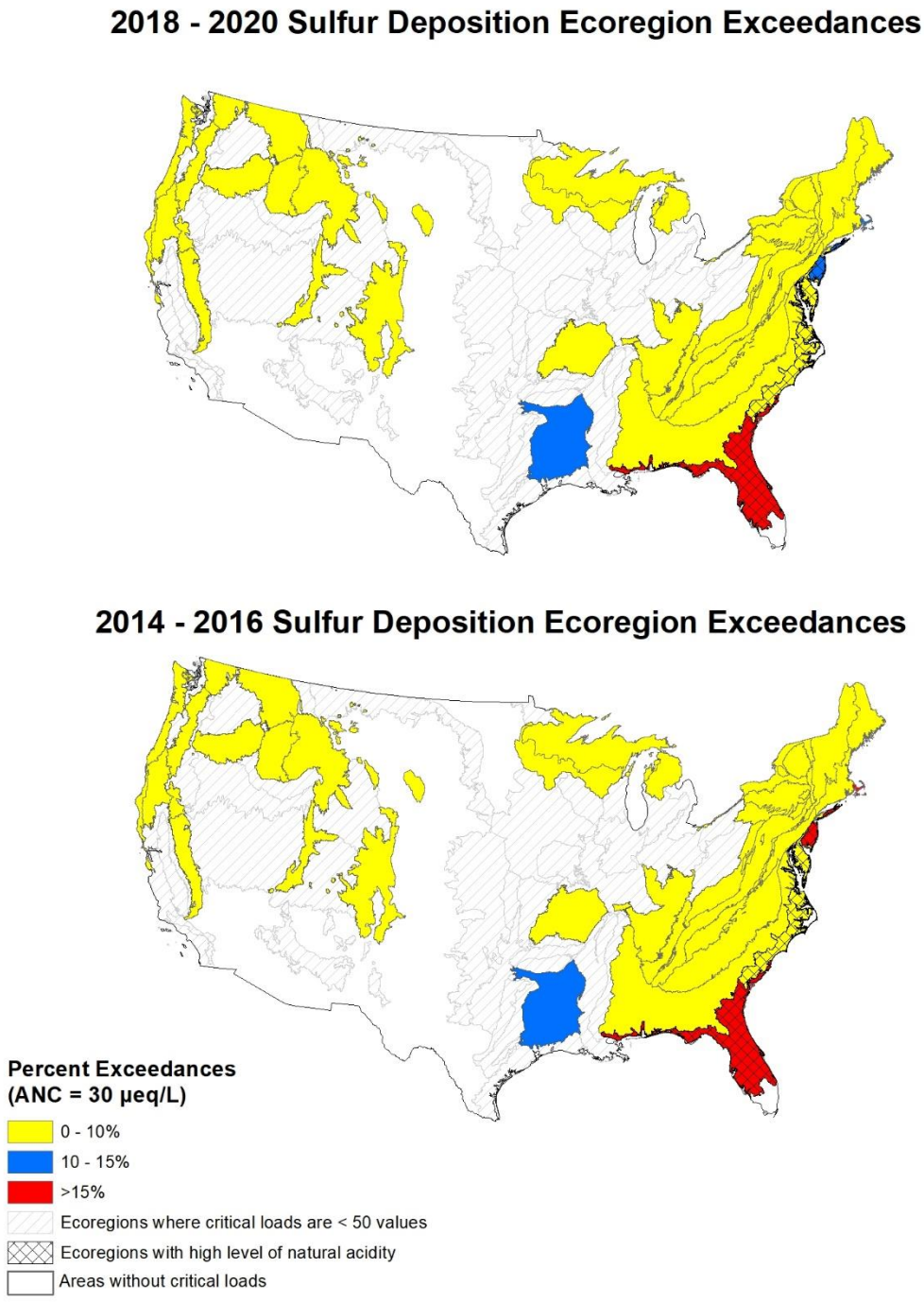

Figure 5-18. Map of critical load exceedances for S only deposition from 2018-20 (top) and 2014-16 (bottom) for an ANC threshold of 30 µeq/L.

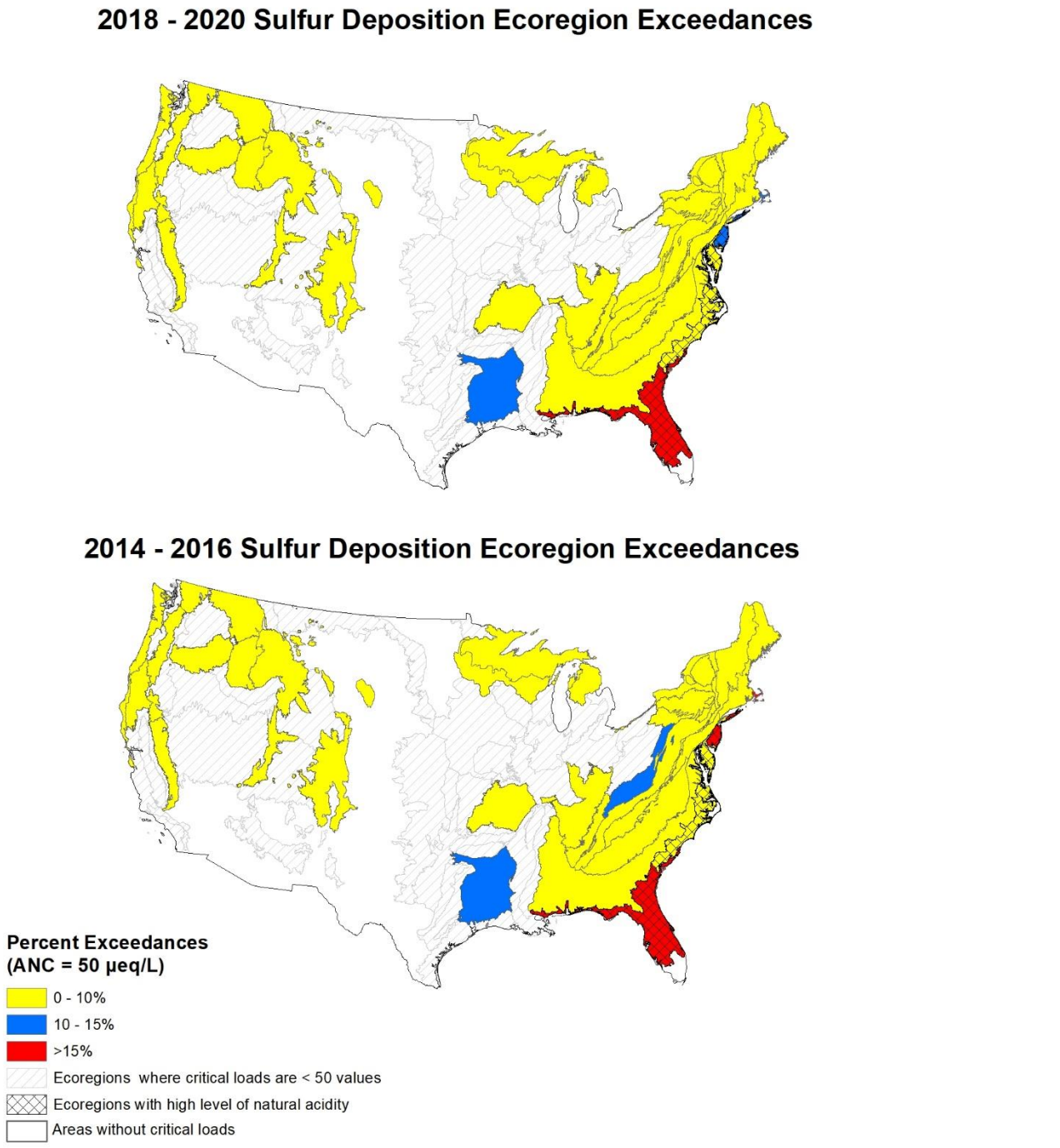

Figure 5-19. Map of critical load exceedances for S only deposition from 2018-20 (top) and 2014-16 (bottom) for an ANC threshold of 50 µeq/L.

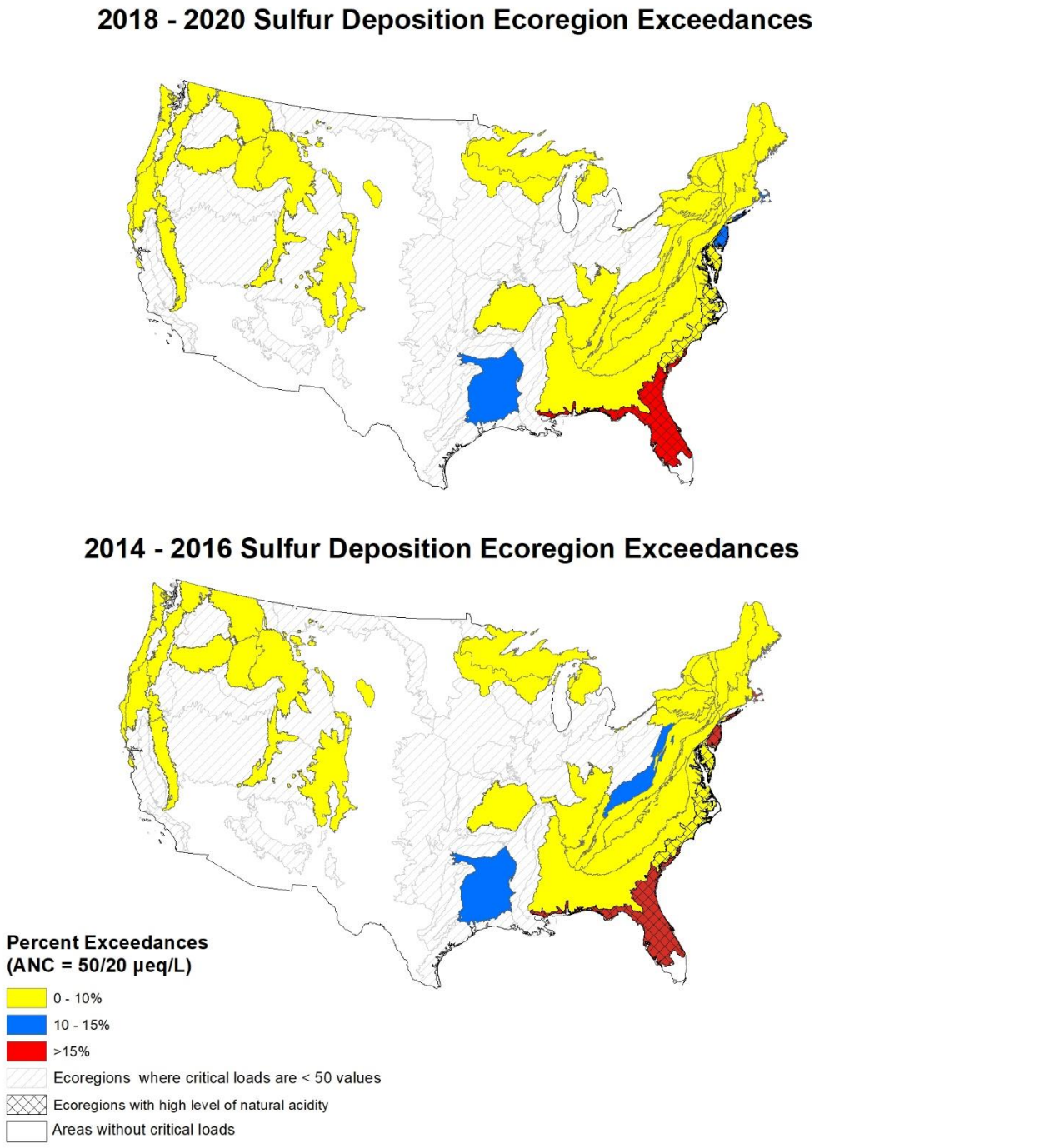

Figure 5-20. Map of critical load exceedances for S only deposition from 2018-20 (top) and 2014-16 (bottom) for an ANC threshold of 50 µeq/L for East and 20 µeq/L for the West.

5.1.3.3 Case Study Analyses

The case study areas are geographically diverse acid-sensitive areas across the CONUS that have sufficient data to complete the quantitative analyses. Five case study areas were identified that meet the criteria (Figure 5-21): White Mountain National Forest (WHMT), Shenandoah Valley Area (SHVA), Northern Minnesota (NOMN), Rocky Mountain National Park (ROMO) and Sierra Nevada Mountains (SINE). Three of these areas are in the eastern U.S. (NOMN, SHVA and WHMT) and two areas are in the western U.S. (ROMO and SINE). Class I areas occur in three of the five case study areas (SHVA, ROMO and SINE). Additional aquatic acidification analyses using the case studies can be found in Appendix 5A. A total of 523 CLs were identified in four of the five case study areas, while the SHVA case study had complete coverage, with 4977 CLs. The case studies, ROMO, SINE, NOMN, and WHMT, had 119, 139, 190, and 75 CLs, respectively**.** For this discussion, the analyses identified the calculated sulfur deposition values at or below which the case study sites would likely be able to attain the target ANC values of 50, 30 and 20 µeq/L for the eastern case studies and 20 µeq/L for the western case studies.

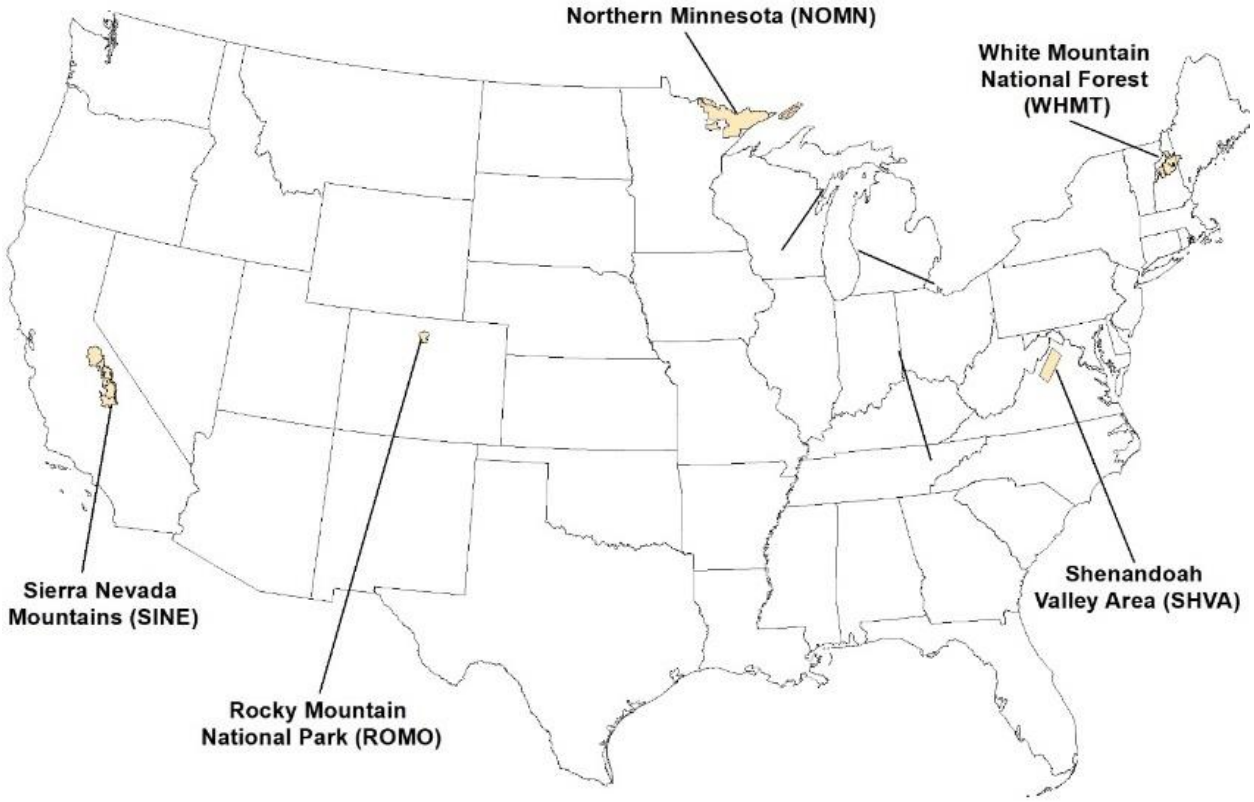

Figure 5-21. Location of the five case study areas.

The steady-state mass balance modeling results summarized in Table 5-6 indicate the average CL for achieving a target ANC of 20 µeq/L in the five study areas ranges from about 10

to 12 kg/ha-yr. For 70 to 90% of sites to achieve an ANC of 20 µeq/L, the estimated CL for S deposition ranges from about 4 to 9 kg/ha-yr. The average CL to achieve an ANC value of 30 µeq/L ranges from about 10 to 11 kg/ha-yr and for 70-90% of sites to achieve an ANC of 30 µeq/L, the estimated CL for S deposition ranges from about 3 to 8 kg/ha-yr. For an ANC target of 50 µeq/L, the average CL for sites in the five case studies ranges from about 7 to 10 kg/ha-yr. For 70 to 90% of the case study sites to achieve a target ANC of 50 µeq/L, the estimated CL for S deposition ranges between 3 to 4kg/ha-yr, except for White Mountain, which is extremely sensitive. Overall, these findings are slightly lower than the ecoregion-scale results.

Table 5-6. Annual average S deposition at/below which modeling indicates an ANC of 20, 30 or 50 µeq/L can be achieved in the average, 70% and 90% of waterbodies in each study area.

ANC (µeq/L)	Based on average across all sites in area					Based on 70% of sites achieving					Based on 90% of sites achieving				
	------- Eastern -------			--- Western ---		------- Eastern -------			--- Western ---		------- Eastern -------			--- Western ---	
	N. Minn	White Mtns	Shenan-doah	Rocky Mtn NP	Sierra Nev Mtns	N. Minn	White Mtns	Shenan-doah	Rocky Mtn NP	Sierra Nev Mtns	N. Minn	White Mtns	Shenan-doah	Rocky Mtn NP	Sierra Nev Mtns
	(kg/ha-yr)	(kg/ha-yr)	(kg/ha-yr)	(kg/ha-yr)	(kg/ha-yr)	(kg/ha-yr)	(kg/ha-yr)	(kg/ha-yr)	(kg/ha-yr)	(kg/ha-yr)	(kg/ha-yr)	(kg/ha-yr)	(kg/ha-yr)	(kg/ha-yr)	(kg/ha-yr)
20	11	11	12	9.5	12	5.5	6.9	9.4	5.4	4.1	4.2	4.4	7.1	3.6	1.8
30	10	10	11			5.3	6.1	8.4			3.9	3.3	6.3		
50	10	10	9.4			4.7	4.1	6.3			3.2	0.7	4.1		

Note: Shaded boxes indicate that consistent with convention followed in the ecoregion analysis above, CLs are not presented for ANC target values of 30 and 50 µg/L in the West.

5.1.4 Characterization of Uncertainty

We have characterized the nature and magnitude of associated uncertainties and their impact on the REA estimates based primarily on a mainly qualitative approach, informed by several quantitative sensitive analyses, all of which are described in Appendix 5A, section 5A.3. The mainly qualitative approach used here and in quantitative analyses in other NAAQS reviews is described by WHO (2008). Briefly, with this approach, we have identified key aspects of the assessment approach that may contribute to uncertainty in the conclusions and provided the rationale for their inclusion. Then, we characterized the magnitude and direction of the influence on the assessment for each of these identified sources of uncertainty. Consistent with the WHO (2008) guidance, we scaled the overall impact of the uncertainty by considering the degree of uncertainty as implied by the relationship between the source of uncertainty and the exposure and risk estimates. A qualitative characterization of low, moderate, and high was assigned to the magnitude of influence and knowledge base uncertainty descriptors, using quantitative observations relating to understanding the uncertainty, where possible. Where the magnitude of uncertainty was rated low, it was judged that large changes within the source of uncertainty

would have only a small effect on the assessment results (e.g., an impact of few percentage points upwards to a factor of two). A designation of medium implies that a change within the source of uncertainty would likely have a moderate (or proportional) effect on the results (e.g., a factor of two or more). A characterization of high implies that a change in the source would have a large effect on results (e.g., an order of magnitude). We also included the direction of influence, whether the source of uncertainty was judged to potentially over-estimate ("over"), under-estimate ("under"), or have an unknown impact to exposure/risk estimates.

A summary of the overall uncertainty characterization is provided in Appendix 5A, Table 5A-53. Two types of quantitative analyses that informed our understanding of the variability and uncertainty associated with the CL estimates developed in this assessment and support the uncertainty characterization are also presented in Appendix 5A, in sections 5A.1.1 and 5A.1.2. The first type of analysis is a sensitivity analysis using Monte Carlo techniques to quantify CL estimate uncertainty associated with several model inputs, and the second is an analysis of the variation in CL estimates among the three primary modeling approaches on which the CLs used in this assessment were based.

As overarching observations regarding uncertainty associated with this REA, we take note of two overarching aspects of the assessment. The first relates to interpretation of specific thresholds of ANC and the second to our understanding of the biogeochemical linkages between deposition of S and N compounds and waterbody ANC (implemented in modeling used in this assessment), and the associated estimation of CLs. While ANC is an established indicator of aquatic acidification risk, there is uncertainty in our understanding of relationships between ANC and risk to native biota, particularly in waterbodies in geologic regions prone to waterbody acidity. Such uncertainties relate to the varying influences of site-specific factors other than ANC. Uncertainty associated with our understanding of the biogeochemical linkages between deposition and ANC and the determination of steady-state CLs is difficult to characterize and assess. Uncertainty in CL estimates is associated with parameters used in the steady-state CL models. While the SSWS and other CL models are well conceived and based on a substantial amount of research and applications available in the peer-reviewed literature, there is uncertainty associated with the availability of the necessary data to support certain model components.

The strength of the CL estimates and the exceedance calculation rely on the ability of models to estimate the catchment-average base-cation supply (i.e., input of base cations from weathering of bedrock and soils and air), runoff, and surface water chemistry. Key parameters in this modeling include estimates of the catchment-average base-cation supply (i.e., input of base cations from weathering of bedrock and soils and air), runoff, and surface water chemistry. The uncertainty associated with runoff and surface water parameters relates to availability of measurements; however, the ability to accurately estimate the catchment supply of base cations

to a water body is still difficult, and uncertain (Appendix 5A, section 5A.3). This area of uncertainty is important because the catchment supply of base cations from the weathering of bedrock and soils is the factor with the greatest influence on the CL calculation and has the largest uncertainty (Li and McNulty, 2007). For example, the well-established models generally rely on input or simulated values for base cation weathering (BCw) rate, a parameter the ISA notes to be "one of the most influential yet difficult to estimate parameters in the calculation of critical acid loads of N and S deposition for protection against terrestrial acidification" (ISA, section IS.14.2.2.1). Obtaining accurate estimates of weathering rates is difficult because weathering is a process that occurs over very long periods of time, and the estimates on an ecosystem's ability to buffer acid deposition rely on accurate estimates of weathering. Although the approach to estimate base-cation supply for the national case study (e.g., F-factor approach) has been widely published and analyzed in Canada and Europe, and has been applied in the U.S. (e.g., Dupont et al., 2005 and others), the uncertainty in this estimate is unclear and could be large in some cases.

In light of the significant contribution of this input to the CL estimates, a quantitative uncertainty analysis of CL estimates based on state-steady CL modeling was performed (Appendix 5A, section 5A.3.1). This analysis, involving many model simulations for the more than 14,000 waterbodies, drawing on Monte Carlo sampling, provided a description of the uncertainty around the CL estimate in terms of the confidence interval for each waterbody mean result. The size of the confidence interval ranged from 0.37 meq/m^2-yr at the 5^{th} percentile to 33.2 meq/m^2-yr at the 95^{th} percentile. Lower confidence intervals were associated with CLs determined with long-term water quality data and low variability in runoff measurements. Estimates of CL determined by one or very few water quality measurements, and in areas where runoff is quite variable (e.g., the western U.S.) had larger confidence intervals, indicating greater uncertainty. Critical load estimates with the lowest uncertainty were for waterbody sites in the eastern U.S., particularly along the Appalachian Mountains, in the Upper Midwest, and in the Rocky Mountains. Greater uncertainty is associated with CLs in the Midwest and South and along the CA to WA coast. This uncertainty in the Midwest is associated with most of the CLs in waterbodies in this area being based on one or a few water quality measurements, while the high uncertainty for sites along the CA and WA coasts relates to variability in runoff values. On average, the size of the confidence interval for all SSWC CLs was 7.68 meq S/m^2-yr or 1.3 kg S/ha-yr, giving a confidence level of ±3.84 meq/m^2-yr or ±0.65 kg S/ha-yr. While a comprehensive analysis of uncertainty has not been completed for these estimates prior to this REA, expert judgment suggested the uncertainty for combined N and S CLs to be on average about ±0.5 kg/ha-yr (3.125 meq/m^2-yr), which is generally consistent with the range of determined from this quantitative uncertainty analysis.

At the ecoregion scale, fifty-one ecoregions had sufficient data to calculate the 5th to 95th percentile (Appendix 5A, Table 5A-56). Smaller confidence intervals around the mean CL (i.e., lower uncertainty CLs) were associated with ecoregions in the Appalachian Mountains (e.g., Northern Appalachian and Atlantic Maritime Highlands (5.3.1), Blue Ridge (8.4.4), Northern Lakes and Forests (5.2.1), and North Central Appalachians (5.3.3) and Rockies (e.g. Sierra Nevada (6.2.14), Southern Rockies (6.2.14), and Idaho Batholith (6.2.15). Ecoregions with more uncertain CLs included the Northeastern Coastal Zone (8.1.7), Cascades (6.2.7), Coast Range (7.1.8), Interior Plateau (8.3.3), and Klamath Mountains/California High North Coast Range (6.2.11).

Although the vast majority of CLs in this assessment were based on the SSWC model, an analysis was conducted to understand differences in the CLs calculated with the different methods. There are three main CL approaches all based on the watershed mass-balance approach where acid-base inputs are balanced. The three approaches include: (1) SSWC model and F-Factor that is based on quantitative relationships to water chemistry (Dupont et al., 2005; Scheffe et al., 2014; Lynch et al., 2022), (2) Statistical Regression Model that extrapolated weathering rates across the landscape using water quality or landscape factors (Sullivan et al., 2012a; McDonnell et al., 2014), and (3) Dynamic Models (MAGIC or Pnet-BGC). Critical load values were compared between these models to determine model biases. Results from the comparison between different CL methods that were used to calculate the critical loads in the NCLD are summarized in Appendix 5A, section 5A.3.3, for lakes in New England and the Adirondacks and streams in the Appalachian Mountains. Overall, good agreement was found between the three methods used to calculate CLs, indicating there was not a systematic bias between the methods and that they should produce comparable results when used together as they were in these analyses.

5.1.5 Summary of Key Findings

Quantitative analyses were performed to assess acidification risks of S deposition in waterbodies across the U.S. using a critical load approach. Due to the finding of a negligible influence of N deposition on acidification under the S deposition levels in this assessment, we focused on S deposition solely (Appendix 5A, section 5A.2.1). In this assessment, ANC was used as the water quality indicator of acidification, based on its longstanding use for this purpose (ISA, Appendix 7, section 7.1.2.6). We also focused on acid-deposition-sensitive areas for which the available CL modeling estimates indicated that the target ANC values of 50, 30 and 20 µg/L could be reached. Analyses were performed at three different spatial scales: nationwide, ecoregion (level III), and case studies.

Critical load estimates for specific waterbody sites across the contiguous U.S. were drawn from the NCLD (version 3.2.1) for comparison to total deposition estimates in the same locations from TDep for five time periods since 2000. Comparisons were only performed for critical load estimates greater than zero. The results of these analyses are summarized with regard to spatial extent and severity of deposition-related acidification effects and the protection from these effects associated with a range of annual S deposition.

Between the three-year period 2000-2002, which was the analysis year for the 2009 REA, and 2018-2020, the latest period considered in the present analyses, national average sulfur deposition has declined by 68% across the U.S. This decline in deposition is reflected in the very different aquatic acidification impact estimates for the two periods. Unlike the findings for 2000-2002 in the last review (concluded in 2012), few waterbody sites are estimated to be receiving deposition in excess of their critical loads for relevant ANC targets under recent deposition levels. While recognizing inherent limitations and associated uncertainties of any such analysis, the national-scale assessment performed as part of this review, indicates that under deposition scenarios for the 2018-2020 time period, the percentage of waterbodies nationwide that might not be able to maintain an ANC of 50 µg/L in the east and an ANC of 20 µg/L in the west would be less than 5% (see Table 5-1).

The ecoregion-level analyses of ANC levels and deposition estimates for the five periods from 2001-2003 through 2018 -2020 illustrate the spatial variability and magnitude of the impacts that might be expected for several target ANC levels (50, 30 and 20 µg/L) and the temporal changes across the 20-year period. For example, during the two most recent 3-year periods, the ecoregion median S deposition estimates in 2014-16 were below 5 kg/ha-yr in all ecoregions and the estimates for 2018-20 were all below 4 kg/ha-yr. In this analysis, we summarized the ecoregion-level exceedances of CLs for each of the ANC targets in each of the five time periods. While recognizing limitations and associated uncertainties of these analyses, we note several key observations.

Although the ecoregion S deposition estimates in the 18 eastern ecoregions analyzed were all below 5 kg/ha-yr in the two most recent time periods (2014-16 and 2018-20), the full dataset of five time periods ranges from below 2 up to nearly 18 kg/ha-yr. Across this dataset of CL exceedances for the three ANC targets for all 90 eastern ecoregion-time period combinations, 73% of the combinations had at least 90% of waterbodies per ecoregion estimated to achieve ANC at or above 20 µeq/L, and 60% had at least 90% of the waterbodies estimated to achieve ANC at or above 50 µeq/L. In the early ecoregion-time period combinations fewer than half of the eastern ecoregion-time period combinations (and all of the western combinations) had an S deposition estimate below 4 kg/ha-yr.

Ninety percent of the eastern ecoregion-time period combinations were for ecoregion deposition estimates at or below 13 kg/ha-yr. For these combinations (at or below 13 kg/ha-yr), at least 90% of waterbodies per ecoregion were estimated to achieve an ANC at or above 20, 30 and 50 µeq/L in 80%, 73% and 65% of all ecoregion-time period combinations, respectively. For S deposition estimates at or below 9 kg/ha-yr (approximately three quarters of the combinations), at least 90% of all waterbodies per ecoregion were estimated to achieve ANC at or above 20, 30 and 50 µeq/L in 87%, 81% and 72% of combinations. respectively. For S deposition estimates at or below 5 kg S/ha-yr, these values are 96%, 92% and 82% of combinations. For the 75 western ecoregion-time period combinations, all of which had an S deposition estimate below 4 kg/ha-yr, at least 90% of waterbodies per ecoregion were estimated to achieve an ANC at or above 50 µg/L.

The case study analyses provide estimates of S deposition that might be expected to allow these geographically diverse locations, including several Class I areas, to meet the three ANC targets. In reviewing these estimates, we recognize inherent limitations and associated uncertainties. Focusing on the three eastern case studies, the CL modeling indicates that at an annual average S deposition of 9-10 kg/ha-yr, the sites in these areas, on average, might be expected to achieve an ANC at or above 50 µeq/L. At an annual average S deposition of about 6-9 kg/ha-yr, 70% of the sites in the areas are estimated to achieve an ANC at or above 20 µeq/L and at about 5-8 kg/ha-yr, 70% are estimated to achieve an ANC at or above 30 µeq/L. Lower S deposition values are estimated to achieve higher ANC across more sites. Across the three eastern areas, the CL estimates for each ANC target are lowest for the White Mountains National Forest study area, and highest for the Shenandoah Valley study area.

5.2 NITROGEN ENRICHMENT IN AQUATIC ECOSYSTEMS

There are several other categories of effects to aquatic ecosystems from deposition of nitrogen and sulfur for which there is significant scientific evidence and causality judgements, as described in Chapter 4. These include N enrichment in various types of aquatic systems, including freshwater streams and lakes, estuarine and near-coastal systems, and wetlands, as described in section 4.3.1.[9] Separate quantitative analyses were not performed for these categories of effects in this review due to recognition of a number of factors, including modeling and assessment complexities, and site- or waterbody-specific data requirements, as well as, in some cases, issues of apportionment of atmospheric sources separate from other influential sources.

[9] Two other categories of effects assessed in the ISA (and for which causal determinations are made) are mercury methylation, and sulfide toxicity (ISA, Appendix 12), as summarized in sections 4.4.1 and 4.4.2 above.

5.2.1 Freshwater Wetlands

New information has become available since the 2008 ISA on N critical loads for U.S. wetlands. While critical loads have previously been identified for European wetlands such as bogs, fens, and intertidal wetlands for a variety of endpoints including plant growth and species composition, peat and peat water chemistry, decomposition of organic material, and nutrient cycling (Bobbink et al., 2003), recent studies have shown that CLs for *Sphagnum* moss effects in European bogs may not be directly relevant or transferrable to North American and/or U.S. wetlands (ISA, Appendix 11, section 11.3.1.6). With regard to North American freshwater wetlands, some limited new information is available in this review. For example, a CL for wetland C cycling, quantified as altered peat accumulation and net primary productivity, has been estimated between 2.7 and 13 kg N/ha-yr based on four studies (Greaver et al., 2011; ISA, Appendix 11, section 11.9.1). Additionally, N loading between 6.8-14 kg N/ha-yr has been suggested by empirical evidence and modeling to be protective of populations of purple pitcher plants (*Sarracenia purpurea*) based on morphology and population dynamic endpoints (Gotelli and Ellison, 2002, 2006). At the lowest experimental addition level (16 kg N/ha-yr), which has been assessed in several studies, there are observations of altered C and N cycling and altered biodiversity (ISA, Appendix 11). The endpoints affected include decreases in moss cover, increased peat biomass, decreased N retention efficiency, and altered/damaged leaf stoichiometry in vascular plants (ISA, Appendix 11, section 11.10.2).

5.2.2 Freshwater Lakes and Streams

Since the 2008 ISA, empirical and modeled critical loads for the U.S. have been estimated based on surface water NO_3^- concentration, diatom community shifts, and phytoplankton biomass growth nutrient limitation shifts. A critical load ranging from 3.5 to 6.0 kg N/ha-yr was identified for high-elevation lakes in the eastern U.S. based on the nutrient enrichment inflection point (where NO_3^- concentrations increase in response to increasing N deposition). Another critical load of 8.0 kg N/ha-yr was estimated by Pardo et al. (2011) for eastern lakes based on the value of N deposition at which significant increases in surface water NO_{3^-} concentrations occur. In both Grand Teton and Yellowstone national parks, critical loads for total N deposition ranged from $<1.5 \pm 1.0$ kg N/ha-yr to $>4.0 \pm 1.0$ kg N/ha-yr (Nanus et al., 2017; ISA, Appendix 9, section 9.5).

Additional critical loads have been identified since the 2008 ISA for eastern Sierra Nevada lakes, Rocky Mountain lakes, the Greater Yellowstone Ecosystem, and Hoh Lake, Olympic National Park (ISA, Appendix 9, Table 9-4). The identified values fall near or within the range of 1.0 to 3.0 kg N/ha-yr for western lakes (Baron et al., 2011). An empirical critical load of 4.1 kg/TN/ha-yr above which phytoplankton biomass P limitation is more likely than N

limitation was identified by Williams et al. (2017) for the western U.S. using univariate regression modeling of available water chemistry data from 2006-2011 for 208 western U.S. mountain lakes, with prediction of a ratio of dissolved inorganic N to total phosphorus as the response variable (ISA, Appendix 9, section 9.5); the lake-specific estimates ranged from 2.8 to 5.2 kg/TN/ha-yr. This evidence is geographically specific, perhaps even waterbody specific, and is not available for most of the U.S.

Larger freshwater lakes, such as the Great Lakes, and freshwater portions of large river systems are also susceptible to eutrophication from N loading (ISA, Appendix 9, section 9.1). In these larger systems, atmospheric N from direct deposition, runoff, and leaching from terrestrial ecosystems combines with other diffuse and point sources of N. The contribution from other terrestrial sources of N, such as fertilizer, livestock waste, septic effluent, and wastewater treatment plant outflow, often becomes much more important in these large waterbodies than in headwater and upland areas (ISA, Appendix 9, section 9.1.1.1). Further, N limitation appears to have become increasingly common in freshwater systems, likely due to alteration of nutrient dynamics from increased agricultural and urban P inputs (Appendix 9, section 9.1; Paerl et al., 2016; Grantz et al., 2014; Paerl et al., 2014; Finlay et al., 2013).

5.2.3 Estuaries, Coastal Waters and Coastal Wetlands

Information newly available in this review includes new applications of models that have quantified eutrophication processes in estuaries and near-coastal marine ecosystems (ISA, section IS.7). These have included applications of N cycling or hypoxia models, as well as modeling the apportionment of N loads in these systems.

In U.S. coastal wetlands, two studies are available that have considered N loads below 100 kg N/ha-yr. Wigand et al. (2003) observed associations of estimated N loading with plant community structure in 10 saltmarsh sites around Narragansett Bay but indicated that confounding effects of marsh physical characteristics made unclear the extent to which N enrichment contributed to variation in plant structure. A N addition experiment in a Narragansett Bay saltmarsh by Caffrey et al. (2007) provided evidence that 80 kg N/ha-yr can alter microbial activity and biogeochemistry.

The relationship between N loading and algal blooms, and associated water quality impacts, has led to numerous water quality modeling projects to inform water quality management decision-making in multiple estuaries, including Chesapeake Bay, Narraganset Bay, Tampa Bay, Neuse River Estuary and Waquoit (ISA, Appendix 7, section 7.2). These projects often utilize indicators of nutrient enrichment, such as chlorophyll a, dissolved oxygen and abundance of submerged aquatic vegetation, among others (ISA, section IS.7.3 and Appendix 10, section 10.6). For these estuaries, the available information regarding atmospheric deposition

and the establishment of associated target loads varies across the various estuaries (ISA, Appendix 7, Table 7-9).

The establishment of target loads is in many areas related to implementation of the total maximum daily load (TMDL) requirements of section 303(d) of the Clean Water Act. Under the CWA, section 303(d), every two years, states and other jurisdictions are required to list impaired waterbodies not meeting water quality standards. For waterbodies on the list, a TMDL must be developed that identifies the maximum amount of pollutant a waterbody can receive and still meet water quality standards, e.g., standards for dissolved oxygen and chlorophyll a (which are indicators of eutrophication).

Nutrient load allocation and reduction activities in some large estuaries predate development of CWA 303(d) TMDLs. The multiple Chesapeake Bay Agreements signed by the U.S. EPA, District of Columbia, and states of Virginia, Maryland, and Pennsylvania first established the voluntary government partnership that directs and manages bay cleanup efforts and subsequently included commitments for reduction of N and P loading to the bay. Efforts prior to 2000 focused largely on point-source discharges, with slower progress for nonpoint-source reductions via strategies such as adoption of better agricultural practices, reduction of atmospheric N deposition, enhancement of wetlands and other nutrient sinks, and control of urban sprawl (2008 ISA, section 3.3.8.3).

Studies since 2000 estimate atmospheric deposition to contribute substantially to the overall N budget for Chesapeake Bay (ISA, Appendix 7, section 7.2.1; Howarth, 2008b; Boyer et al., 2002). In the TMDL established for Chesapeake Bay in 2010, atmospheric deposition was recognized as the major N source to the Chesapeake Bay watershed, greater than the other sources of fertilizer, manures, or point sources (U.S. EPA, 2010). The TMDL modeling estimated seventy-five percent of the atmospheric N loading to the Chesapeake watershed to originate from sources within the Bay airshed (U.S. EPA, 2010). The 2010 TMDL included a loading allocation for atmospheric deposition of N directly to tidal waters of 15.7 million lbs/year (7.1 million kg/yr), which was projected to be achieved by 2020 based on air quality progress under existing Clean Air Act regulations and programs (U.S. EPA, 2010). With that projection in reduced atmospheric loading, water quality modeling was used to identify the reductions across the subbasins and tributaries that were needed to enable water quality standards for dissolved oxygen to be achieved in the mainstem of the Bay and the major tidal river segments. The total additional N loading reduction is 185.93 million lbs/year, to be achieved by actions of the seven jurisdictions in the Chesapeake Bay watershed, which includes six States and the District of Columbia (U.S. EPA, 2010).

Jurisdictions for other U.S. estuaries have also developed TMDLs to address nutrient loading causing eutrophication. For example, atmospheric deposition in 2000 was identified as

the third largest source of N loading to Narragansett Bay (via the watershed and directly to the water body), which, to Narragansett Bay in the year 2000, was atmospheric deposition (20%) (ISA, Appendix 7, section 7.2.1). Similarly atmospheric deposition was estimated to account for approximately a third of N input to several small- to medium-sized estuaries of southern New England, with the percentage varying widely for individual estuaries (ISA, Appendix 7, section 7.2.1; Latimer and Charpentier, 2010). Another modeling study in the Waquoit Bay estuaries in Cape Cod, MA, using data since 1990, estimated atmospheric deposition to have decreased by about 41% while wastewater inputs increased 80% with a net result that total loads were concluded to not have changed over that time period (ISA, Appendix 7, section 7.2.1). Another well studied estuarine system is Tampa Bay, for which a 2013 study estimated atmospheric sources to account for more than 70% of total N loading based on 2002 data (ISA, Appendix 7, section 7.2.1). The TMDL for Tampa Bay allocates 11.8 kg/ha-yr N loading to atmospheric deposition (ISA, Appendix 16, section 16.4.2; Janicki Environmental, 2013). The Neuse River Estuary is another for which modeling work has investigated the role of atmospheric N deposition nutrient enrichment and associated water quality indicators, including chlorophyll a (ISA, Appendix 10, section 10.2).

Nitrogen loading to estuaries has also been considered with regard to impacts on submerged aquatic vegetation. For example, eelgrass coverage was estimated to be markedly reduced in shallow New England estuaries with N loading at or above 100 kg N/ha-yr (ISA, Appendix 10, section 10.2.5). Another study estimated loading rates above 50 kg/ha-yr as a threshold at which habitat extent may be impacted (ISA, Appendix 10, section 10.2.5; Latimer and Rego, 2010). Factors that influence the impact of N loading on submerged vegetation includes flushing and drainage in estuaries (ISA, Appendix 10, section 10.6).

5.2.4 Summary: Key Findings and Associated Uncertainties

The eutrophication of wetlands and other aquatic systems is primarily associated with nitrogen inputs whether from deposition or other sources. The ranges of deposition associated with these effects is very broad and ranges from levels on the order of a few kg N/ha-yr for impacts to diatom communities in high elevation lakes to over 500 kg N/ha-yr for some effects of interest in some wetland N addition studies. While the information available on these types of impacts is sufficient for causal determinations it is often very localized and difficult to utilize more broadly, such as for the purpose of quantitative assessment relating deposition to waterbody response at an array of U.S. locations. Accordingly, in this review, this information was considered from a more descriptive perspective in characterizing conditions reported in the evidence as associated with various effects described in Chapter 4.

There is also a wealth of information available for estuaries and coastal systems. Over the past few decades, modeling analyses have been conducted in multiple estuaries and large river systems to relate N loading to various water quality indicators, including chlorophyll a, dissolved oxygen and also prevalence of habitat, such as SAV. While a focus is identification of total N loading targets for purposes of attaining water quality standards for such indicators, the modeling work also includes apportionment of sources, which vary by system. The assignment of targets to different source types (e.g., groundwater, surface water runoff and atmospheric deposition) in different waterbodies and watersheds the also varies for both practical and policy reasons. Further, during the multi-decade time period across which these activities have occurred, atmospheric deposition of N in coastal areas has declined. In general, however, atmospheric deposition targets for N for the large systems summarized above have been on the order of 10 kg/ha-yr, with some somewhat lower and some somewhat higher.

5.3 EFFECTS OF S AND N DEPOSITION IN TERRESTRIAL ECOSYSTEMS

As noted in the introduction to this chapter, analyses in the 2012 review that related atmospheric deposition in recent times (e.g., since 2000) to terrestrial effects, or indicators of terrestrial ecosystem risk, were generally considered to be more uncertain than conceptually similar modeling analyses for aquatic ecosystems (e.g., "aquatic acidification is clearly the targeted effect area with the highest level of confidence" (2009 REA, section 7.5; 2011 PA, section 1.3). The terrestrial analyses in the 2012 review were comprised of a critical load-based quantitative modeling analysis focused on BC:Al ratio in soil (the benchmarks for which are based on laboratory responses rather than field measurements) and a qualitative characterization of nutrient enrichment (2009 REA). The more qualitative approach taken for nutrient enrichment in the 2012 review involved describing deposition ranges identified from observational or modeling research as associated with potential effects/changes in species, communities and ecosystems, with recognition of uncertainties associated with quantitative analysis of these depositional effects (2011 PA, section 3.2.3).

In this review, rather than performing new quantitative analyses focused on terrestrial ecosystems, we draw on prior analyses (e.g., in the 2009 REA) and published studies recognized in the ISA that provide information pertaining to deposition levels associated with effects related to terrestrial acidification and N enrichment. This approach considers the available studies and with investigation into various assessment approaches. Unlike aquatic acidification where a full quantitative exposure and risk assessment has been conducted (see section 5.1) at multiple scales because the available information, tools and assessment approaches provide strong support for analyses that are targeted to the needs in this review, we determined that such an approach is not

warranted for terrestrial effects related to N and S deposition in this review based on our assessment of the available information and tools and current review needs. Therefore, this section draws on the wealth of quantitative information relating deposition to consideration of terrestrial ecosystem effects, as described below and in the following subsections.

Since the 2012 combined review of the secondary NAAQS for N oxides and SO_X, in addition to publications of analyses that apply steady-state (and dynamic) modeling to predict future soil acidity conditions in various regions of the U.S. under differing atmospheric loading scenarios (ISA, Appendix 4, section 4.6.2), several publications have analyzed large datasets from field assessments of tree growth and survival, as well as understory plant community richness, with estimates of atmospheric N and/or S deposition (ISA, Appendix 6, section 6.5). These latter studies investigate the existence of associations of variations in plant community or individual measures (e.g., species richness, growth, survival) with a metric for deposition during an overlapping time period, generally of a decade or two in duration. Soil acidification modeling and observational studies, as well as experimental addition studies, are, to various extents, informative in considering N and S deposition levels of interest in the review.

In general, observational or gradient studies differ from the chemical mass balance modeling approach in a number of ways that are relevant to their consideration and utilization for our purposes in this review. One difference of note is the extent to which their findings reflect or take into account the ecosystem impacts of historical deposition. Observational studies are describing variation in indicators in the current context (with any ecosystem impacts, including stores of deposited chemicals that remain from historical loading). Historical loading, and its associated impacts, can also contribute to effects analyzed with estimates of more recent deposition in observational studies. Mass balance modeling, in the steady-state mode that is commonly used for estimating critical loads for acidification targets, does not usually address the complication of historical deposition impacts that can play a significant role in timing of system recovery. In this type of modeling, timelines of the various processes are not addressed. While this provides a simple approach that may facilitate consideration unrelated to timelines, it cannot address the potential for changes in influential factors that may occur over time with different or changed deposition patterns.

For example, in considering the potential for terrestrial ecosystem impacts associated with different levels of deposition, the simple mass balance models common for estimating critical acid loads related to BC:Al ratio are often run for the steady state case. Accordingly, the underlying assumption is that while historic deposition, and the various ways it may affect soil chemistry into the future (e.g., through the stores of historically deposited sulfur), may affect time to reach steady state (e.g., as the system processes the past loadings), it would not be expected to affect the steady state solution (i.e., the estimated critical load for the specified soil

acidification indicator target). The complexities associated with site-specific aspects of ecosystem recovery from historic depositional loading (which contribute uncertainties to interpretation of steady-state solutions) become evident through application of dynamic models.

Observational studies, on the other hand, due to their focus on an existing set of conditions, are inherently affected by the potential influence of historical deposition and any past or remaining deposition-related impacts on soil chemistry and/or biota, in addition to other environmental factors. The extent of the influence of historical deposition (and its ramifications) on the associations reported in these studies with metrics quantifying more recent deposition is generally not known. Where patterns of spatial variation in recent deposition are similar to those for historic deposition, there may be potential for such influence. This is an uncertainty associated with interpretation of the observational studies as to the deposition levels that may be contributing to the observed variation in plant or plant community responses. Thus, while observational studies contribute to the evidence base on the potential for N/S deposition to contribute to ecosystem effects (and thus are important evidence in the ISA determinations regarding causality), their uncertainties (and underlying assumptions) differ from those of modeling analyses, and they may be somewhat less informative with regard to identification of specific N and S deposition levels that may elicit ecosystem impacts of interest. Both types of studies, as well as N addition experiments, which are not generally confounded by exposure changes beyond those assessed yet may have other limitations (see section 5.3.4 below), are considered in the sections below.

5.3.1 Soil Chemistry Response

Quantitative linkages between N and S deposition and soil chemistry responses vary across the geography of the U.S. As summarized in sections 4.2 and 4.3 above, acidification and N enrichment processes can alter the biogeochemistry in terrestrial ecosystems (ISA, Appendix 4). There are several indicators of acidification and N enrichment that also have linkages to biological responses that are commonly used in quantitative analyses (ISA, Appendix 4, Table 4-1). These indicators are soil characteristics strongly associated with specific aspects of soil acidification or nutrient enrichment. Uncertainties in the estimates of these indicators in quantitative analyses for specific areas will generally be associated with limitations in the estimation approach and the associated parameter values for those locations.

A number of soil characteristic metrics have been identified to have relationships with biological responses, making them useful indicators for assessing potential soil acidification impacts. One commonly used indicator for quantitative modeling analyses of the effect of acidifying deposition on forests (see section 5.2.2 below) is the ratio of base cations to aluminum (BC:Al), with higher ratios indicating a lower potential for acidification-related biological effects

(ISA, Table IS-2). The ratio in soil solution can be reduced by release of base cations from the soil (e.g., through the process of neutralizing drainage water acidity), which reduces the base saturation of the soil. Soil base saturation[10] and changes to it can also be an indicator of acidification risk (ISA, Appendix 4, section 4.3.4). The accelerated loss of base cations through leaching can cause a decrease in base saturation and decreases in soil solution Ca:Al ratio, which are all indicators of soil acidification. Inorganic and organic acids can be neutralized by soil weathering or base cation exchange, in addition to denitrification (ISA, Appendix 4, section 4.3). Some studies have indicated soil base saturation to be a better indicator than BC:Al ratio, and one for which metrics associated with potential risk may have a more well-founded basis as a more robust indicator for field assessment (e.g., Sullivan et al., 2013).

There are many indicators of N enrichment and potential eutrophication, including N accumulation, e.g., increased soil N concentrations or decreased carbon to nitrogen (C:N) ratios (ISA, section IS.5.1.1). The ratio of soil C to soil N can be indicative of ecosystem N status; it is a "reliable and relatively straightforward measure for identifying forest ecosystems that may be experiencing soil acidification and base leaching as a result of N input and increased nitrification" (ISA, Appendix 4, p. 4-39). Accordingly, the C:N ratio can be useful in informing assessments of the potential for accelerated nitrification and nitrate leaching (ISA Appendix 4, section 4.3.6; Aber et al., 2003).

Increases in soil N can lead to nitrate leaching, potentially imposing a drain on base cations and a potential for increased acidity (ISA, Appendix 4, section 4.3). Thus, nitrate leaching can be an indicator of potential for increased aquatic acidity, as well as for terrestrial or aquatic N enrichment. Studies in various locations throughout the eastern U.S. and in the Rocky Mountains have reported estimates of N deposition associated with an onset of increased nitrate leaching (ISA, Appendix 4, sections 4.3.2 and 4.6.2). For example, based on monitoring results for an 8-year experimental addition experiment in an alpine dry meadow in the Rocky Mountains, with annual additions of 20, 40 and 60 kg N/ha-yr (Bowman et al., 2006), Bowman et al. (2014) reported 10 kg N/ha-yr to be associated with enhanced nitrate leaching at this location (ISA, Appendix 4, section 4.6.2.2).

Thus, the response of a terrestrial system, and the associated biota, to N additions as through atmospheric deposition, can be one of acidification and/or nutrient enrichment depending on the geology and soil chemistry (e.g., base cation weathering rate or base cation exchange capacity), residual impacts of historic deposition (e.g., SO_4^{2-}/NO_3^- stored in soil) and organic content, as well as acid sensitivity or growth limitations of the resident species. With

[10] As described in the ISA, "[s]oil base saturation expresses the concentration of exchangeable bases (Ca, Mg, potassium [K], sodium [Na]) as a percentage of the total cation exchange capacity (which includes exchangeable H^+ and inorganic Al)" (ISA, Appendix 4, p. 4-27).

regard to soil indicators of nutrient enrichment (i.e., levels associated with particular risk of harm or degree of protection), there is little research in the U.S. on which to base target values for indicators such as soil N accumulation or NO_3^- leaching (Duarte et al., 2013). This and uncertainties associated with site-specific characteristics (e.g., carbon and organic content of soils) may affect the use of soil modeling for identifying deposition targets aimed at controlling nutrient enrichment.

5.3.2 Effects on Trees

In this section we summarize the findings related to quantitative evaluation of S and N deposition effects on trees. While S deposition contributes to acidification and its associated negative effects on terrestrial systems, N deposition, as described in Chapter 4 and section 5.3.1 above, may contribute to acidification and/or nutrient enrichment, with associated effects on tree growth and survival that, for acidification, can be negative and, for nutrient enrichment, can be positive or negative. While the response is influenced by site-specific characteristics, some species-specific patterns have also been observed (ISA, Appendix 6, section 6.2.3.1). For example, conifer species, particularly at high elevations, were more likely to exhibit negative growth responses or mortality in response to added N and less likely to demonstrate increased growth (ISA, Appendix 6, section 6.2.3.1; McNulty et al., 2005; Beier et al., 1998; Boxman et al., 1998). Variation in response can also be related to site-specific factors contributing to variations associated with location. For example, while some long-term N addition experiments indicate that broadleaf species more commonly exhibit increased growth (than conifers), there is variation across studies as seen in Appendix 5B (Table 5B-1). The extent to which species-specific observations are related to the site-specific characteristics of areas where species are distributed or to species-specific sensitivities is not clear.

In the subsections below, we draw on three main categories of studies: steady-state mass balance modeling of soils, experimental addition studies, and observational or gradient studies of trees. As noted in section 5.3. above, each of these categories of studies has associated strengths and limitations/uncertainties for our purposes here. For example, while the mass balance modeling studies are explicitly focused on acidic deposition effects, observational studies, given their real-world settings, may reflect patterns of deposition contributing to both acidic deposition and/or the effects of nutrient enrichment. Thus, the subsections below are organized by study category within which the findings with regard to both types of effects are discussed.

5.3.2.1 Steady-State Mass Balance Modeling of Terrestrial Acidification

Consistent with assessment of aquatic acidification (see section 5.1 above), steady-state mass balance modeling is also utilized to identify N/S deposition rates associated with conditions posing differing risks to tree health. The evidence base evaluating such modeling, however, is

less robust than for aquatic ecosystems, such that the foundation for identifying target conditions for neutralizing acidification, and for identifying appropriate values for some model parameters, is more limited and uncertain, as noted below.

As noted in section 5.3.1 above, an indicator commonly utilized to identify conditions associated with protection from acidifying deposition risks to tree growth and survival is BC:Al (ISA, Appendix 5, section 5.2.1). There are limitations, however, in the ability of this ratio for indicating tree health risk. Accordingly, some more recent studies have emphasized other indicators such as exchangeable Ca or soil base saturation (e.g., Sullivan et al., 2013).

Limitations associated with use of BC:Al ratios include those related to their interpretation. More specifically, the two meta-analyses often referenced to inform interpretation of estimated BC:Al ratios with regard to associated potential risks to tree health – Sverdrup and Warfvinge (1993) and Cronan and Grigal (1995) – were largely based on soil solution concentrations derived from laboratory and greenhouse studies (Sverdrup and Warfvinge, 1993; Cronan and Grigal, 1995).[11] For example, the literature review by Cronan and Grigal (1995), which reported the Ca:Al ratios in 35 studies in which a response in seedling roots (e.g., change in nutrient content) were reported, is also often cited as a basis for selection of a target BC:Al value for use in simple mass balance models. Nearly all of the 35 studies were conducted in hydroponic or sand systems, in which aluminum is generally more freely available than in a soil substrate (Cronan and Grigal, 1995). As would be expected, there are limitations and uncertainties associated with findings involving artificial substrates and growing conditions (ISA, Appendix 5, section 5.2.1).[12] In consideration of these analyses, the BC:Al targets used in the 2009 REA for identifying acidifying deposition loads that might provide different levels of protection range from less than 1 to 10. Use of such target values (of 0.6, 1 and 10) in steady state simple mass balance modeling in the last review resulted in the identification of acidifying deposition loads ranging from 487 to 2009 eq/ha-yr, across two areas of the Northeast for BC:Al target values differing by a factor of nearly 20 (Table 5-7 and Table 5-8).

[11] Ratios of BC:Al were identified using the cumulative percentage of experiments for tree seedling species grown in solution reporting a 20% growth reduction (Sverdrup and Warfvinge, 1993). For example, at cumulative percentage of 50% the BC:Al ratio was 1.2, and at 100% the ratio was on the order of 8 (Sverdrup and Warfyinger, 1993). The 2009 REA concluded that this analysis reported critical BC:Al ratios ranging from 0.2 to 0.8 (2009 REA, p. 4-54).

[12] Based on the distribution Ca:Al ratios in the studies, Cronan and Grigal (1995) estimated a 50% risk of tree growth response for a molar ratio of 1.0 based on fact that 17 of the 35 studies had ratio at/above 1.0. The percentage of studies with a ratio at/above 1.8 was 25%., and it was approximately 5% at a ratio of 5, based on there being 33 of 35 or 94% of studies reporting a response for a Ca/Al ratio above 5. Only two of the 35 studies, both in conifers, reported a response, a change to root nutrient content (Cronan and Grigal (1995). In this assessment, "plant toxicity or nutrient antagonism was reported to occur at Ca/Al ratios ranging from 0.2 to 2.5" (2009 REA, p. 4-54).

Table 5-7. Acid deposition levels estimated for BC:Al targets in 24-state range of red spruce and sugar maple using steady-state simple mass balance model (2009 REA).

Target BC:Al	Critical Loads for Acid Deposition for Different BC:Al Targets		
	In terms of S+N (eq/ha-yr)	*In terms of S (kg S/ha-yr)*	*In terms of N (kg N/ha-yr)*
0.6	1237- 2009	*40-64*	*17-28*
1	892-1481	*29 - 48*	*13-21*
10	487-910	*16- 29*	*7-13*

The 2009 REA (that informed the 2012 review of the NAAQS for N oxides and SO_X review) used the Simple Mass Balance (SMB) model for forest soil acidification, in steady-state mode, to assess the extent to which atmospheric S and N deposition for the year 2002 might be expected to contribute to soil acidification of potential concern (with BC:Al ratio used as an indicator) for the sensitive species of sugar maple and red spruce in areas of 24-states where they are native (2011 PA, section 3.1.3; 2009 REA, section 4.3). The critical load analysis for the three target BC:Al ratio values (identified for different levels of risk for growth impacts) drawn from an estimated relationship between tree growth effect for different species and BC:Al ratio yielded an array of estimates of acidifying deposition with potential to affect the health of at least a portion of the sugar maple and red spruce growing in the United States (2009 REA, section 4.3 and Appendix 5; 2011 PA).

In addition to the uncertainty associated with characterization of risk for target BC:Al ratio values, uncertainties were recognized in the SMB model calculations for the 2009 REA analyses. For example, uncertainty recognized with the findings related to the use of default values for several key parameters (e.g., denitrification, nitrogen immobilization, the gibbsite equilibrium constant, and rooting zone soil depth), and dependence of the SMB calculations on assumptions made in its application (2009 REA, section 4.3.9). Similarly, the ISA discussion of SMB equations summarized findings of Li and McNulty (2007), who found uncertainty to come primarily from components of the estimates for base cation weathering and acid-neutralizing capacity (ISA, Appendix 4, section 4.5.1.2).

Since the 2009 REA, an updated approach to estimating one particularly influential parameter in the soil BC:Al modeling (cation weathering) has been reported (Phelan et al., 2014). Use of the new approach at 51 forested sites in Pennsylvania yielded rates consistent with soil properties and regional geology. The updated rates were generally higher, indicating a greater buffering capacity for sites in this area to acidifying deposition than previously determined (Phelan et al., 2014). The recent study by Duarte et al. (2013) also used updated values for cation weathering for a study extending across New England and New York. For a soil

BC:Al target of 10, this study reported a range of deposition estimates slightly higher than those from the 2009 REA (see Table 5-8 below).

Table 5-8. Acidic deposition levels estimated for several BC:Al ratio targets by steady-state mass balance modeling for sites in northeastern U.S.

Endpoint, Species, Location	**Deposition/Addition** (loading)	**Notes**
---------- *Modeling Analyses – Steady-state mass balance* ----------		
Range of risk for reduced growth (sugar maple and red spruce) in areas of 24 states in Northeast, based on soil BC:Al targets of 0.6, 1 and 10	487 to 2009 eq/ha-yr (7-28 kg N/ha-yr or 16-64 kg S/ha-yr)	2009 REA
Soil BC:Al target of 10 for forest protection at >4000 plots in New England and New York.	For a BC:Al target of 10, 850-2050 eq/ha-yr (27-66 kg S/ha-yr or 12-29 kg N/ha-yr), range for 80% of sites (for a BC:Al target of 10) total range was 11 to 6,540 eq ha−1yr−1, the lowest loads in Maine, NH and VT	Duarte et al. (2013)

5.3.2.2 Experimental Addition Studies

A number of experimental addition studies, conducted primarily in the eastern U.S., have reported mixed results for growth and survival (see Appendix 5B, Table 5B-1). The species studied have included oaks, spruce, maples, and pines. (Magill et al., 2004; McNulty et al., 2005; Pregitzer et al., 2008; Wallace et al., 2007). Some multiyear S or N addition experiments (involving additions greater than 20 kg/ha-yr) with a small set of eastern species, including sugar maple, aspen, white spruce, yellow poplar, black cherry, have not reported tree growth effects (ISA, Appendix 5, section 5.5.1; Bethers et al., 2009; Moore and Houle, 2013; Jung and Chang, 2012; Jensen et al., 2014). Studies described in Appendix 5B are summarized here, including the annual amounts of N added (in addition to the background deposition occurring during these times):

- Additions of 25 to as high as 150 kg N/ha-yr for 8-14 years (dating back to 1988) were associated with increased growth reported in sugar maple and oaks, at sites in MI, MA, NY, ME.
- Additions of 15.7 and 31.4 kg N/ha-yr for 14 years (beginning in 1988) were associated with reduced basal area (red spruce) or growth (red maple, tulip poplar and black cherry, red pine) at sites in VT, MA, WV.
- Additions of 25 kg N/ha-yr for 13 years (beginning in 1989) were associated with increased growth rates for sugar maple but not for red spruce.

The N deposition levels simulated in experimental addition studies that report tree effects, including either increased or reduced growth, are generally greater than 10 kg N/ha-yr (Appendix 5B, Table 5B-1).

5.3.2.3 Observational or Gradient Studies

Since the last review of the NAAQS for N oxides and SO_X, several observational studies have been published that investigate the existence of statistical associations between tree growth or survival, as assessed at U.S. Forest Service, Forest Inventory and Analysis program (USFS/FIA)[13] sites across the U.S., and estimates of average deposition of S or N compounds averaged over multiyear time periods (Appendix 5B, section 5B.2.2; ISA, Appendix 5, section 5.5.2 and Appendix 6, section.6.2.3.1; Dietze and Moorcroft, 2011; Horn et al., 2018). The standardized protocols employed in the FIA program make the use of the FIA plot data a strength of these studies. These studies generally utilized the tree measurement data collected by the USFS from periodic assessments at each site, and data for other factors analyzed, including metrics for atmospheric deposition (Table 5-9; Dietze and Moorcroft, 2011; Thomas et al., 2010; Horn et al., 2018).

The study by Dietze and Moorcroft (2011) statistically evaluated the influence of a number of factors, in addition to $SO_4^{2-}$and NO_3^- wet deposition (site-specific estimates of average of 1994-2005 annual averages), on tree mortality (assessed over 5-15-year measurement intervals within the period from 1970s through early 2000s) in groups of species characterized by functional type (267 species categorized into 10 groups) at sites in the eastern and central U.S. (Appendix 5B, section 5B.2.2.1; ISA, Appendix 5, section 5.5.2). The full range of average SO_4^{2-} deposition was 4 to 30 kg S/ha-yr (Dietze and Moorcroft, 2011). Other factors assessed (which were all found to have statistically significant associations with more than one of the tree species groups) were precipitation, minimum and maximum temperature, ozone, topographic factors (elevation, slope and variation in solar radiation and soil moisture), and biotic interaction factors (stand basal area and age, and focal-tree diameter at breast height). The authors reported that the strongest effect on mortality was due to acidifying deposition (specifically SO_4^{2-}), particularly in the northeast sites (Dietze and Moorcroft, 2011). Negative associations were reported with tree survival for 9 of the 10 functional groups. Survival for the same 9 groups was also negatively associated with long-term average ozone concentrations. The third highest influence was for N deposition (range across sites was 6 to 16 kg N/ha-yr), with mortality in all but one species group having a negative association (i.e., lower probability of mortality with higher NO_3^- deposition). Regarding the significant associations with S and N deposition, the authors recognized that "[t]he impacts of both acidification and nitrogen deposition on tree mortality result from cumulative,

[13] The FIA Program's forest monitoring component involves periodic assessments of an established set of plots distributed across the U.S. This component includes collection of data at field sites (one for every 6,000 acres of forest). The data include forest type, site attributes, tree species, tree size, and overall tree condition. At a subset of the plots, a broader suite of forest health attributes including tree crown conditions, lichen community composition, understory vegetation, down woody debris, and soil attributes are also assessed (USFS, 2005).

long-term deposition, and the patterns presented [in their paper] should be interpreted in that light" (Dietze and Moorcroft, 2011).

The study by Thomas et al. (2010) focused on relationships of tree growth and survival (assessed at FIA plots from 1978 through 2001, with measurement interval ranging from 8.3 to 14.4 years) with N deposition (mean annual average for 2000-04) as the only pollutant included in the statistical analyses (Appendix 5B, section 5B.2.2.2). Increased growth was associated with higher N deposition in 11 of 23 species in northeastern and north-central U.S and with lower N deposition in three species (Thomas et al., 2010). Eight species showed negative associations of survival rates with N deposition and three showed positive associations. The other factors analyzed included temperature, precipitation, and tree size, but did not include other pollutants (Thomas et al., 2010).

The third study utilizing measurements at USFS plots, reported on statistical modeling of tree growth and survival of 71 species at USFS plots across the U.S. with site-specific estimates of average S and N deposition across the measurement interval (generally 10 years) within the period from 2000-2013 (Horn et al., 2018; Appendix 5B, section 5B.2.2.3). The study focused on 71 of 94 species for which covariance between N and S deposition metric values and other factors was a lower concern (Horn et al., 2018). Of the 71 species on which the analysis focused, negative associations were reported for survival and growth with S deposition estimates for 40 and 31 species, respectively. Sulfur deposition at sites of these species ranged from a minimum below 5 kg/ha-yr to a site maximum above 40 kg/ha-yr, with medians for these species generally ranging from around 5 to 12 kg/ha-yr (Appendix 5B, section 5B.2.3).

The study by Horn et al. (2018) also reported associations of growth and survival with N deposition estimates that varied positive to negative across the range of deposition at the measurement plots for some species, and also among species (Horn et al., 2018). For the six species, for which survival was negatively associated with the N deposition metric across the full range of values, the site-specific deposition metric ranged from below 5 to above 50 kg/ha-yr, with medians ranging from 8 to 11 kg N/ha-yr (Appendix 5B, Figure 5B-7). The median values for the 19 other species with unimodal (or hump-shaped) associations that were negative at the species median deposition value (and for which sites were not limited to the western U.S.) ranged from 7 to 11 kg N/ha-yr. The deposition metric ranges were generally similar for the species for which survival was positively associated with the metric (across full range or at the median). Of the 39 species for which growth was significantly associated with N deposition, the association was negative across the full range for two species (with sample sites predominantly in the Atlantic coastal pine barrens and northern plains and forests, respectively). The median deposition across sites for these two were nine and ten kg N/ha-yr (Appendix 5B, Figure 5B-5 and Attachment 2). The median deposition values for the two other species with hump shaped

functions that were negative at the median were seven and eight kg N/ha-yr, respectively (Appendix 5B, Figure 5B-5).

Observational studies newly available in this review include two smaller studies in the Adirondacks of New York that investigated relationships of forest plot characteristics with N and S deposition metrics. These locations are well documented to have received appreciable acidic deposition over the past several decades. The studies report negative associations of forest health metrics with N and/or S deposition metrics (see Appendix 5B, Table 5B-2). These include the study by Sullivan et al. (2013), in which mean growth rates of sugar maple were positively correlated with exchangeable Ca and base saturation at the watershed level, indicating the influence of these soil acidification indicators. Also, newly available in this review are studies that analyzed potential for associations of tree growth of sensitive species with temporal changes in SO_X and/or NO_X emissions. For example, a study by Soulé (2011) reported increased red spruce growth in North Carolina to be associated with reductions in emissions of SO_X and N oxides from utilities in the southeastern U.S., among other factors, over the period from 1974 to 2007 (Soulé, 2011; ISA, Appendix 5, section 5.5.1).

Another observational study newly available in this review documented recovery of a stand of eastern redcedar (in the Appalachian Mountains of West Virginia) from historical S pollution using an analysis of tree ring chronology from 1909 to 2008, and a multivariate correlation analysis involving historical climate variables, atmospheric CO_2 concentrations and U.S. emissions estimates for SO_2 and N oxides (ISA, Appendix 5, p. 5-18; Thomas et al., 2013). Tree growth has increased significantly since 1970 and the analysis indicates it is explained by increases in atmospheric CO_2 and NO_X emissions and reductions in SO_2 emissions (ISA, Appendix 5, section 5.2.1.3; Thomas et al., 2013). The authors described the response as an indirect result of reductions in acid deposition, while other researchers have suggested that, given the speed of the response, it may more likely be related to reduced gaseous SO_2 than acid deposition (ISA, Appendix 5, section 5.2.1.3; Schaberg et al., 2014).

Table 5-9. Tree effects and associated S/N deposition levels from observational studies using USFS/FIA data.

Endpoint, Species, Location	Deposition/Addition	Reference
---------- S Deposition Metric Analyses ----------		
Survival in 7 of 10 species' groups in eastern and central U.S. negatively associated with SO_4^{2-} deposition	SO_4^{2-} wet deposition estimates (average, 1994-2005) varied 4 to 30 kg S/ha-yr across all sites.	Dietze and Moorcroft (2011)
Survival in 40 species across U.S. was negatively associated with S deposition estimates.	Median average S deposition estimates (2000-16) for these species: 3[A] to 12 kg S/ha-yr.	Horn et al. (2018)
Growth in 31 species across U.S. was negatively associated with S deposition estimates.	Median S deposition estimates for these species varied 4[A] to 12 kg S/ha-yr, when western species are excluded.	
---------- N Deposition Metric Analyses ----------		
Mortality in 1 species' group in eastern/central U.S. positively associated with NO_3^- deposition Mortality in 9 of 10 species' groups in eastern and central U.S. negatively associated with NO_3^- deposition (reduced mortality with increased NO_3^-)	NO_3^- wet deposition estimates (average, 1994-2005) varied from 6 to 16 kg N/ha-yr across all sites analyzed	Dietze and Moorcroft (2011)
Survival of 8 species negatively associated with N deposition. Survival of 3 species positively associated with N deposition. Growth of 3 species (all conifers) negatively associated with N deposition, Growth of 11 of 24 species positively associated with N deposition,	Estimates of average N deposition across the full set of study sites ranged from 3 to 11 kg N/ha-yr for the period 2000-2004.	Thomas et al. (2010)
Survival of 6 species was negatively associated with N deposition across deposition ranges Survival of 21 other species (2 limited to the West), with hump-shape associations, also negatively associated with N deposition at median deposition across species' sites. Survival of one species positively associated with N deposition across deposition range Survival of 4 other species, with hump-shape associations, also positively associated with N deposition at median deposition for species' sites.	For species with negative associations, median N deposition estimates varied from 8 to 11 kg N/ha-yr. For 19 species with negative association at median deposition, western species excluded, median N deposition varied 7 to 12 kg N/ha-yr. For species with positive association, median N deposition estimate was 11 kg N/ha-yr. For species with positive association at median deposition, median N deposition varied from 7 to 12 kg N/ha-yr.	Horn et al. (2018)
Growth of 2 species was negatively associated with N deposition across all species' sites. Growth of 2 other species (with hump-shape associations) also negatively associated with N deposition at the median deposition across sites Growth of 20 species (17 nonwestern species) was positively associated with N deposition across all species' sites. Growth of 15 other species with hump-shape associations (14 nonwestern species) was also positively associated with N deposition at the median deposition across those species' sites.	The median average deposition estimates for the measurement interval (during 2000-16) varied from 9 and 10 kg N/ha-yr. The median estimates for the other 2 species were 7 and 8. The 17 nonwestern species assessed at sites for which the median average deposition estimate for the measurement interval (during 2000-16) varied from 7 to 12 kg N/ha-yr. The median estimates for the other 14 nonwestern species were 7 to 11 kg N/ha-yr.	

[A] The two values below 5 kg S/ha-yr were for species with 60-80% of samples from the Northern Forests ecoregion.
Details of information summarized here are provided in Appendix 5B, section 5B.2.2.3 and Tables 5B-2 and 5B-6.

5.3.3 Other Effects

The studies available that may inform consideration of S or N deposition levels of potential interest for deposition-related effects on terrestrial biota other than trees include both addition experiments and observational or gradient studies. In addition to effects on individual species, these studies often report metrics related to changes in communities of particular plant or lichen populations. Information from both types of studies and with regard to species-level or community-level effects is discussed in the subsections below. The focus in these studies, however, is predominantly on N deposition.

5.3.3.1 Effects on Herbs and Shrubs

Observational/Gradient Studies

Since the 2012 review, new observational studies have investigated relationships between deposition and community composition for understory plants. One of the largest studies, Simkin et al. (2016), investigated relationships between species richness (number of species) of herbaceous plants[14] and values of a N deposition metric at more than 15,000 forest, woodland, shrubland and grassland sites across the U.S. (Appendix 5B, section 5B.3.2). The study grouped the sites into open- or closed-canopy sites, with forest sites falling into the closed-canopy category and the rest in open-canopy. The data for sites in each of the two categories were analyzed for relationships of species richness (number of herbaceous species) with values of the N deposition metric, soil pH, temperature, and precipitation (Simkin et al., 2016). The species richness assessments were conducted across a 23-year period (1990-2013) by multiple researchers, at sites clustered most prominently in portions of the 14-state study area, e.g., MN, WA, OR, VA, NC and SC (Appendix 5B, Figure 5B-13). The N deposition metric for each site was a 10-year average of dry N deposition (2002-2011) added to a 27-year average (1985-2011) of wet deposition (Simkin et al., 2016; Appendix 5B, section 5B.3.2).

Different relationships among the analyzed factors were observed for the two categories of sites, with a hump-shaped relationship of species richness with the deposition metric at open-canopy sites and a strong influence of soil pH at the closed-canopy (forest) sites (Simkin et al., 2016).

- At open-canopy sites, the association of herbaceous species richness with the N deposition metric was somewhat dependent on soil pH, precipitation and temperature. Herbaceous species richness was positively associated with the N deposition metric at the lower end of the deposition range and negatively associated with N deposition at the higher end of the deposition range, on average for metric values above 8.7 kg N/ha-yr (Simkin et al., 2016).

[14] Herbaceous plants are nonwoody vascular plants, including annuals, biannual and perennials.

- At closed-canopy (forest) sites, the association of herbaceous species richness with the N deposition metric was highly dependent on soil pH. Across sites with acid soil pH at/above 4.5, species richness was negatively associated with N deposition metric values greater than 11.6 kg N/ha-yr, but among sites with basic soils there was no point in the data set at which N deposition had a negative effect on species richness (the analysis included deposition values up to ~20 kg N/ha-yr).

The long time period over which the N deposition estimates are averaged in this study provides for an N deposition metric generally representative of long-term N deposition over a time period of temporally changing rates, particularly in areas of the Midwest south to the Gulf and eastward (e.g., ISA, Appendix 2, section 2.7). The impact of the differing time periods for the wet versus dry deposition estimates, however, is unclear. Notably, the study did not consider potential roles for other pollutants with a potential influence on the observations, including ozone and S deposition. Overall, the study by Simkin et al. (2016) indicates an effect of N deposition on herbaceous species richness, with a number of uncertainties that limit interpretations regarding identification of specific deposition levels of potential concern with regard to impacts on herbaceous species number.

Studies in southern California, particularly in grassland or coastal sage scrub communities, have investigated the role of past N deposition in documented alterations of community composition and increases in the presence of invasive species (ISA, Appendix 6, section 6.3.6). In light of the changes in vegetation that have occurred in this area since the early 20th century, a recent study by Cox et al. (2014) utilized a landscape-level analysis in investigating the risk of coastal sage scrub communities converting to exotic annual grasslands and potential associations with N deposition. These analyses further considered the factors that might influence or facilitate community recovery. Results of these analyses indicated that recovery of coastal sage shrub communities[15] from exotic grass invasion was most likely in sites with N deposition below 11.0 kg N/ ha-yr (in 2002, based on CMAQ modeling) and that had experienced relatively low invasion (Cox et al., 2014).

Experimental Addition Studies

Several addition studies have focused on California coastal sage scrub communities (ISA, Appendix 6, section 6.3.6). A study of 13 years of 50 kg N/ha-yr additions reported no significant effects on plant cover for the first 11 years of the 13-year period (ISA, Appendix 6, p. 6-81; Appendix 5B, Table 5B-7). Community composition was changed after five years, reflecting changes in the relative abundance of dominant shrubs, and in the 11th through 13th years, increases in an exotic plant and decreases in one of the native shrubs were reported (Vourlitis, 2017; Vourlitis and Pasquini, 2009).

[15] Coastal sage scrub is a shrubland community that occurs in Mediterranean-climate areas in southern California.

Experimental addition experiments have also reported variable relationships between N additions and impacts for herb or shrub communities (ISA, Appendix 6, section 6.3; Appendix 5B, section 5B.3.1). For example, a study by Bowman et al. (2012) in a dry sedge meadow in Colorado reported no shifts in species richness or diversity in response to N additions of 5, 10 and 30 kg/ha-yr, but also found increases in cover of one species (*Carex rupestris*) that ranged from 34 to 125% across the treatments (ISA, Appendix 6, section 6.3.4). Changes in the relative abundance of this species was the authors' basis for their CL estimate of 4 kg N/ha-yr.

At Joshua Tree National Park in the Mojave desert of California, non-native grass biomass increased significantly at three of the four study sites receiving 30 kg N/ha-yr for two years but experienced no significant change with an addition of 5 kg N/ha-yr (Allen et al., 2009). No significant change in community composition or species richness was reported in a semi-arid grassland in Utah in response to smaller additions of 2, 5 and 8 kg N/ha-yr over two years (ISA, Appendix 6, Table 6-21; McHugh et al., 2017). Much higher additions, of 10, 20, 34, 54 and 95 kg N/ha-yr over 23 years, in prairie grasslands resulted in reduced species richness. Ceasing those additions after 10 years resulted in recovery of species number back to control numbers[16] after 13 years (Clark and Tillman, 2008).

5.3.3.2 Effects on Lichen

The available information on N, S or PM exposure conditions associated with effects on lichen is primarily focused on nitrogen species (available evidence summarized in the ISA, Appendix 6, section 6.5.2). Limited information regarding effects of SO_X on lichen species is summarized in section 5.4.1 below, and the extent to which the effects relate to airborne SO_X or dry deposition of SO_2 (*vs* associated acidic deposition) is not clear. Somewhat similarly, section 5.4.2 below summarizes the available information regarding N oxides exposure conditions, including associated deposition, for which effects are reported on lichen species. We address below several observational or gradient studies newly available in this review that analyzed relationships between lichen community characteristics and N and/or S deposition metrics at sites in the Northeast and Northwest (Table 5B-9; ISA, Appendix 5, section 5.5.1 and Appendix 6, section 6.5).

In the northeastern U.S., past studies have concluded that in areas distant from industrial or urban sources, atmospheric deposition alters chemistry of tree bark (that provides substrate for lichen species) through acidification or eutrophication (Cleavitt et al., 2011; van Herk, 2001; ISA, Appendix 6, section 6.2.3.3). A study of relationships between lichen metrics and metrics for annual and cumulative N and S deposition from 2000 to 2013 at plots in four Class I areas of

[16] Species number changes in control plots contributed to this finding (Clark and Tillman, 2008; Isbell et al., 2013).

the northeastern U.S. reported that "lichen metrics were generally better correlated with cumulative deposition than annual deposition" (Cleavitt et al., 2015). Further, cumulative dry deposition of S yielded the best fit to decreases in thallus condition, poorer community-based S Index values, and absence of many S-sensitive species, indicating a stronger role for legacy of historical deposition than recent deposition patterns (Cleavitt et al., 2015). Across the years studied, annual S and N deposition in the four areas declined, from roughly 6-15 kg S/ha-yr to 3-6 kg S/ha-yr and from roughly 4-15 kg N/ha-yr to 3-8 kg N/ha-yr (Cleavitt et al., 2015, Figure 4).

Two more recent studies involve sites in the Northwest and focus on assessing relationships between metrics for lichen community composition and estimated N deposition. The study by Geiser et al. (2010) related lichen air scores assigned based on relative abundance of oligotrophic and eutrophic species in assessments (conducted from 1994 to 2002) to N deposition metric values (based on 1990-99 average N deposition). The authors identified a breakpoint between the third and fourth air scores which was associated with 33-43% fewer oligotrophic species and 3 to 4-fold more eutrophic species than sites with scores in the "best" bin; at sites reflecting this scoring breakpoint, total N deposition estimates ranged from 3 to 9 kg N/ha-yr (Geiser et al., 2010). Using a different score or index to characterize lichen communities (based on assessments 1993-2011), Root et al. (2015) analyzed particulate N estimated from speciated $PM_{2.5}$ monitoring data and throughfall N deposition estimated from lichen N content. Several aspects of these studies complicate interpretation of exposure conditions and identification of N deposition levels associated with particular risks to lichen communities. For example, the methods for utilizing N deposition differ from current commonly accepted methods. There is also uncertainty regarding the potential role of other unaccounted-for environmental factors (including ozone, SO_2, S deposition and historical air quality and associated deposition). There is uncertainty concerning the independence of any effect of deposition levels from residual effects of past N deposition. And there are few controlled N addition experiments that might augment or inform interpretation of the findings of observational/gradient studies (fumigation studies are summarized in section 5.4.2 below). Other studies in Europe and Canada have not reported such associations with relatively large N deposition gradients.

5.3.4 Summary: Key Findings and Associated Uncertainties

Key findings related to ambient air concentrations and S and N deposition levels associated with terrestrial effects discussed in prior sections are summarized below.

5.3.4.1 Deposition and Risks to Trees

Soil Acidification Analyses and Risk to Trees

Steady-state modeling analysis performed in the 2009 REA estimated annual amounts of acid deposition at or below which one of three BC:Al targets would be met in a 24-state area in which the acid-sensitive species, red spruce and sugar maple, occur. While the two least restrictive targets (BC:Al of 0.6 and 1) differed by less than a factor of two, the two most restrictive targets (BC:Al of 1 and 10) differed by a factor of 10. A range of acid deposition was estimated for each of the three targets. For a BC:Al target of 0.6, the range was 1237-2009 eq/ha-yr; for a BC:Al target of 1, the range was 892-1481 eq/ha-yr; and for a BC:Al target of 10, the range was 487-910 eq/ha-yr. Estimates of total S and N deposition in regions of the U.S. for the 2019-2021 period appear to meet all but the most restrictive of these targets (e.g., section 2.5.3 above; ISA, Appendix 2, sections 2.6 and 2.7).

Uncertainties associated with these analyses include those associated with the limited dataset of laboratory-generated data on which the BC:Al targets are based. These data are derived from an array of studies of tree seedlings in artificial substrates and responses ranging from changes in plant tissue components to changes in biomass. In addition to the uncertainty associated with the basis for the BC:Al targets, there are uncertainties in the steady-state modeling parameters, most prominently those related to base cation weathering and acid-neutralizing capacity (2009 REA, section 4.3.9). As discussed in section 5.3.2.1 above, more recent publications have employed a new approach to estimating these parameters, including the weathering parameter, with reduced uncertainty. For the Pennsylvania study area where this was tested, a greater buffering capacity was estimated, and for a larger study area of the Northeast, the deposition estimates for the BC:Al target of 10 were slightly higher than those for the 2009 REA (Phelan et al., 2014; Duarte et al., 2013).

Tree Growth and Survival in Experimental Addition Studies

Experimental addition studies of S, or S plus N, with additions greater than 20 kg/ha-yr, have been performed in eastern locations and focused on a small set of species, including sugar maple, aspen, white spruce, yellow poplar, black cherry; these studies generally have not reported growth effects (Appendix 5B, section 5B.2.1). A study involving both S and N additions greater than 20 kg/ha-yr for each substance reported increased growth rate for sugar maple but not for the second species (Bethers et al., 2009), while another study of similar dosing of S and N reported reduced growth in three species after 10 years that resolved in two of the species after 22 years (Jensen et al., 2014). In both situations background deposition contributions were also appreciable (Appendix 5B, Table 5B-1).

Uncertainties associated with these analyses include the extent to which the studies reflect steady-state conditions. Given the variability in the durations across these studies and the relatively short durations for some (e.g., less than five years), it might be expected that steady-state conditions have not been reached, such that the S/N loading is within the buffering capacity of the soils. A related limitation of some of these studies is the lack of information regarding historic deposition at the study locations that might inform an understanding of the prior issue. However, many of the studies have assessed soil characteristics and soil acidification indicators, which also informs this issue.

With regard to N addition, the available studies have reported mixed results for growth and survival for several eastern species including oaks, spruce, maples and pines (Table 5B-1; Magill et al., 2004; McNulty et al., 2004; Pregitzer et al., 2008; and Wallace et al., 2007). Some studies have suggested that this variation in responses is related to the dominant mycorrhizal association of the species (e.g., Thomas et al., 2010). It is not clear the extent to which such findings may be influenced by species-specific sensitivities or soils and trees already impacted by historic deposition, or other environmental factors. Uncertainties for N addition experiments and interpretation of their results include this complexity, as well as the uncertainties identified above for S or S+N addition studies.

Observational/Gradient Studies of Tree Growth/ Survival

With regard to S deposition, the two large studies that analyzed growth and/or survival measurements in tree species at sites in the eastern U.S. or across the country report negative associations of tree survival and growth with the S deposition metric for nearly half the species individually and negative associations of tree survival for 9 of the 10 species' functional type groupings (Dietze and Moorcroft, 2011; Horn et al., 2018[17]). Interestingly, survival for the same 9 species groups was also negatively associated with long-term average ozone (Dietze and Moorcroft, 2011).

- The full range of average SO_4^{2-} deposition estimated for the 1994-2005 time period assessed by Dietze and Moorcroft (2011) for the eastern U.S. study area was 4 to 30 kg S $ha^{-1}yr^{-1}$.
- Median S deposition (2000-13) estimated at sites (measurement interval average [occurring within 2000-13]) of nonwestern species with negative associations with growth or survival ranged from 5 to 12 kg S $ha^{-1}yr^{-1}$, with few exceptions (Horn et al., 2018).

The S deposition metrics for the two studies were mean annual average deposition estimates for total S or sulfate (wet deposition) during different, but overlapping, time periods of

[17] The study by Horn et al. (2018) constrained the S analyses to preclude a positive association with S.

roughly 10-year durations. Additionally, S deposition in the U.S. across the full period of these studies (1994-2013) generally exhibited a consistent pattern of appreciable declines. Further, the study plots, particularly in the eastern U.S., have experienced decades of much higher S deposition in the past. The extent to which the differences in growth or survival across sites with different deposition estimates are influenced by historically higher deposition (e.g., versus the magnitude of the average over the measurement interval) is unknown. There are few available studies describing recovery of historically impacted sites (e.g., ISA, section IS.4.1, IS.5.1, IS.11.2).

Regarding N deposition, the three large studies that analyzed growth and/or survival measurements in tree species at sites in the northeastern or eastern U.S., or across the country, report associations of tree survival and growth with several N deposition metrics (Dietze and Moorcroft, 2011; Thomas et al., 2010; Horn et al., 2018).

- Estimates of average N deposition across the full set of sites analyzed by Thomas et al. (2010) in 19 states in the northeastern quadrant of the U.S. ranged from 3 to 11 kg N/ha-yr for the period 2000-2004.
- The full range of average NO_3^- deposition estimated for the 1994-2005 time period assessed by Dietze and Moorcroft (2011) for the eastern U.S. study area was 6 to 16 kg N $ha^{-1}yr^{-1}$.
- Median N deposition estimated (measurement interval average [falling within 2000-13]) at sites of nonwestern species for which associations with growth or survival were negative (either over full range or at median for species) ranged from 7 to 12 kg N $ha^{-1}yr^{-1}$ (Horn et al., 2018).
- Median N deposition estimated (measurement interval average [within 2000-13]) at sites of nonwestern species for which associations with growth or survival were positive (either over full range or at median for species) ranged from 7 to 12 kg N $ha^{-1}yr^{-1}$ (Horn et al., 2018).

The N deposition metrics for these three studies were mean annual average deposition estimates for total N or nitrate (wet deposition) during different, but overlapping, time periods that varied from 5 to more than 10 years and include areas that have experienced decades of much higher deposition. Further, N deposition during the combined time period (1994-2013) has changed appreciably at many sites across the country, with many areas experiencing declines and a few areas experiencing increases in deposition of some N species and in total N deposition.

In considering what can be drawn from these studies with regard to identification of deposition levels of potential concern for tree species effects, a number of uncertainties are recognized. For example, several factors with potential influence on tree growth and survival were not accounted for. For example, although ozone was analyzed in one of the three studies, soil characteristics and other factors with potential to impact tree growth and survival (other than climate) were not assessed, contributing uncertainty to their interpretations. Further, differences

in findings for the various species (or species' groups) may relate to differences in geographic distribution of sampling locations, which may contribute to differences in ranges of deposition history, geochemistry etc. Additionally, as noted above, the extent to which associations reflect the influence of historical deposition patterns and associated impact is unknown.

As summarized in Appendix 5B, Table 5B-6, there is a general similarity in findings among the studies, particularly of Horn et al. (2018) and Dietze and Moorcroft (2011), even though the time period and estimation approach for S and N deposition differ. Given the role of deposition in causing soil conditions that affect tree growth and survival, and a general similarity of spatial variation of recent deposition to historic deposition, an uncertainty associated with quantitative interpretation of these studies is the extent to which the similarity in the two studies' finding may indicate the two different metrics to both be reflecting geographic variation in impacts stemming from historic deposition. Although the spatial patterns are somewhat similar, the magnitudes of S and N deposition in the U.S. has changed appreciably over the time period covered by these studies (e.g., Appendix 5B, Figures 5B-9 through 5B-12). The appreciable differences in magnitude across the time periods also contribute uncertainty to interpretations related to specific magnitudes of deposition associated with patterns of tree growth and survival.

5.3.4.2 Deposition Studies of Herbs, Shrubs and Lichens

The available studies that may inform our understanding of exposure conditions, including N deposition levels, of potential risk to herb, shrub and lichen communities include observational or gradient studies and experimental addition conducted in different parts of the U.S. Among the studies of plant communities are observational studies of herbaceous species richness at sites in a multi-state study area and of grassland or coastal sage scrub communities in southern California, and experimental addition experiments in several western herb or shrub ecosystems. The experimental addition studies indicate effects on community composition associated with annual N additions of 10 kg N/ha-yr (in the context of background deposition on the order of 6 kg N/ha-yr) and higher (section 5.3.3.1 above). Experiments involving additions of 5 kg N/ha-yr variously reported no response or increased cover for one species (in context of background deposition estimated at 5 kg N/ha-yr). The landscape-level analysis of coastal sage scrub community history in southern California observed a greater likelihood of recovery of sites with relatively low invasion of exotic invasive grasses when the N deposition metric level was below 11 kg N/ha-yr. Lastly, the multi-state analysis of herbaceous species richness reported a negative association with N deposition metric values above 8.7 kg N/ha-yr at open-canopy sites and above 11.6 kg N/ha-yr at forest sites with acidic soil pH at or above 4.5.

Limitations and associated uncertainties vary between the two types of studies (experimental addition and observational). Both are limited with regard to consideration of the

impacts of long-term deposition. While there are some experimental addition studies lasting more than 20 years, many are for fewer than 10 years. Additionally, such studies are necessarily limited with regard to the number and diversity of species and ecosystems that can be analyzed. In the case of observational studies, the many decades-long history of S and N deposition, as well as elevated levels of airborne pollutants, including ozone and nitrogen oxides, in the U.S. is their backdrop, and its influence on associations observed with more recent deposition metrics is generally unaccounted for. Further, given the very nature of observational studies as occurring in real time, there is uncertainty associated with characterization, including quantification, of the particular exposure conditions that may be eliciting patterns of ecosystem metrics observed.

The few studies of lichen species diversity and deposition-related metrics, while contributing to the evidence that relates deposition, including acidic deposition in eastern locations, to relative abundance of different lichen species, are more limited with regard to the extent that they inform an understanding of specific exposure conditions in terms of deposition levels that may be of concern. As summarized in section 5.3.3.2 above, a number of factors limit such interpretations of the currently available studies. These factors include uncertainties related to the methods employed for utilizing estimates of N deposition, the potential role of other unaccounted-for environmental factors (including ozone, SO_2, S deposition and historical air quality and associated deposition), and uncertainty concerning the independence of any effect of deposition levels from residual effects of past patterns of deposition. We additionally note the summary in section 5.4.2 below, of information on exposure conditions associated with effects on lichen species of oxides of N such as HNO_3.

5.4 OTHER EFFECTS OF OXIDES OF N AND S AND OF PM IN AMBIENT AIR

The evidence related to exposure conditions for other effects of SO_X, N oxides and PM in ambient air includes concentrations of SO_2 and NO_2 associated with effects on plants, concentrations of NO_2 and HNO_3 associated with effects on plants and lichens and quite high concentrations of PM that affect plant photosynthesis. The PM effects described in the evidence are nearly all related to deposition. With regard to oxides of N and S, we note that some effects described may be related to dry deposition of SO_2 and HNO_3 onto plant and lichen surfaces. These exposure pathways would be captured in observational studies and could also be captured in some fumigation experiments.

With regard to SO_2 the evidence comes from an array of studies, primarily field studies for the higher concentrations associated with visible foliar injury and laboratory studies for other effects. With regard to oxides of N, the evidence indicates that effects on plants and lichens occur at much lower exposures to HNO_3 (than to NO_2). The laboratory and field studies of

oxides of N vary with regard to their limitations; field studies are limited with regard to identification of threshold exposures for the reported effects and uncertainties associated with controlled experiments include whether the conditions under which the observed effects occur would be expected in the field. With regard to the latter, as described in section 5.4.2 below, the elevated concentrations of NO_2 and HNO_3 in the Los Angeles area in the 1970s-90s is well documented as is the decline of lichen species in the Los Angeles Basin during that time.[18]

5.4.1 Sulfur Oxides

As summarized in section 4.1 above, other welfare effects of SO_X in ambient air include effects on vegetation, such as foliar injury, depressed photosynthesis and reduced growth or yield. Within the recently available information are observational studies reporting increased tree growth in association with reductions in SO_2 emissions. These studies, however, do not generally report the SO_2 concentrations in ambient air or account for the influence of changes in concentrations of co-occurring pollutants such as ozone (ISA, Appendix 3, section 3.2). The available data that include exposure concentrations are drawn from experimental studies or observational studies in areas near sources, with the most studied effect being visible foliar injury to various trees and crops (ISA, Appendix 3, section 3.2; 1982 AQCD, section 8.3). Based on controlled laboratory exposures in some early studies (assessed in the 1982 AQCD), concentrations greater than approximately 0.3 ppm SO_2 for a few hours were required to induce slight injury in seedlings of several pine species, with sensitive species exposed in conducive conditions being more likely to show visible injury (1982 AQCD, section 8.3). With regard to foliar injury, the current ISA states there to be "no clear evidence of acute foliar injury below the level of the current standard" (ISA, p. IS-37). For effects on plant productivity and growth, studies described in the 1982 AQCD that involve experimental exposures in the laboratory have reported depressed photosynthesis by 20% or more from one week of continuous exposure to 0.5 ppm SO_2 for 3 weeks to 3 hours/day at 0.5 ppm. Few studies report yield effects from acute exposures, with the available ones reporting relatively high concentrations. For example, a study with soybeans reported statistically significant yield reductions (more than 10%) after a 4.2-hour exposure to concentrations greater than 1 ppm SO_2 (1982 AQCD, section 8.3).

The evidence presented in the ISA also includes effects on lichen species, such as those reported in laboratory fumigation experiments that have assessed effects on photosynthesis and other functions in a few lichen groups (ISA, Appendix 3, section 3.2). For example, a study of

[18] For example, concentrations of HNO_3 reported in forested areas of California in the 1980s ranged up to 33 ug/m^3, and annual average NO_2 concentrations in the Los Angeles area ranged from 0.078 ppm in 1979 to 0.053 ppm in the early 1990s (section 5.4.2). Ambient air concentrations of HNO_3 in the Los Angeles metropolitan area have declined markedly, as can be seen from Figure 2-23 (in section 2.4.1), which compares concentrations at CASTNET monitoring sites between 2019 and 1996.

two lichens in Spain by Sanz et al. (1992) found photosynthesis to be significantly depressed in the more sensitive species after 4 to 6 hours at 0.1 ppm SO_2, with recovery occurring within 2 hours following exposure. After shorter exposures to 0.25, 0.5 and 0.9 ppm, photosynthesis recovered within two weeks. After exposures to higher concentrations, photosynthesis in the more sensitive species was significantly reduced and did not recover. The second species tested was appreciably less sensitive to SO_2 exposure (Sanz et al., 1992).

5.4.2 Nitrogen Oxides

The direct welfare effects of N oxides in ambient air include effects on plants and lichens. For plants, studies reported in the ISA did not report effects on photosynthesis and growth resulting from exposures of NO_2 concentrations below 0.1 ppm (ISA, Appendix 3, section 3.3). For example, five days of 7-hour/day exposures of soybean plants reduced photosynthesis at 0.5 ppm and increased photosynthesis at 0.2 ppm NO_2 (ISA, Appendix 3, section 3.3). Exposures to 0.1 ppm NO_2 continuously for eight weeks and for six hours/day over 28 days elicited reduced growth of Kentucky blue grass and seedlings of three tree species, respectively (ISA, Appendix 3, section 3.3). A study of five California native grasses and forbs exposed to 0.03 ppm NO_2 continuously for 16 weeks found no significant effects on shoot or root biomass, photosynthesis or stomatal conductance (ISA, Appendix 3, section 3.3). Visible foliar injury has not been reported to occur with NO_2 exposure concentrations below 0.2 ppm except for exposures of durations lasting 100 hours or longer (ISA, Appendix 3, section 3.3). The ISA notes that for most plants, "injury occurred in less than 1 day only when concentrations exceeded 1 ppm" (ISA, Appendix 3, p. 3-10). The information is more limited with regard to exposures to other oxides of N. A study involving three 4-hr exposures to 30 ppb PAN on alternating days in a laboratory setting reported statistically significant reduction in growth of kidney bean and petunia plants (ISA, Appendix 3, section 3.3).

The evidence for HNO_3 includes controlled exposure studies describing foliar effects on several tree species. For example, 12-hour exposures to 50 ppb HNO_3 (~75 µg/m^3) in light, and to 200 ppb (~530 µg/ m^3) in darkness, affected ponderosa pine needle cuticle (ISA, Appendix 3, section 3.4). Nitric acid has also been found to deposit on and bind to the leaf or needle surfaces. Continuous 32- or 33-day chamber exposure of ponderosa pine, white fir, California black oak and canyon live oak to 24-hour average HNO_3 concentrations generally ranging from 10 to 18 µg/m3 (moderate treatment) or 18 to 42 µg/ m^3 (high treatment), with the average of the highest 10% of concentrations generally ranging from 18 to 42 µg/ m^3 (30-60 µg/ m^3 peak) or 89 to 155 µg/ m^3 (95-160 µg/ m^3 peak), resulted in damage to foliar surfaces of the 1 to 2-year old plants (ISA, Appendix 3, section 3.4; Padgett et al., 2009). The moderate treatment reflects exposure concentrations observed during some summer periods in the Los Angeles Basin in the mid-

1980s, including a high HNO_3 concentration of 33 ug/ m^3 in August 1986 (Padgett et al., 2009; Bytnerowicz and Fenn, 1996), when annual average NO_2 concentrations in the Basin ranged up to 0.058 ppm (U.S. EPA, 1987).

The available evidence for lichens includes a recent laboratory study, involving daily HNO_3 exposures for 18 to 78 days, with daily peaks near 50 ppb (~75 µg/ m^3) that reported decreased photosynthesis, among other effects (ISA, Appendix 6, section 6.2.3.3; Riddell et al., 2012). Based on studies extending back to the 1980s, HNO_3 has been suspected to have had an important role in the dramatic declines of lichen communities that occurred in the Los Angeles basin (ISA, Appendix 3, section 3.4; Nash and Sigal, 1999; Riddell et al., 2008; Riddell et al., 2012). For example, lichen transplanted from clean air habitats to analogous habitats in the Los Angeles basin in 1985-86 were affected in a few weeks by mortality and appreciable accumulation of H^+ and NO_3^- (ISA, Appendix 3, section 3.4; Boonpragob et al., 1989). As described in Appendix 5B, section 5B.4.1, the Los Angeles metropolitan area experienced NO_2 concentrations well in excess of the NO_2 secondary standard during this period. For example, annual average NO_2 concentrations in Los Angeles ranged up to 0.078 ppm in 1979 and remained above the standard level of 0.053 ppm into the early 1990s (Appendix 5B, section 5B.4.1). The magnitude and spatial extent of declines over the last several decades, in both dry deposition of HNO_3 and annual average HNO_3 concentration in this area and nationally, are illustrated in Figure 2-23 above (and the ISA, Appendix 2, Figure 2-60). As assessed in the ISA, the evidence indicates NO_2, and particularly, HNO_3, as "the main agent of decline of lichen in the Los Angeles basin" (ISA, Appendix 3, p. 3-15), thus indicating a role for the elevated concentrations of nitrogen oxides documented during the 1970s to 1990s (and likely also occurring earlier). More recent studies indicate variation in eutrophic lichen abundance to be associated with variation in N deposition metrics (ISA, Appendix 6, section 6.2.3.3). The extent to which these associations are influenced by residual impacts of historic air quality is unclear.

5.4.3 Particulate Matter

The extent to which quantitative information is available for airborne PM concentrations associated with ecological effects varies for the various types of effects. The concentrations at which PM has been reported to affect vegetation (e.g., through effects on leaf surfaces, which may affect function or through effects on gas exchange processes) are generally higher than those associated with conditions meeting the current standards and may be focused on specific particulate chemicals rather than on the mixture of chemicals in PM occurring in ambient air. For example, reduced photosynthesis has been reported for rice plants experiencing fly ash particle deposition of 0.5 to 1.5 g/m^2-day, which corresponds to loading greater than 1000 kg/ha-yr (ISA, Appendix 15, sections 15.4.3 and 15.4.6). Studies involving ambient air PM have generally

involved conditions that would not be expected to meet the current secondary standards, e.g., polluted locations in India or Argentina (ISA, Appendix 15, sections 15.4.3 and 15.4.4). Studies in the U.S. that have looked at the effects of airborne PM on plant reproduction near roadway locations in the U.S. have not reported a relationship between PM concentrations and pollen germination (ISA, Appendix 15, section 15.4.6). In summary, little information is available on welfare effects of airborne PM in exposure conditions likely to meet the current standards, and that which is available does not indicate effects to occur under those conditions.

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6 RELATIONSHIPS OF DEPOSITION TO AIR QUALITY METRICS

6.1 OVERVIEW

To address the framing questions that guide the scope of this review, this section focuses on characterizing the relationship between deposition of S and N compounds and air quality concentrations of S oxides, N oxides and $PM_{2.5}$. This characterization is a key aspect of the approach taken in this PA for assessing deposition-related effects and the adequacy of the current secondary standards, as summarized in section 3.2 (Figure 6-1).

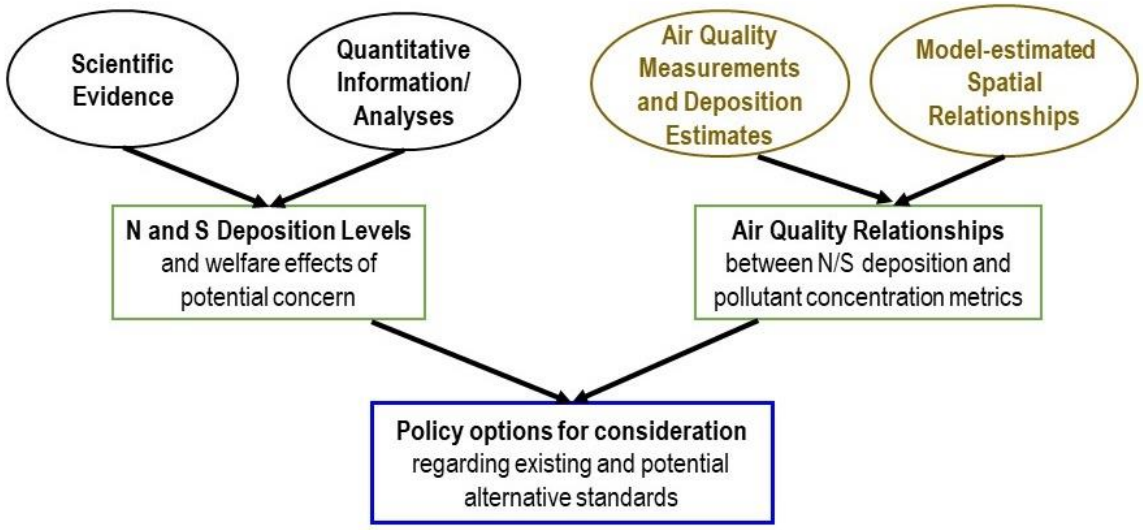

Figure 6-1. General approach for assessing the currently available information with regard to consideration of protection provided against deposition-related ecological effects on the public welfare.

While the ecological effects examined in this review include those associated with deposition of S and N, the NAAQS are set in terms of pollutant concentrations. The goal of this section is to examine the relationship between atmospheric deposition of S and N with ambient air concentrations of criteria air pollutants, over a range of conditions (e.g., pollutants, regions, time periods). An evaluation of this relationship can then help inform how changes in air concentrations, and the emissions from which they result, could lead to changes in the amounts of S and N deposited. This understanding can then help inform decisions on which air quality metric(s) to consider for a standard designed to protect against S and N deposition-related effects.

However, there are fundamental difficulties in establishing quantitative relationships between air quality concentrations and deposition, stemming from the complex atmospheric processes that govern the lifecycle of pollutant emissions to eventual deposition to the surface. As described in more detail below, multiple pollutants can contribute to S and N deposition. Additionally, there are multiple deposition pathways (i.e., dry deposition and wet deposition) that can influence the spatial and temporal scales at which deposition occurs and which can vary by pollutant and pollutant phase. Further, deposition measurements are relatively limited and are largely available only for wet deposition. There are relatively few sites that collect collocated air concentration and pollutant deposition data. We can use air quality models to estimate deposition where there is a lack of monitors, but these models are limited by our understanding of the processes that influence deposition and have their own uncertainties and error.

6.1.1 Review of the Processes Affecting Atmospheric Deposition

Atmospheric deposition occurs when a pollutant is transferred from the atmosphere to the earth's surface through dry deposition (settling onto the surface through direct contact) or wet deposition (aqueous uptake, or scavenging by rain, clouds, snow, or fog). There are a variety of factors that determine how much of the pollutant is deposited. For example, the rate at which a pollutant dry deposits (i.e., the dry deposition velocity) depends on the physical properties of the chemical compound, meteorological conditions, and the properties of the surface to which the pollutant is being deposited. The rate of wet deposition is influenced by the chemical and physical properties of the pollutant, the precipitation rate, and the vertical distribution of the pollutant in the atmosphere.

For dry deposition, the physical properties of a chemical compound can be especially important in determining its deposition velocity and the rates of deposition can vary substantially across the nitrogen and sulfur containing compounds in the atmosphere (ISA, Appendix 2, section 2.5). For example, NO_2 can oxidize to form nitric acid (HNO_3), which deposits more readily than NO_2. However, HNO_3 can also partition into the particle phase in the presence of ammonia to form ammonium nitrate (NH_4NO_3) which is deposited primarily through wet processes. Fine particles have a slower dry deposition velocity and generally remain in the atmosphere longer than gases, i.e., days for nitrate $PM_{2.5}$ versus hours for HNO_3 (Table 6-1; Xu and Penner, 2012). On the other hand, HNO_3 can also absorb onto larger, coarse particles, whose dry deposition velocity is faster than $PM_{2.5}$ (e.g., Zhang et al., 2001; Emerson et al., 2020). Thus, differences in the chemical and physical forms of nitrogen and sulfur contribute variability in the rate of deposition and in the relationship between total air concentrations and atmospheric deposition. Furthermore, the dry deposition velocity is influenced by meteorological conditions and interaction with the deposition surface properties (for example, the density of leaf area).

Meteorological factors such as wind speed, humidity, atmospheric stability, and temperature all affect the rate of settling for particles and gases. There are also micrometeorological parameters that impact dry deposition rates of particles, such as friction velocity, roughness height, and surface wetness (ISA, Appendix 2, section 2.5.2; Wesely, 2007).

For wet deposition, the amount of nitrogen and sulfur transferred to cloud water and falling precipitation is largely driven by the air concentration. The vertical distribution of the pollutant can influence deposition amounts. Air pollutant concentrations have historically been measured near ground level where health and ecological effects occur. Sulfur and nitrogen higher in the troposphere can be scavenged by clouds and falling precipitation via wet deposition. While dry deposition is directly related to the ground-level concentration, it is important to recognize that wet deposition is affected by concentrations throughout the troposphere (ISA, Appendix 2, section 2.5.2) which highlights the role of atmospheric transport of pollution.

6.1.2 Scales of Influence for Depositional Pathways Amid a Changing Chemical Environment

Near emission sources, where there is an abundance of nitrogen and sulfur compounds in the gas phase prior to chemical conversion to products like $PM_{2.5}$, it is anticipated that dry deposition will have a relatively larger influence over total deposition. On the other hand, wet deposition is expected to have a larger influence downwind following transport and transformation of gaseous species into longer-lived aerosol forms. Changes in chemical regimes and in the sensitivity of $PM_{2.5}$ formation may affect when, where and how pollution deposits. For example, NO_X and SO_X emission reductions over the past several decades have shifted the sensitivity of $PM_{2.5}$ toward an acid gas limitation, such that a greater portion of emitted NH_3 now remains in the gas phase. This will reduce the atmospheric lifetime of NH_X and increase the influence of NH_3 dry deposition on local scales.

Atmospheric humidity and the frequency of precipitation is also influential. For example, desert areas receive very little precipitation and hence contribution from wet deposition is low. Much of the western U.S. has drought years that result in very low wet deposition amounts, followed by years with higher amounts of precipitation and higher wet deposition. The eastern U.S. has less interannual variability in rainfall. The frequency of precipitation affects the relative contributions of wet and dry deposition and therefore can also cause variability in the relationship between ground-level air concentrations and deposition.

Figure 6-2 is a simplified illustration of the primary pathways by which different pollutants contribute to total deposition of S and N and is intended to summarize the discussion above. This schematic differentiates the role of criteria pollutants and their indicator compounds from the non-criteria pollutants (i.e., ammonia). Additionally, this illustration highlights the

primary loss pathways for each pollutant on a generalized national basis. It is an overview schematic focused on the relationships between the criteria pollutants under consideration, and in this case does not illustrate the role for meteorology or of other atmospheric constituents (e.g., organic species). Table 6-1 provides a summary of the expected atmospheric lifetimes of various N and S containing pollutants based on a literature review.

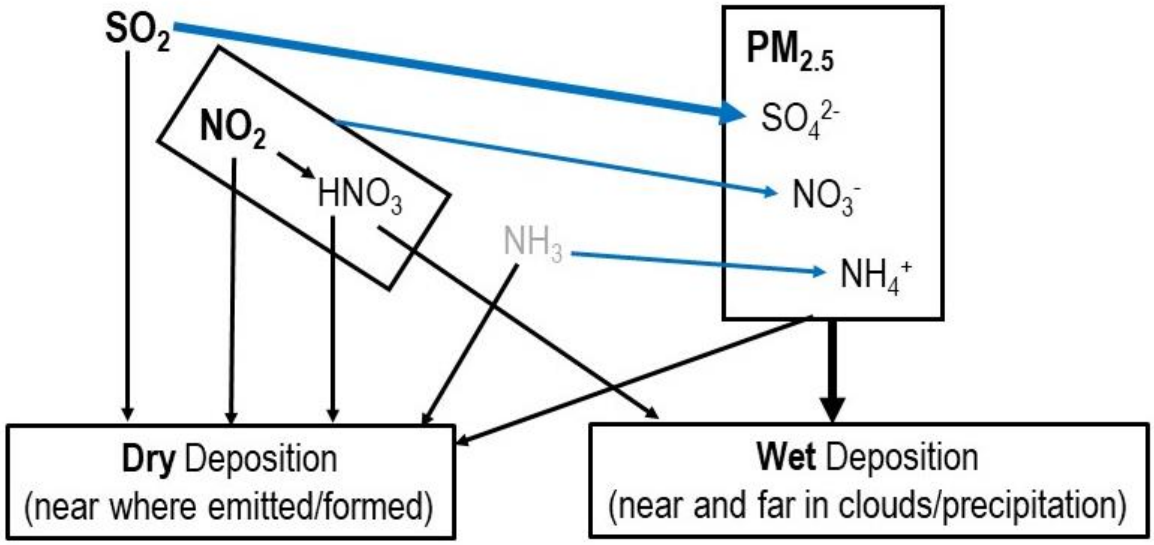

Figure 6-2. Primary pathways by which emitted pollutants are transformed and deposited. Blue arrows indicate that chemical transformation can occur during transport. Bold arrow indicates primary loss mechanism pathway. Bolded pollutants are NAAQS indicators; grey font is for non-criteria pollutant (ammonia).

Table 6-1. Estimated atmospheric lifetimes of S- and N-containing species based on literature review.

Pollutant	Lifetime	Conditions	Reference
SO_2	19 ± 7 hours	Summer, Eastern USA	Lee et al., 2011
	58 ± 20 hours	Winter, Eastern USA	Lee et al., 2011
$NO + NO_2$	4 to 21 hours	Surface	Shah et al., 2020; Liu et al., 2016; Laughner and Cohen, 2019
	6 hours	Nighttime, Winter, Eastern USA	Kenagy et al., 2018
	29 hours	Daytime, Winter, Eastern USA	Kenagy et al., 2018
NH_3	~11 hours	Global troposphere	Xu and Penner, 2012
	1.5 to 12 hours	Canadian wildfire plume	Adams et al., 2019
HNO_3	1.5 to 12 hours	Power plant plumes	Neuman et al., 2004
	4.8 days	Global troposphere	Xu and Penner, 2012
NH_4 aerosol	3.2 days	Global troposphere	Xu and Penner, 2012
NO_3 aerosol	3.9 days	Global troposphere	Xu and Penner, 2012
SO_4 aerosol	4.6 days	Global troposphere	Banks et al., 2022
Isoprene nitrates	One to a few hours	Forests	Lockwood et al., 2010; Paulot et al., 2009
First-generation organic nitrates	~2 hours	Southeastern USA	Zare et al., 2018
Second-generation organic nitrates	3 hours to 5 days	Eastern USA	Mao et al. 2013; Zare et al., 2018

6.1.3 Analyses in the 2012 Review (Transference Ratio)

The PA for the 2012 review of the NAAQS for N and S oxides introduced the term "transference ratio" which was defined as the ratio of deposition to air concentration (2011 PA, section 7.2.3). This was calculated from annual average values and was spatially averaged over ecoregions that spanned distances on the order of 10,000 km^2. While generally capturing the average relationship between air concentrations and atmospheric deposition over larger areas of the country, the transference ratio approach had some important limitations, especially at local scales. For example, the transference ratio approach did not capture the spatial variability across an area due to the proximity to sources, chemical composition, frequency of precipitation, and vertical distribution of nitrogen and sulfur (ISA, Appendix 2, section 2.5.2.4). Studies completed since the 2011 PA have examined how the use of different models to calculate concentration and deposition can yield very different estimates of the transference ratio, despite having comparable error statistics when compared to measurements of air concentrations and wet deposition (ISA, Appendix 2, section 2.5.2.4). As noted earlier, there are fundamental difficulties in establishing quantitative relationships between air quality concentrations and deposition; the analyses in this review are designed to go beyond the transference ratio by considering these relationships across multiple geographic scales and through multiple analytical approaches.

6.1.4 Organization of this Chapter

The challenges noted above have been considered in the analyses performed in the current review to investigate the relationships between criteria pollutant concentrations and deposition rates. Section 6.2 below describes four separate analyses completed as part of this review: a review of recent trends in air pollutant concentrations and deposition (section 6.2.1), an assessment of concentrations and deposition amounts at collocated sites (specific Class I areas in section 6.2.2; national SLAMS monitors in section 6.2.3), and a trajectory-based modeling analysis that enables an assessment of the association between upwind concentrations and downwind deposition (section 6.2.4). Key uncertainties associated with these analyses are characterized in section 6.3, and the key observations from this work are summarized in section 6.4.

6.2 RELATING AIR QUALITY TO DEPOSITION

This PA recognizes the limitations mentioned above, and as described in Chapter 2, also recognizes that emissions, air concentrations, and deposition have all declined for sulfur and oxidized nitrogen in recent years. This assessment examines the historical record of observations, multi-decadal CMAQ simulations, and hybrid model-measurement TDep estimates to assess the relationship between air concentrations of a specific compound, or combination of compounds, and estimates of N and S deposition in specific locations.

6.2.1 Historical Trend Analyses of Emissions, Concentrations, and Deposition

Total anthropogenic NO_X emissions (as represented by emissions of NO and NO_2) have trended strongly downward across the U.S. between 2002 and 2022 (Figure 6-3). Nationwide estimates indicate a 70% decrease in these emissions over this time as a result of multiple regulatory programs implemented over the past two decades, as well as changes in economic conditions and domestic energy production. This trend is an opportunity to consider how changes in emissions, air concentrations and deposition levels are correlated. As seen in Figure 6-3, the overall decrease in emissions of NO and NO_2 has been driven primarily by decreases from the three largest emissions sectors (i.e., highway vehicles, stationary fuel combustion, and non-road mobile sources). Specifically, compared to the 2002 start year, estimates for 2022 (from the 2020 NEI) indicate an 84% reduction in emissions from highway vehicles, a 68% reduction in emissions from stationary fuel combustion, and a 54% reduction in emissions from non-road mobile sources. Similar to NO_X, and for many of the same reasons, SO_2 emissions have also declined significantly since 2002. Figure 6-4 illustrates the emissions changes over the 2002-2022 period. The data show an 87% decrease in total SO_2 emissions over the period, driven by reductions of 91% in EGU emissions and 96% in mobile source emissions.

In contrast with NO_X and SO_2 emission trends, the annual rate of NH_3 emissions nationally has increased by about 15-20% since 2002 (Figure 6-5). The magnitude and direction of the NH_3 emissions change varies with location across the U.S. and is partly due to growth in agricultural sources of NH_3, which are largely unregulated at the national scale. Variability in local management practices related to animal husbandry makes NH_3 emissions more uncertain than other pollutant emissions derived from, for example, a mobile source model or direct measurements from EGU sources. The EPA has recently improved its models for simulating both livestock waste emissions and the fertilizer application process to inform development of the 2020 NEI, which is expected to have reduced these uncertainties (U.S. EPA, 2023a).

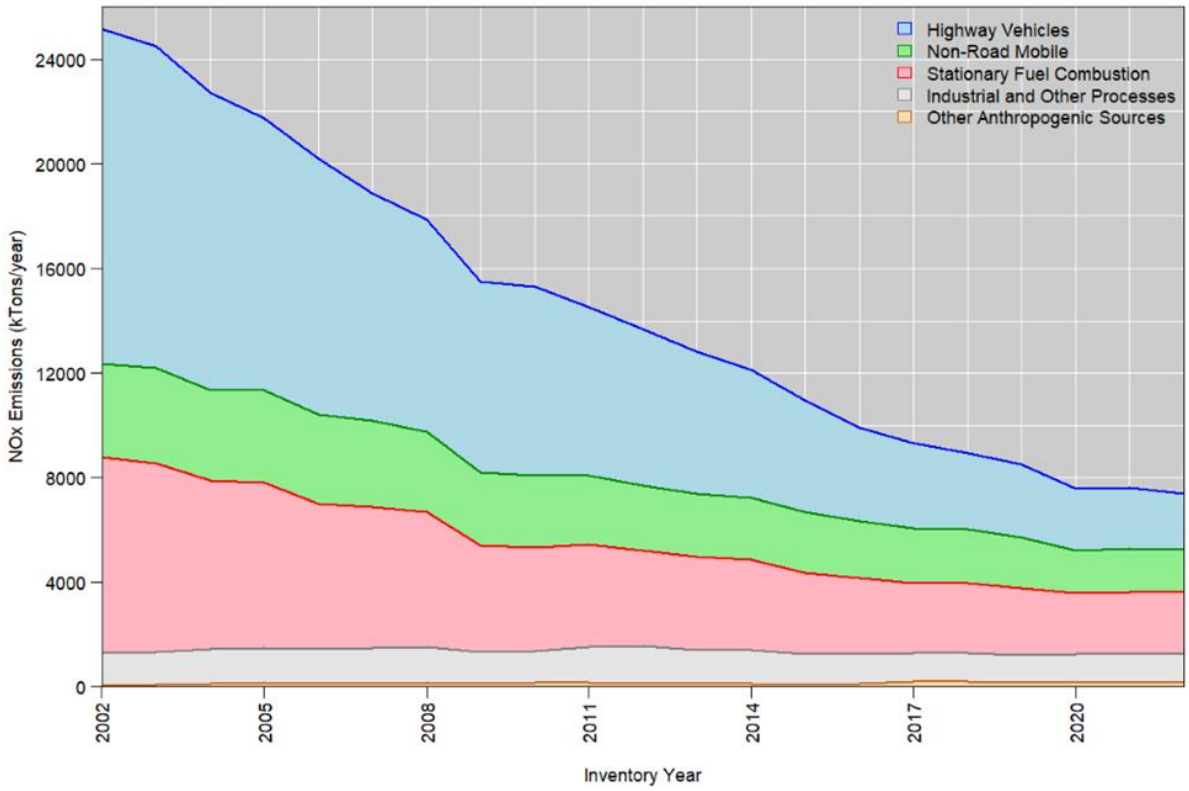

Figure 6-3. Trends in NO + NO_2 emissions by sector from 2002 to 2022 (U.S. EPA, 2023b).

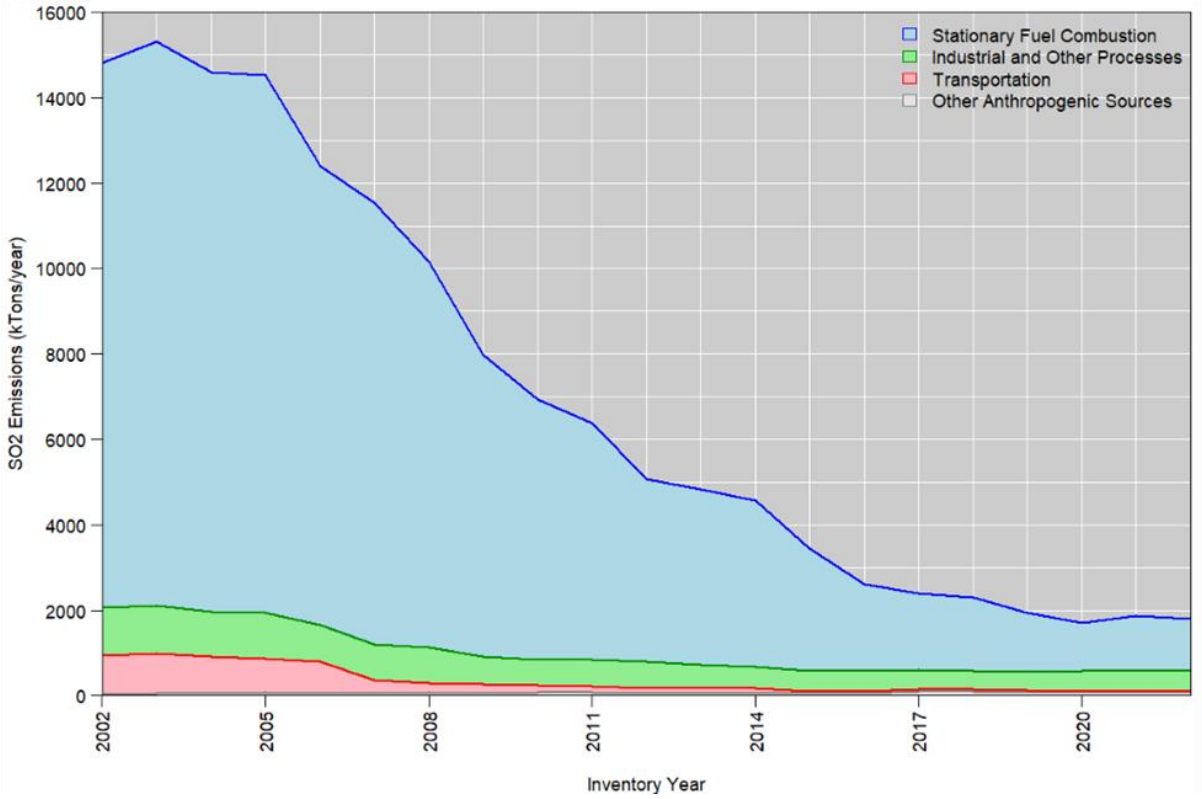

Figure 6-4. Trends in SO_2 emissions by sector from 2002 to 2022 (U.S. EPA, 2023b).

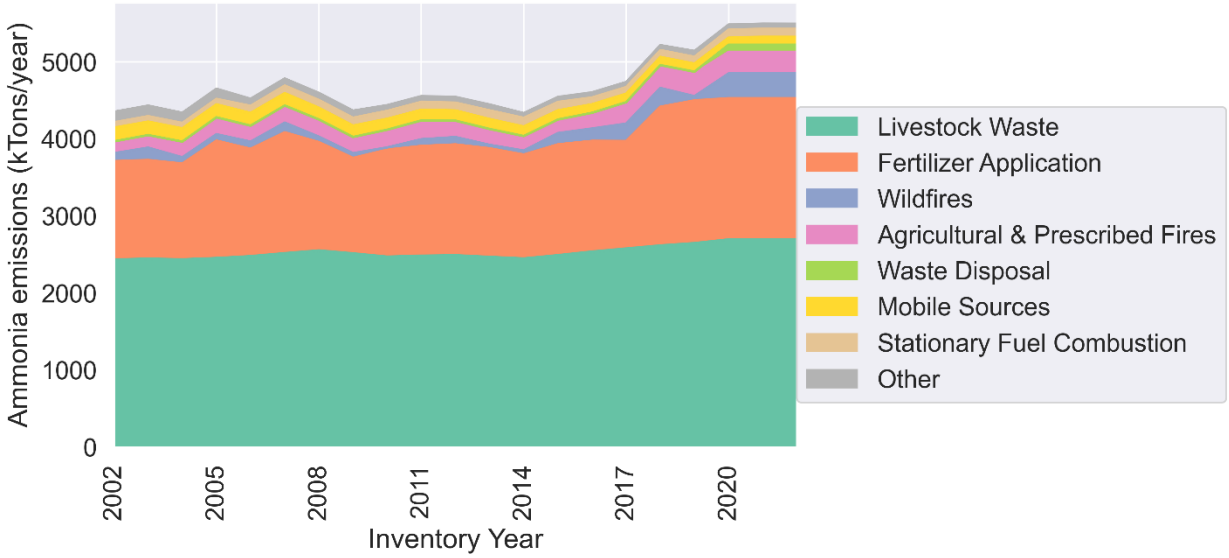

Figure 6-5. Trends in NH_3 emissions by sector from 2002 to 2022.

As discussed in more detail in Chapter 2, coincident with the major reductions in NO_X and SO_2 source emissions, ambient air monitoring data indicate that atmospheric concentrations of NO_2, SO_2, and $PM_{2.5}$ have also trended downward across the U.S. over the past two decades. Figure 6-6 shows the national trends in the annual and 1-hour NO_2 design values based on the

209 sites (annual standard, primary and secondary) and the 135 sites (1-hour standard, primary only) with continuously valid data over the 2000-2022 period. The national median of the annual design values has decreased by 54% from about 15.7 ppb in 2000 to about 7.3 ppb in 2022. The national median of the 1-hour design values has decreased by 38% from 60 ppb in 2000 to 37 ppb in 2022.

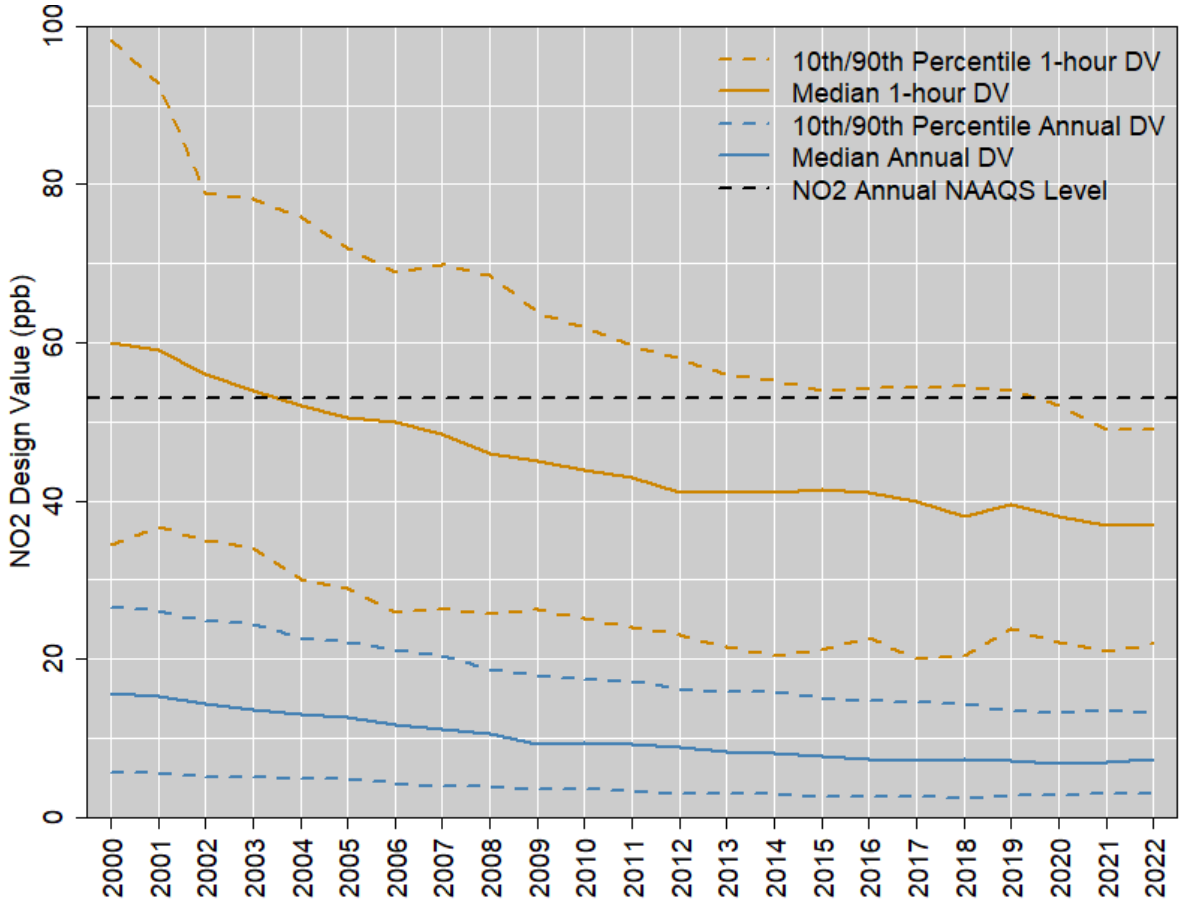

Figure 6-6. Trends in design values for the annual and hourly NO_2 standards (2000 and 2022). (blue = primary and secondary annual standard; brown = primary 1-hour standard).

Figure 6-7 (upper panel) shows the national trends in the 1-hour SO_2 design values (for the primary standard) based on the 162 sites monitoring sites having a valid design value in at least 16 of the 21 three-year periods from 2000 to 2022. The national median of the design values has decreased by about 89% from 69 ppb in 2000 to 8 ppb in 2022. Figure 6-7 (lower panel) presents a similar trend in annual average SO_2 concentrations at SLAMS sites from 2000 to 2021.

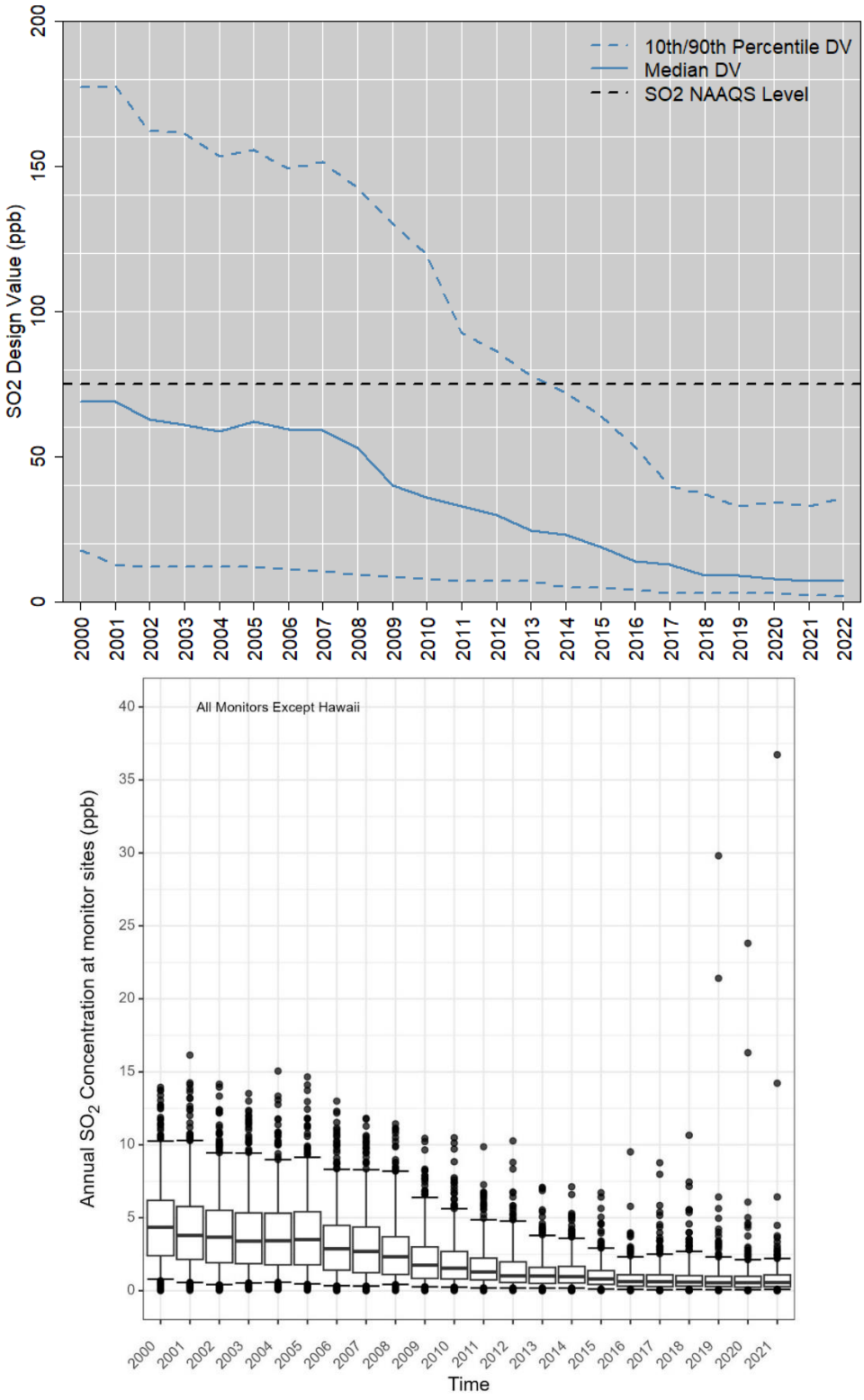

Figure 6-7. Trends in design values for the primary SO_2 standard (99th percentile of 1-hour daily maximum concentrations, averaged over three years) (upper panel) and in annual average SO_2 concentrations at SLAMS in the U.S., excluding Hawaii (lower panel).

Multiple chemicals, including nitrates and sulfates, comprise $PM_{2.5}$. Figure 6-8 shows the national trends in the annual and 24-hour $PM_{2.5}$ design values based on the 395 sites (annual standard) and the 398 sites (24-hour standard) that had valid design values in at least 16 of the 21 three-year periods from 2000-2002 to 2020-2022. Both the annual and 24-hour $PM_{2.5}$ design values exhibited steady decreases from 2002 to 2016. In recent years, the median annual $PM_{2.5}$ design value has remained relatively constant at about 8 µg/m^3 while the 10th and 90th percentile trends have also remained relatively flat at about 6 µg/m^3 and 10 µg/m^3, respectively. The 10th percentile and median of the 24-hour $PM_{2.5}$ design values have also remained relatively constant at about 15 µg/m^3 and 20 µg/m^3, respectively, since 2016. However, the 90th percentile of the 24-hour $PM_{2.5}$ design values has increased substantially in the past six years largely as a result of increased wildfire activity in the western U.S.

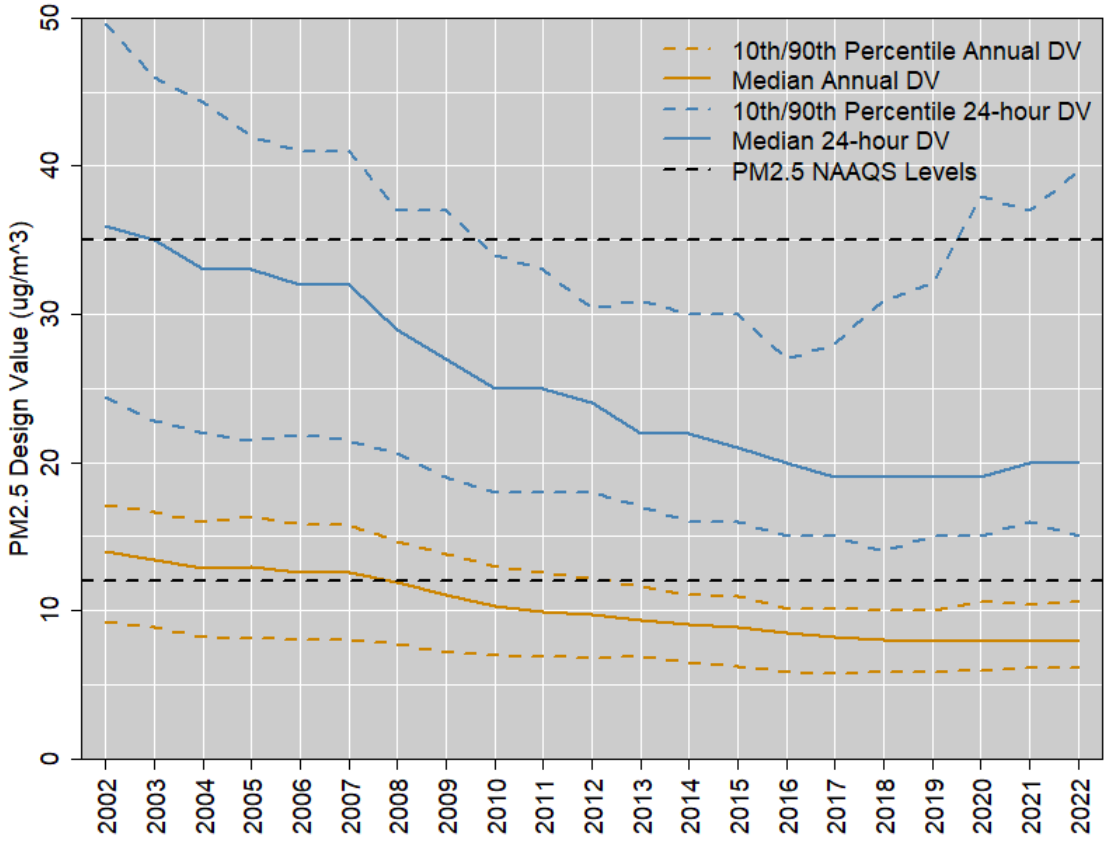

Figure 6-8. Trends in design values for $PM_{2.5}$ standards. The lower black dashed line marks the level of the primary annual standard (12 µg/m^3). The secondary annual $PM_{2.5}$ standard level is 15 µg/m^3. The upper black dashed line marks the level of the primary and secondary 24-hour standards (35 µg/m^3).

Figures 6-9 and 6-10 show trends in annual average concentrations for NO_3^- (nitrate aerosol) and SO_4^{2-} (sulfate aerosol) based on sites that collected data for at least 12 out of 16 years from 2006 to 2021. Broad national reductions in NO_X emissions have resulted in substantial decreasing trends in NO_3^- concentrations over most of the U.S., especially in areas where NO_3^- concentrations were historically highest. Similarly, reductions in SO_2 emissions have resulted in significant reductions in SO_4^{2-} concentrations nationally and especially in the eastern U.S. While not shown here, trends in other $PM_{2.5}$ components like elemental carbon and organic carbon were more variable, with some sites showing substantial decreases and the remaining sites having no clear trend.[1] As discussed in Chapter 2, ammonium sulfate and ammonium nitrate now make up less than one-third of the $PM_{2.5}$ mass at the majority of sites, and only a few sites have more than half of the $PM_{2.5}$ mass from these two compounds.

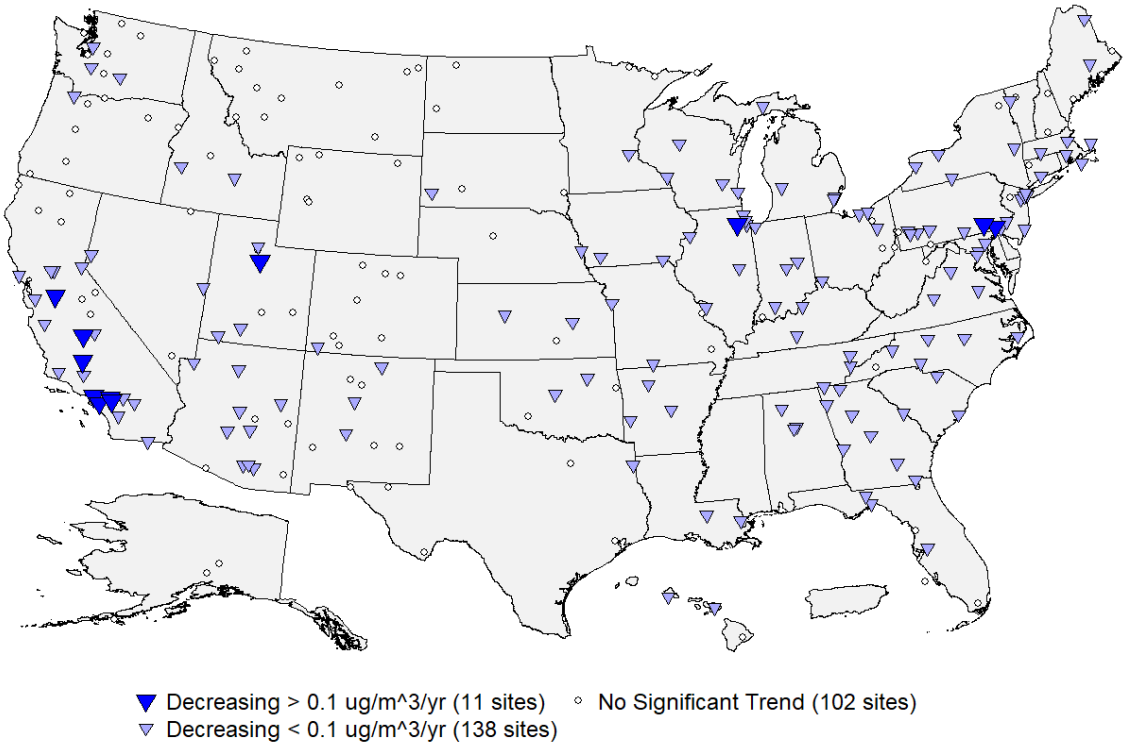

Figure 6-9. Trends in annual average concentrations of NO_3^-, as measured at select NCore, CSN, and IMPROVE sites from 2006 through 2021.

[1] https://gispub.epa.gov/air/trendsreport/2022/#introduction

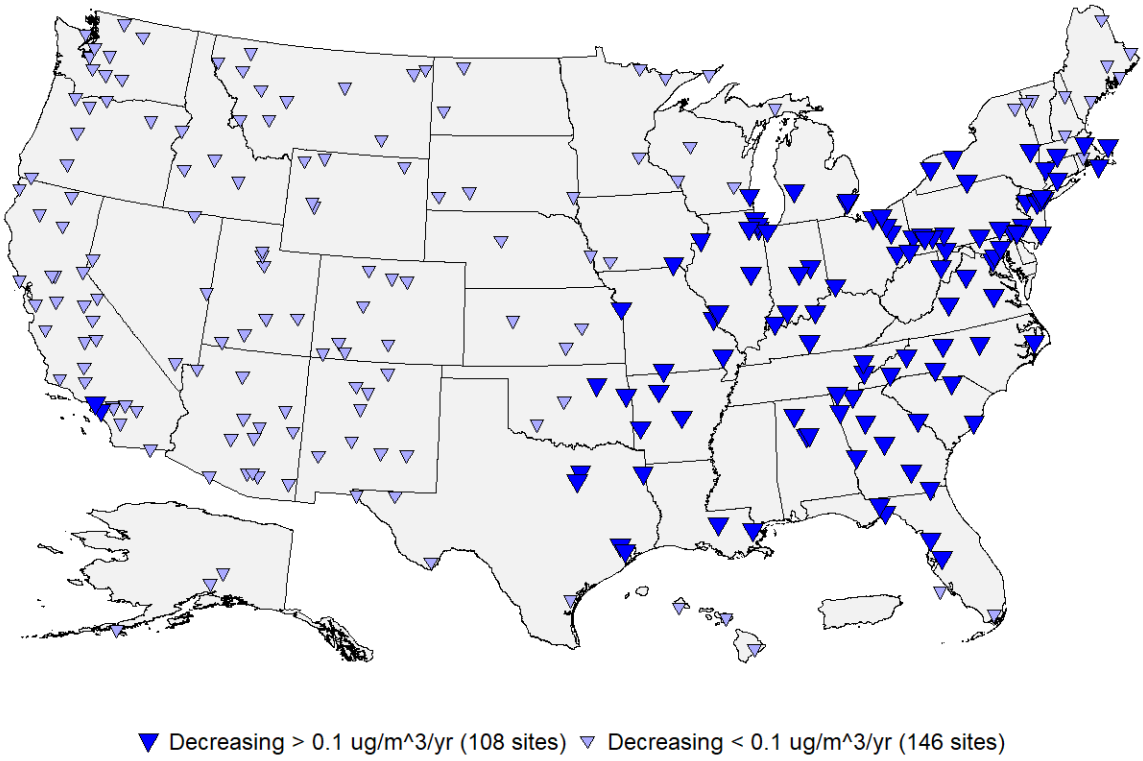

Figure 6-10. Trends in annual average concentrations of SO_4^{2-} as measured at select NCore, CSN, and IMPROVE sites from 2006 through 2021.

Air quality across the U.S. has changed substantially over the past two decades. In response to emissions reductions of NO_X, SO_2, and PM precursor pollutants, concentrations of NO_X, SO_2, and $PM_{2.5}$, including nitrates and sulfates, have decreased sharply. Returning to the examination of the relationship between air concentrations of criteria air pollutants and atmospheric deposition of S and N, these changes in air quality provide an opportunity to assess their simultaneous influence on S and N deposition levels across the U.S. In response to the changes in emissions and air concentrations described above, total deposition of *oxidized* nitrogen and sulfur have also decreased significantly since 2000 (Feng et al., 2020; McHale et al., 2021). Between the two three-year periods of 2000-2002 and 2019-2021, national average estimates of N deposition over the contiguous U.S. have declined by 15% and estimates of total S deposition have declined by 68% (U.S. EPA, 2022). See Table 6-2 for a regional breakout of trends in total S, total N, oxidized N, and reduced N deposition.

The change in total N deposition reflects a combination of declining oxidized N and increasing reduced N, which is consistent with the trends in emissions and air concentrations described above. As expected, the data suggest that dry deposition of nitric acid has decreased significantly over the past two decades and is likely a key contributor to the decrease in total nitrate deposition and decreasing trends in oxidized nitrogen deposition (ISA, Appendix 2,

section 2.7 and Figure 2-60). Emissions decreases of NO_X and downward trends in wet deposition of nitrate have a positive correlation, but because the formation of ammonium is related to the availability of nitrate and sulfate, the correlation between NH_3 emissions and NH_4^+ wet deposition is more complicated (Tan et al., 2020). While dry deposition is more uncertain in magnitude, both surface-based and remote-sensing measurements indicate increasing ammonia concentrations, which is consistent with the increasing trend for ammonia dry deposition, especially in areas with significant agricultural emissions in the Midwest and the Central Valley of California where ammonia dry deposition has become the largest contributor to inorganic N deposition (Li et al., 2016).

The next series of plots further illustrate the changes in deposition patterns across the U.S. over the past two decades. As shown in Figure 6-11,[2] S deposition has decreased sharply across the U.S. over this period due to the significant decreases in sulfur emissions. The changes in sulfur deposition in the Ohio River Valley region are particularly notable. When we restrict the analysis to consider trends only at CASTNET sites, we observe a similar downward trend in total, wet, and dry S deposition, both nationally and over the eastern U.S. (Figure 6-12).

As expected, Figure 6-13 shows that the trends in N deposition are more heterogeneous. Total N deposition has decreased over parts of the Ohio River Valley and in downwind regions such as the northeastern U.S., but there are parts of the country where increases in N deposition are estimated to have occurred over the past two decades (e.g., Texas).

[2] Figures 6-11 and 6-13 through 6-18 were downloaded from the EPA's CASTNET website (https://www.epa.gov/castnet/maps-charts). Figures 6-12 and 6-19 were downloaded from the EPA's CASTNET data download website (https://www.epa.gov/castnet/download-data). Figures 6-11 through 6-19 are based on TDep version v2022.02.

Table 6-2. Regional changes in deposition between 2000-2002 and 2019-2021: (a) total S deposition; (b) total, oxidized and reduced N deposition (U.S. EPA, 2022).

Change in total S deposition

Form of S Deposition	Region	2000-2002	2019-2021	% change
Total Deposition of Sulfur ($kg\ S\ ha^{-1}$)	Mid-Atlantic	15.9	2.1	-87
	Midwest	11.2	2.2	-80
	North Central	3.5	1.5	-56
	Northeast	8.7	1.5	-83
	Pacific	1.0	0.6	-38
	Rocky Mountain	1.0	0.6	-46
	South Central	5.4	2.8	-49
	Southeast	10.3	2.6	-74

Change in total, oxidized and reduced N deposition

Form of N Deposition	Region	2000-2002	2019-2021	% change
Total Deposition of Nitrogen ($kg\ N\ ha^{-1}$)	Mid-Atlantic	13.4	8.5	-36
	Midwest	12.2	9.8	-20
	North Central	8.5	9.5	+11
	Northeast	10.4	6.2	-40
	Pacific	3.8	3.1	-18
	Rocky Mountain	3.0	3.1	+3
	South Central	7.8	9.0	+16
	Southeast	10.8	8.4	-23
Total Deposition of Oxidized Nitrogen ($kg\ N\ ha^{-1}$)	Mid-Atlantic	10.3	4.0	-62
	Midwest	8.0	3.6	-54
	North Central	4.1	2.6	-37
	Northeast	7.7	2.9	-62
	Pacific	2.4	1.4	-42
	Rocky Mountain	1.9	1.3	-35
	South Central	5.0	3.1	-39
	Southeast	7.7	3.4	-56
Total Deposition of Reduced Nitrogen ($kg\ N\ ha^{-1}$)	Mid-Atlantic	3.0	4.6	+51
	Midwest	4.3	6.2	+45
	North Central	4.4	6.9	+56
	Northeast	2.7	3.3	+22
	Pacific	1.4	1.7	+22
	Rocky Mountain	1.1	1.8	+72
	South Central	2.8	6.0	+111
	Southeast	3.1	5.0	+63

The states included in each region are as follows: Mid-Atlantic: DE, MD, NJ, PA, VA, WV; Midwest: IL, IN, KY, MI, OH, WI; North Central: IA, KS, MN, MO, ND, NE, SD; Northeast: CT, MA, ME, NH, NY, RI, VT; Pacific: CA, NV, OR, WA; Rocky Mountain: AZ, CO, ID, MT, NM, UT, WY; South Central: AR, LA, OK, TX; Southeast: AL, FL, GA, MS, NC, TN, SC.

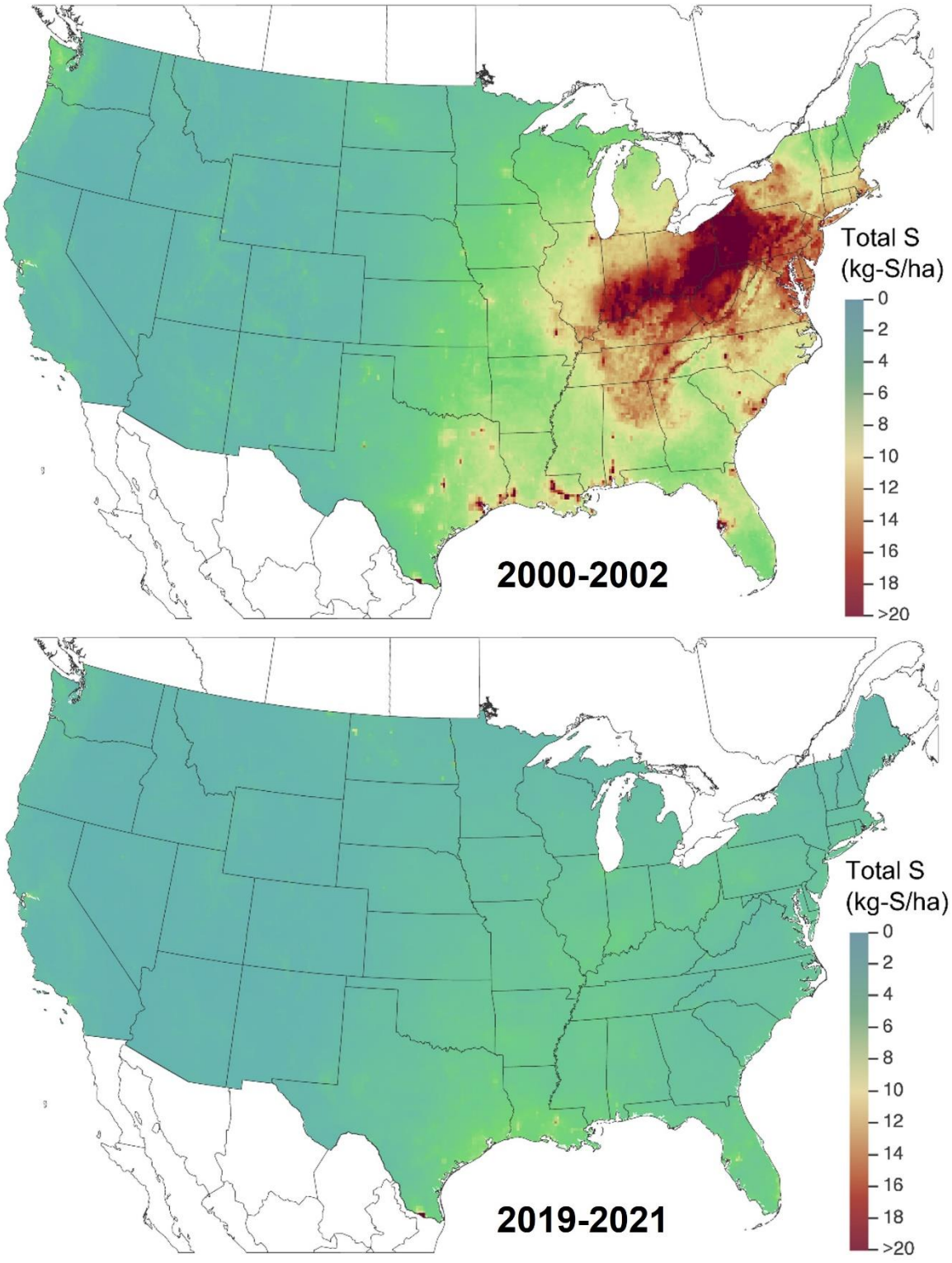

Figure 6-11. TDep-estimated total S deposition: 2000-2002 (top) and 2019-2021 (bottom).

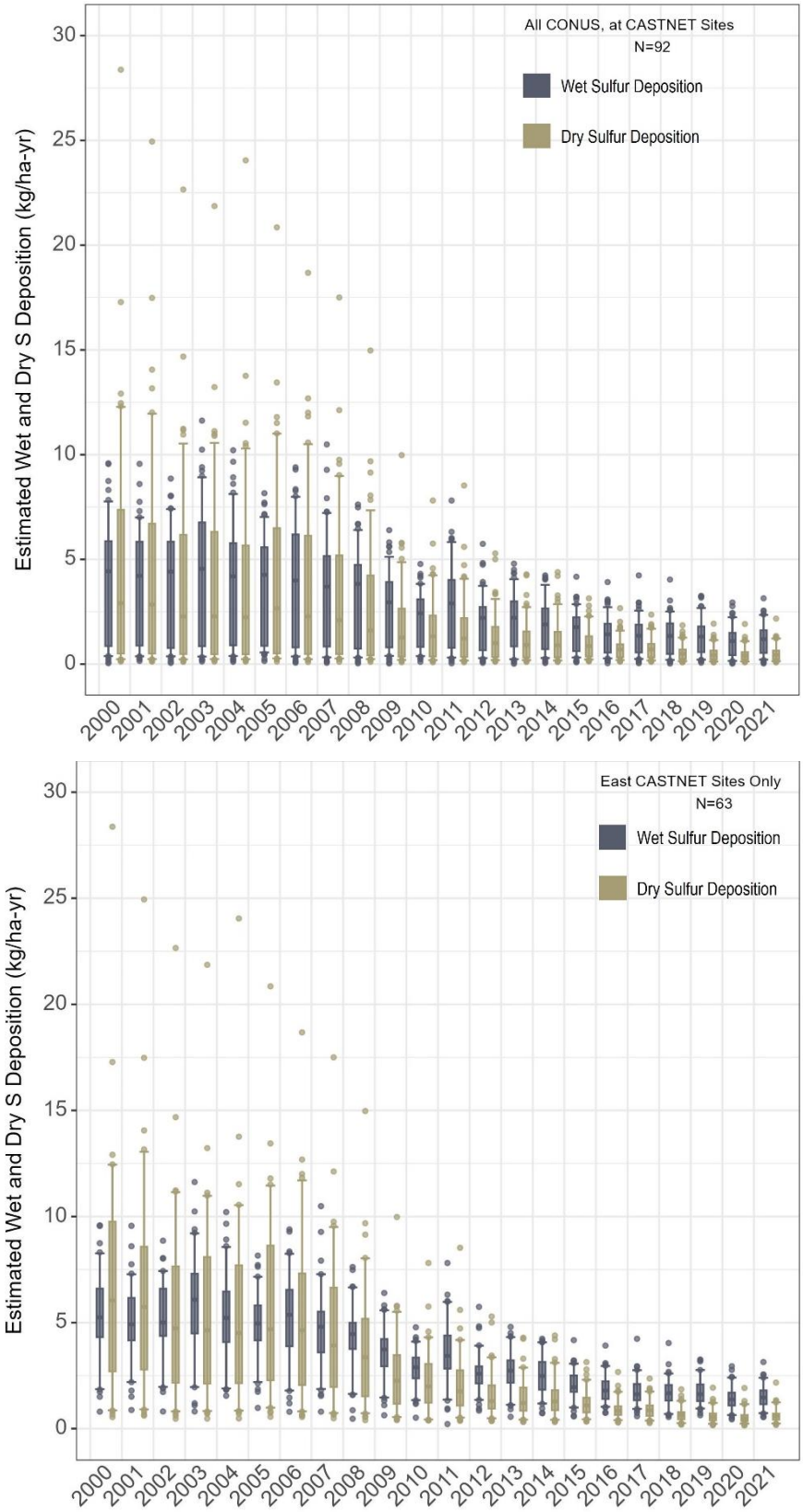

Figure 6-12. Trend in TDep estimates of S deposition (2000-2021) at all 92 CASTNET sites (upper) and the subset of 63 eastern sites (lower).

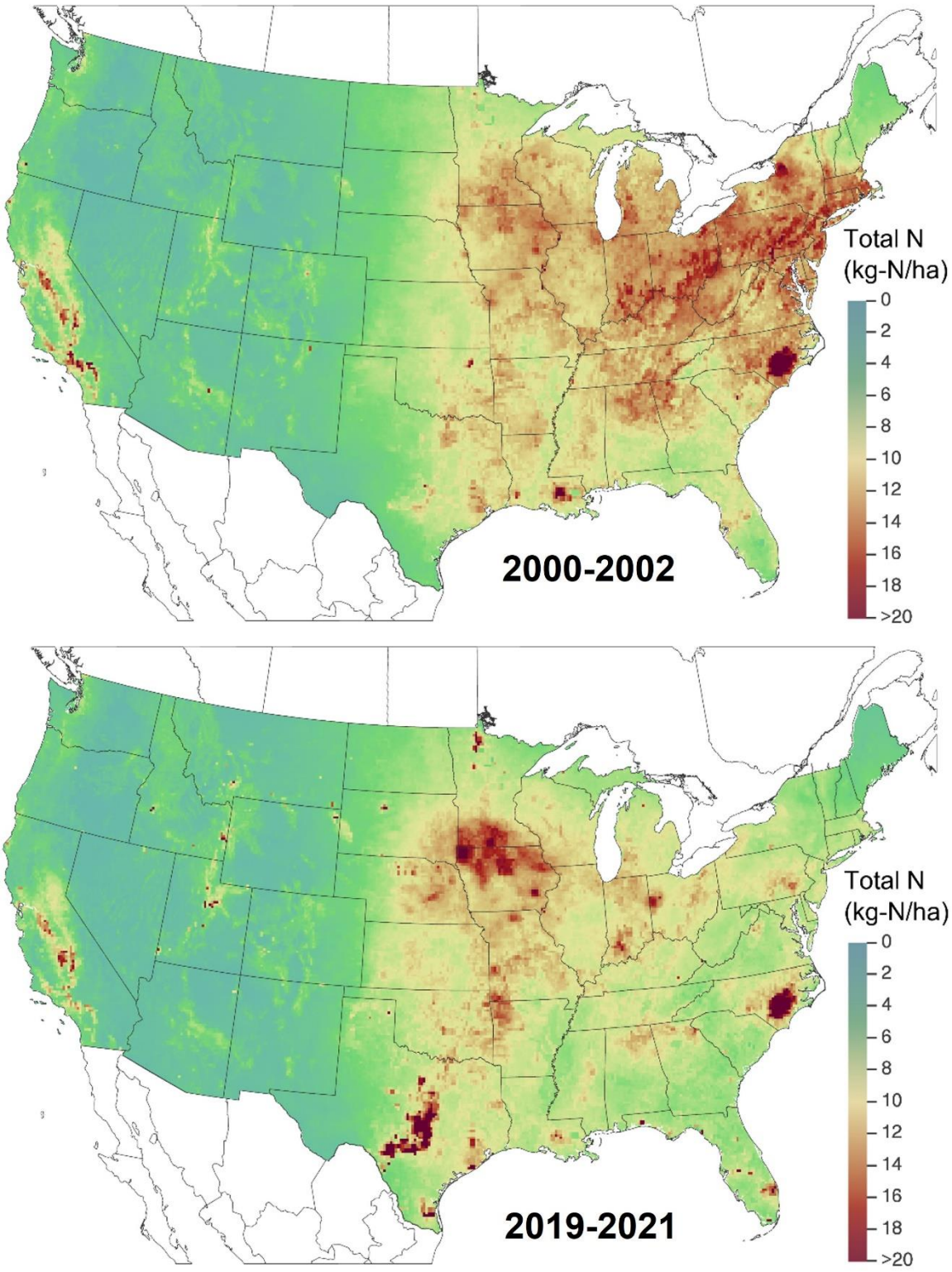

Figure 6-13. TDep-estimated total N deposition: 2000-2002 (top) and 2019-2021 (bottom).

Looking into the components of these trends in N deposition, it can be seen from Figure 6-14 that most of the widespread changes in N deposition across the U.S., both increases and decreases, are due to changes in dry deposition of N. Figure 6-15 shows that while there have been some changes in wet N deposition over the past 20 years (e.g., decreases near Lake Ontario; increases in parts of southern MN), these levels and patterns have remained relatively unchanged compared to dry N deposition.

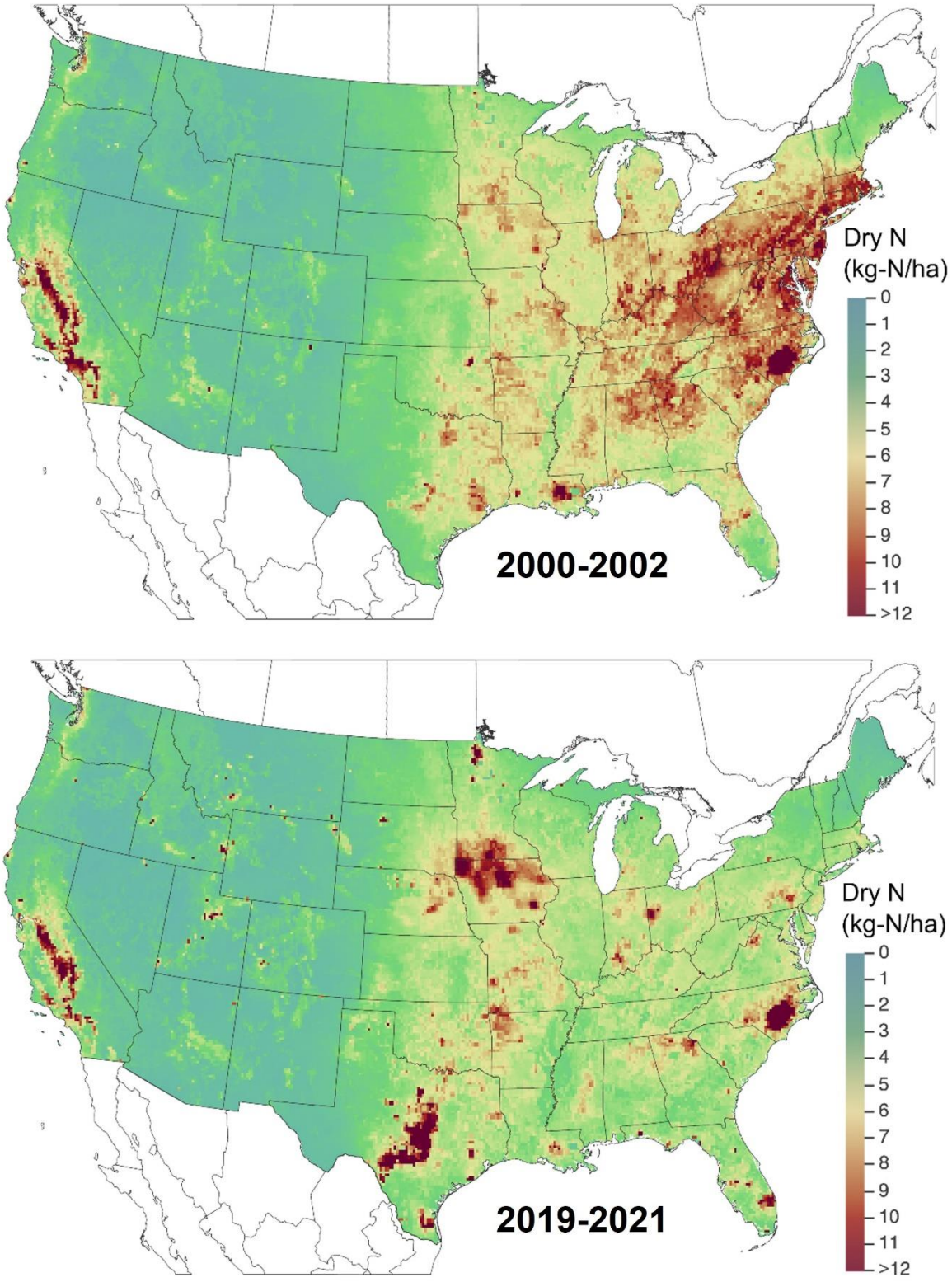

Figure 6-14. TDep-estimated dry N deposition: 2000-2002 (top) and 2019-2021 (bottom).

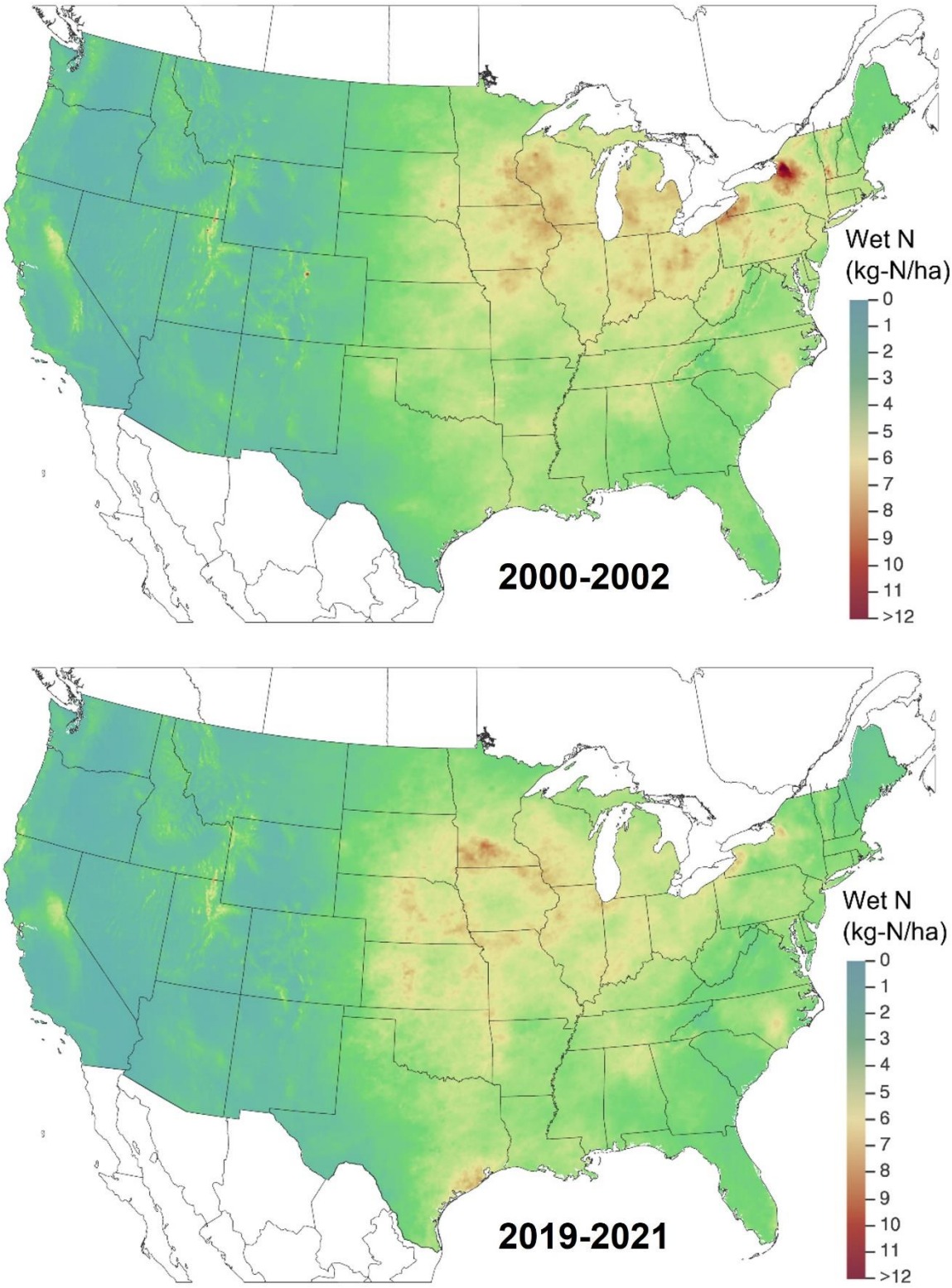

Figure 6-15. TDep-estimated wet N deposition: 2000-2002 (top) and 2019-2021 (bottom).

The aggregate trends in dry deposition of N are driven by two largely opposing trends in the dry deposition of oxidized nitrogen and reduced nitrogen. Two decades ago, there were large amounts of dry oxidized N deposition (5-10 kg N/ha) over much of the eastern U.S. that are not seen in the more current period (< 5 kg N/ha), as shown in Figure 6-16. Conversely, while there were isolated hotspots of dry reduced N deposition in the 2000-2002 timeframe, the number and magnitude of these hotspots has increased substantially in the more recent 2019-2021 period, as shown by Figure 6-17, especially in places like AR, IA, MN, MO and TX. Figure 6-18 confirms that the increases in dry deposition of reduced N are closely linked to increases in NH_3 deposition.

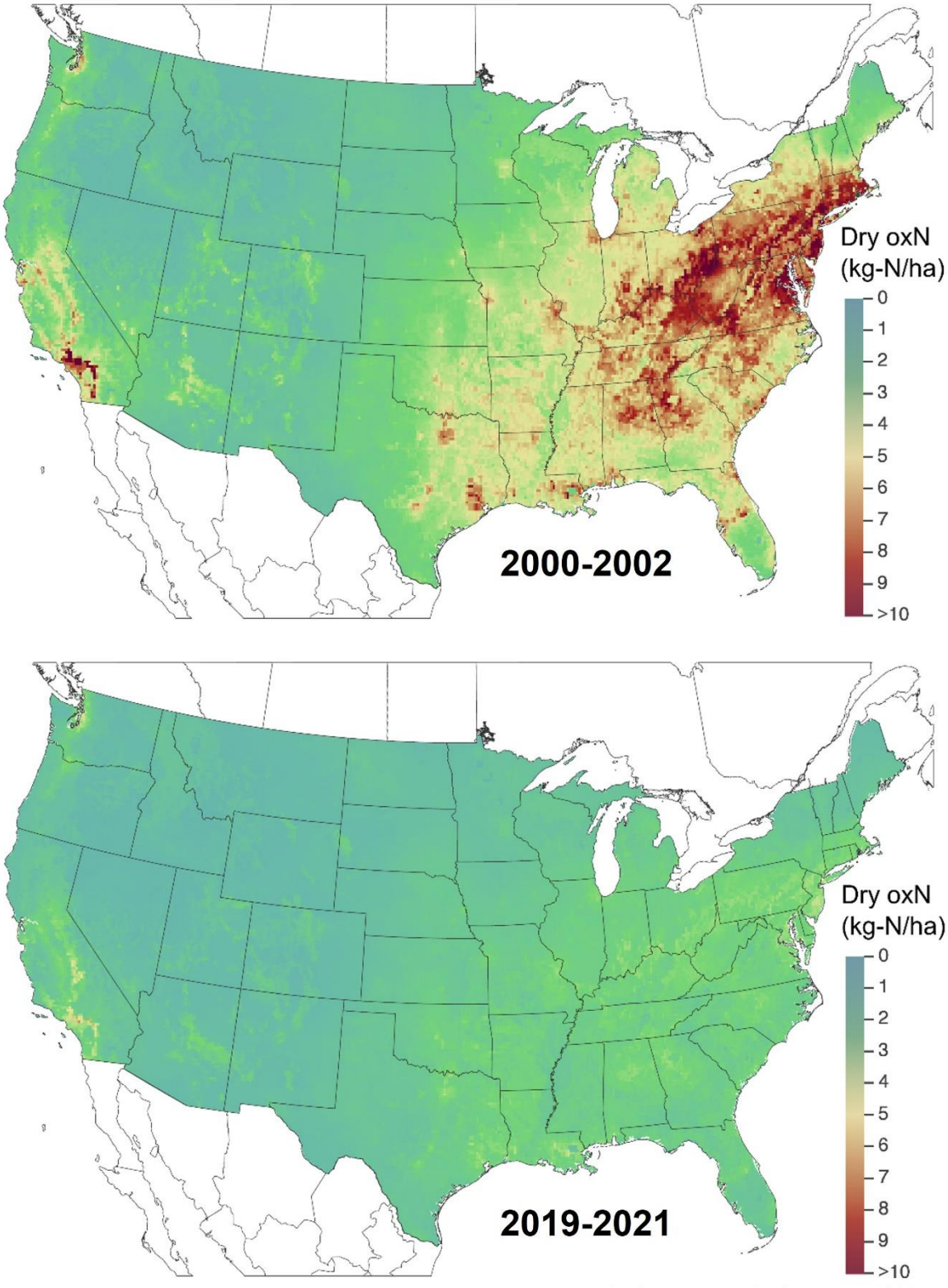

Figure 6-16. Dry oxidized N deposition (TDep estimates): 2000-2002 (top) and 2019-2021 (bottom).

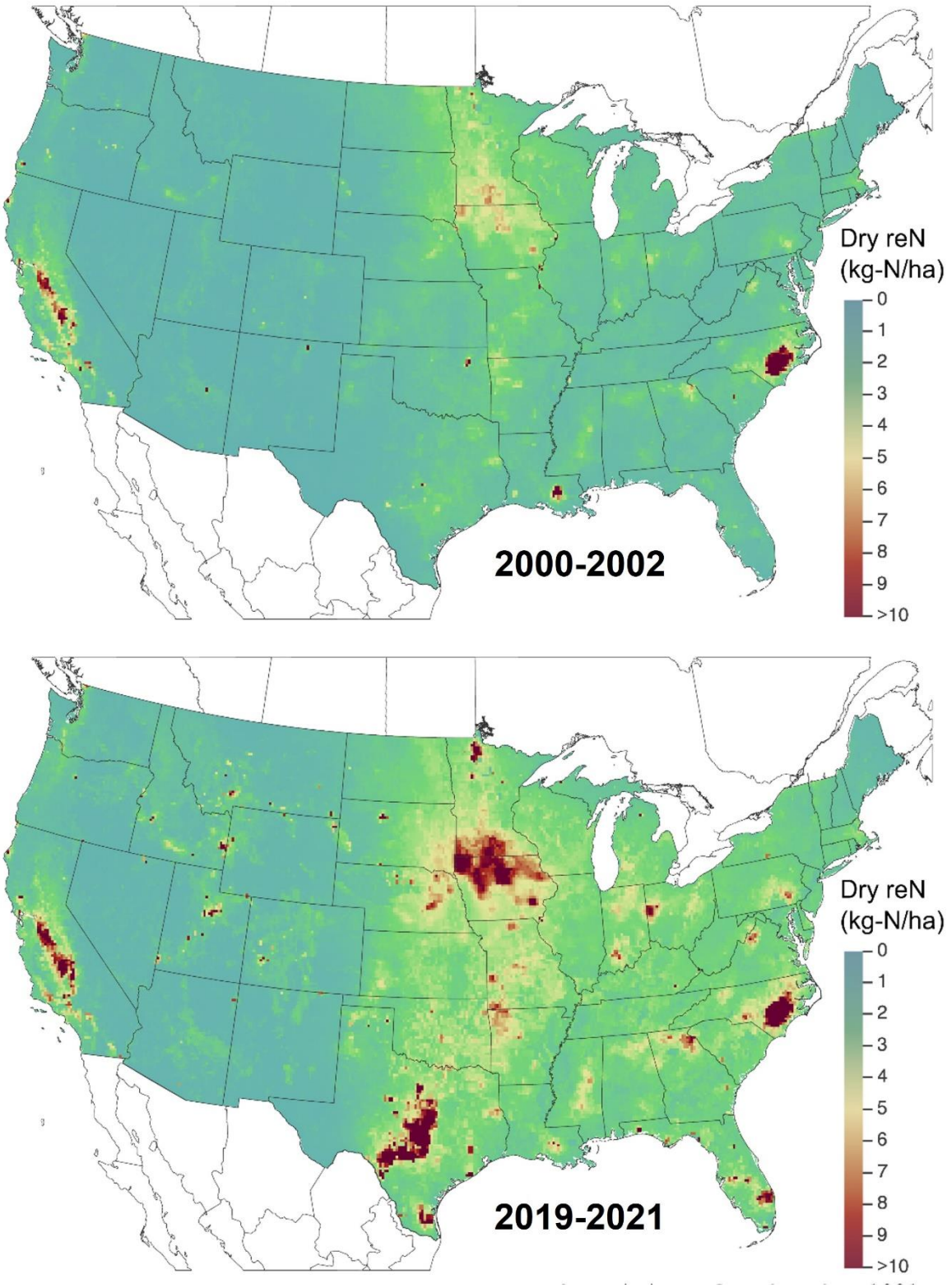

Figure 6-17. TDep-estimated dry reduced N deposition: 2000-2002 (top) and 2019-2021 (bottom).

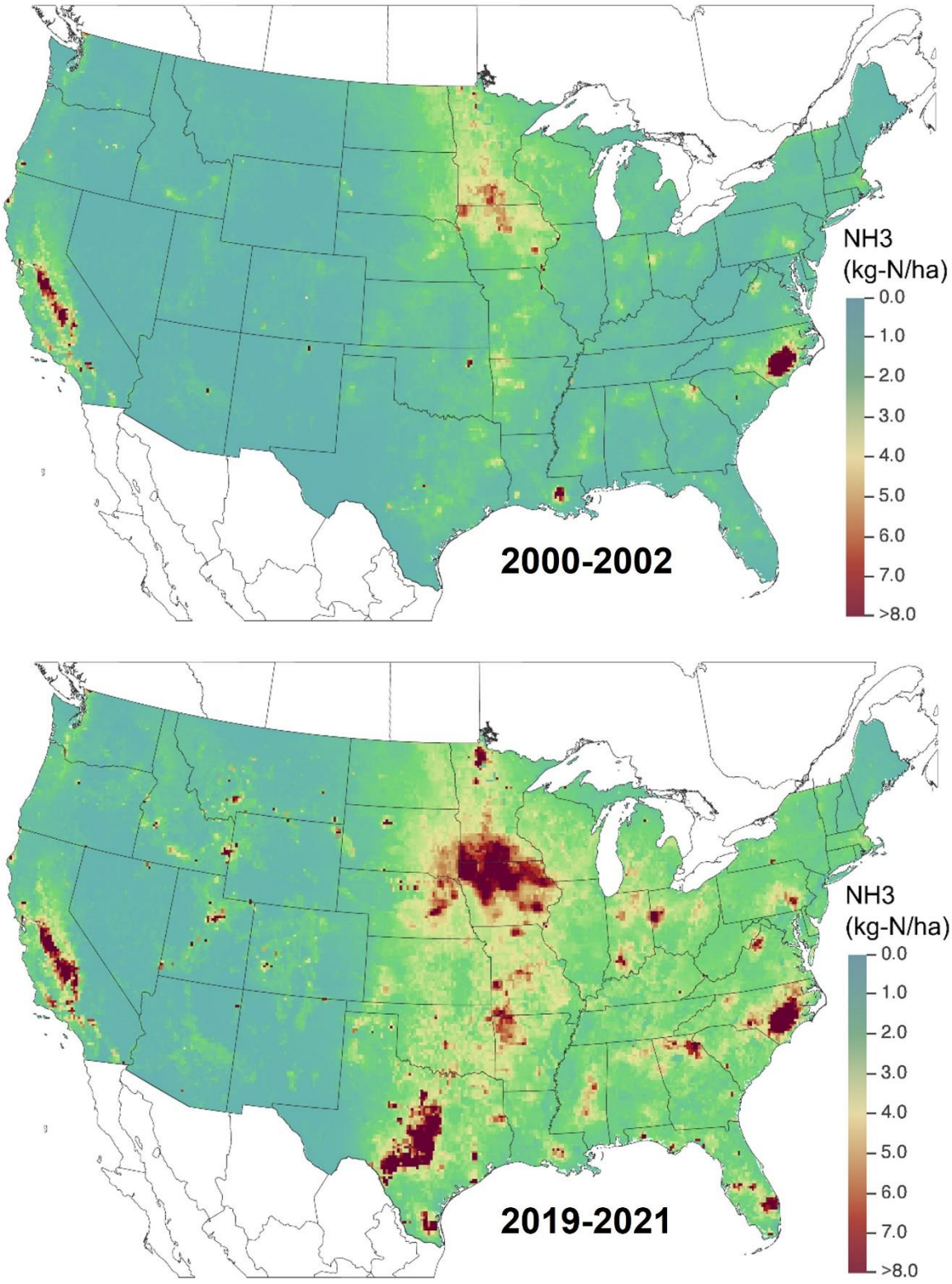

Figure 6-18. TDep-estimated NH_3 deposition: 2000-2002 (top) and 2019-2021 (bottom).

Annual estimates of major components of N deposition at 92 CASTNET sites across the U.S. (for which locations are shown in Figure 2-17) during the period from 2000 through 2021 further confirm the changing trends in the influence of oxidized and reduced N species, as shown in Figure 6-19. Over this period, the relative presence of oxidized species has declined at these monitors, tracking the trends in NO and NO_2 emissions noted above. However, the relative presence of NH_3 has increased appreciably (Figure 6-19).

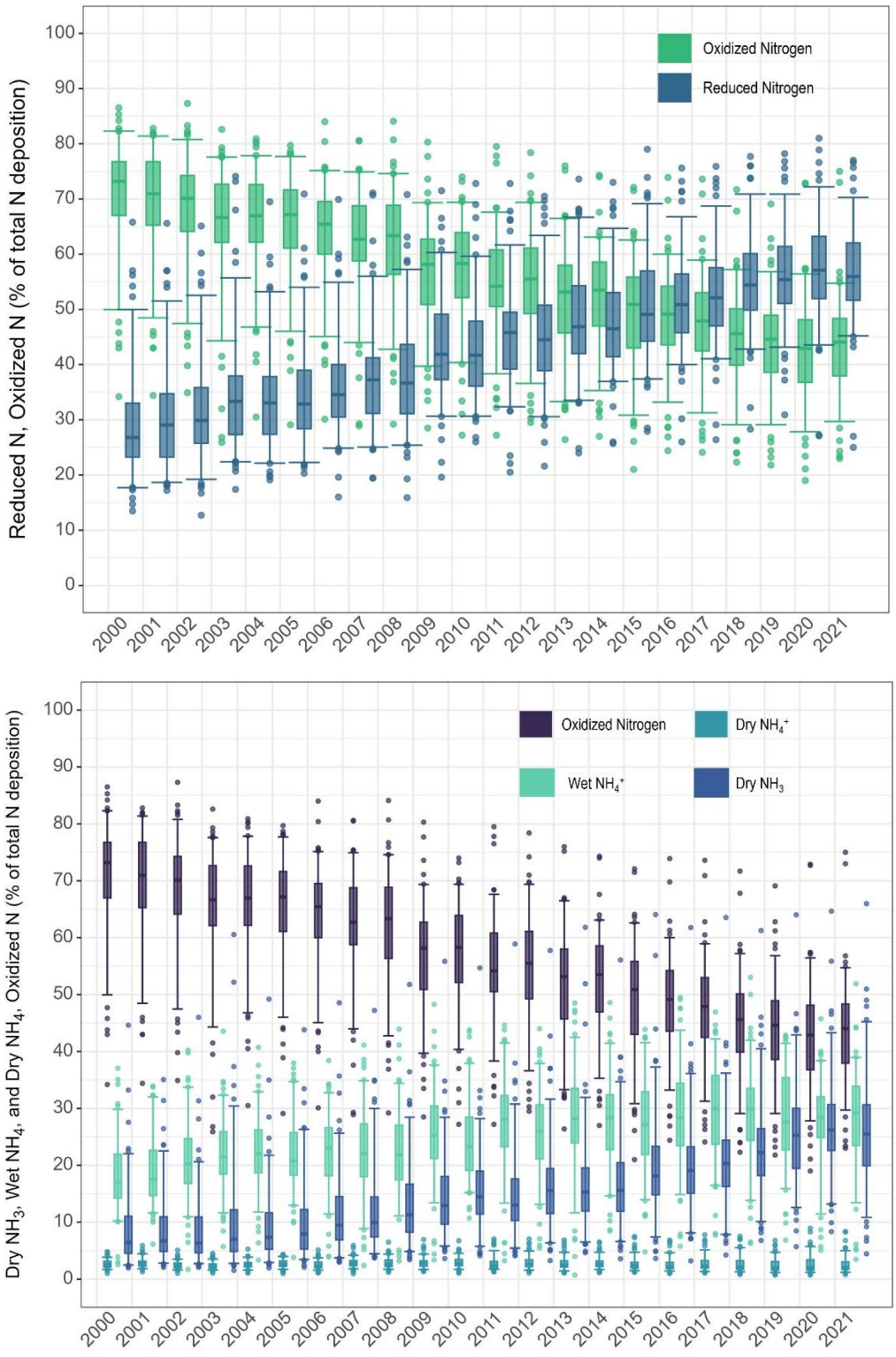

Figure 6-19. TDep estimated components of N deposition at all 92 CASTNET sites (2000-2021): oxidized and reduced N (upper) and by oxidized and reduced N component species (lower).

The longer-term trends in deposition of reduced nitrogen are more challenging to assess because, before 2011, ambient air NH_3 monitoring was rare. For particulate matter, the trend in ammonium (NH_4^+) has followed the downward trends in sulfate and nitrate, because aerosol partitioning to NH_4^+ requires the availability of acid gases, such as sulfur oxides and/or nitrogen oxides, to neutralize NH_3. This increased prevalence of gas phase NH_3 also contributes to the trend of increasing dry fraction of total nitrogen deposition. Satellite-based measurements and chemical transport models have been used to augment the surface-based measurements of ammonia and ammonium to better understand trends. These studies also show increasing ammonia concentrations, especially in parts of the Midwest, Southeast, and West near agricultural sources (Warner et al., 2016; Warner et al., 2017; Yu et al., 2018; Nair et al., 2019; He et al., 2021). These trends are attributed to a combination of warmer temperatures causing greater emissions, increasing agricultural activity, and less available sulfate and nitrate, shifting the prevalence in reduced nitrogen partitioning from particle ammonium toward gas-phase ammonia.

In summary, the analysis of air quality concentrations from criteria pollutants and deposition data over the past two decades show similar trends in the following quantities, implying that there is correlation between:

- SO_2 concentrations and total S deposition
- NO_2 concentrations and *oxidized* N deposition
- $PM_{2.5}$ sulfate concentrations and total S deposition.

However, the spatiotemporal trends between ambient air NO_2 concentrations and total N deposition are inconsistent; due to increases in NH_3 emissions and associated increases in reduced N deposition. Similarly, the trends data show a mixed relationship between $PM_{2.5}$ nitrate concentrations and total N deposition. The subsequent analyses presented in this section are designed to expand upon this simple observation of correlation.

6.2.2 Class I Area Sites – Relationships Between Air Concentrations and Deposition

As a second type of analysis to evaluate the relationship between air quality metrics of interest and the deposition of S and N, we evaluated observational data at 27 sites in 27 remote Class I areas. These areas tend to be further away from emissions sources and are of particular interest for ecological reasons related to the secondary standards, as well. Class I areas have some special federal protections (e.g., focus of efforts to reduce regional haze).[3]

[3] Areas designated as Class I receive special protection status under the CAA, and include all international parks, national wilderness areas that exceed 5,000 acres in size, national memorial parks that exceed 5,000 acres in size, and national parks that exceed 6,000 acres in size, provided the park or wilderness area was in existence on August 7, 1977.

In this section, we first evaluate concentration-to-deposition relationships at these locations from CMAQ simulations (section 6.2.2.1) to consider how the terms are associated within a chemical transport model simulation. We then analyze ambient air observations from CASTNET and IMPROVE sites and measured wet deposition from NADP/NTN sites in these same Class I areas (section 6.2.2.2) to identify S and N-containing compounds for which air concentrations are closely related to S and N deposition. This section also considers TDep estimated total S and N deposition and how total deposition relates to measured ambient air concentrations from CASTNET and IMPROVE monitor sites (section 6.2.2.3). Noting the many factors that can lead to variability in estimated deposition, including frequency of precipitation, and micrometeorological factors relevant to the dry deposition velocity, the analyses focus on multiple years of data to better assess these local relationships. The averaging time for all these comparisons is one year.

The set of 27 Class I areas with co-located CASTNET monitoring stations, chemically-speciated $PM_{2.5}$ from the IMPROVE network, and NADP/NTN wet deposition monitors are identified in Table 6-3 and shown in the map in Figure 6-20. Figure 6-21 shows the distribution of TDep-estimated wet and dry deposition amounts across these 27 areas for the 2017-2019 period. At these locations, N deposition tends to be much greater than S deposition, with both quantities lower than national average values, likely because most of these locations are in the western U.S. Consistent with the national trends, S deposition has also declined more than N deposition over the last few decades at these locations. For nitrogen, at these sites during this time period, dry deposition comprises approximately 60% and wet deposition approximately 40% of total deposition estimates (Figure 6-21). In contrast, for sulfur, wet deposition comprises approximately 60%, on average, and dry deposition, 40% (Figure 6-21). While this presentation reflects estimates at this specific set of sites, we note that patterns here may differ from patterns at other sites across the U.S.

Table 6-3. Collocated CASTNET, NADP/NTN, and IMPROVE monitoring stations used in this analysis of air concentration and deposition.

Class I Area Name	CASTNET	NADP	IMPROVE
Acadia	ACA416	ME98	ACAD1
Big Bend	BBE401	TX04	BIBE1
Canyonlands	CAN407	UT09	CANY1
Chiricahua	CHA467	AZ98	CHIR1
Death Valley	DEV412	CA95	DEVA1
Dinosaur National Monument	DIN431	CO15	DINO1
Everglades	EVE419	FL11	EVER1
Glacier	GLR468	MT05	GLAC1
Great Basin	GRB411	NV05	GRBA1
Grand Canyon	GRC474	AZ03	GRCA2
Great Smokey Mountains	GRS420	TN11	GRSM1
Joshua Tree	JOT403	CA67	JOSH1
Mt. Lassen	LAV410	CA96	LAVO1
Mammoth Cave	MAC426	KY10	MACA1
Mesa Verde	MEV405	CO99	MEVE1
Cascades	NCS415	WA19	NOCA1
Olympic	OLY421	WA14	OLYM1
Petrified Forest	PET427	AZ97	PEFO1
Pinnacles	PIN414	CA66	PINN1
Rocky Mountain	ROM406	CO19	ROMO1
Sequoia	SEK430	CA75	SEQU1
Shenandoah	SHN418	VA28	SHEN1
Theodore Roosevelt	THR422	ND00	THRO1
Voyageurs	VOY413	MN32	VOYA2
Wind Cave	WNC429	SD04	WICA1
Yellowstone	YEL408	WY08	YELL2
Yosemite	YOS404	CA99	YOSE1
Monitored Parameters Included in this Analysis	Gas: SO_2 and HNO_3. Particulate: SO_4, NO_3^-, NH_4^+ [$TNO_3 = HNO_3 + pNO_3^-$]	Wet deposition of: S compounds (SO_4^{2-}) and N compounds (NO_3^-, NH_4^+)	$PM_{2.5}$(total mass)*; $PM_{2.5}(SO_4^{2-})$; $PM_{2.5}(NO_3^-)$; $PM_{2.5}(NH_4^+)$ $PM_{2.5}(NO_3^- + NH_4^+)$

* $PM_{2.5}$ mass monitors at IMPROVE sites employ methods other than FRM/FEM sites (Hand et al., 2023).

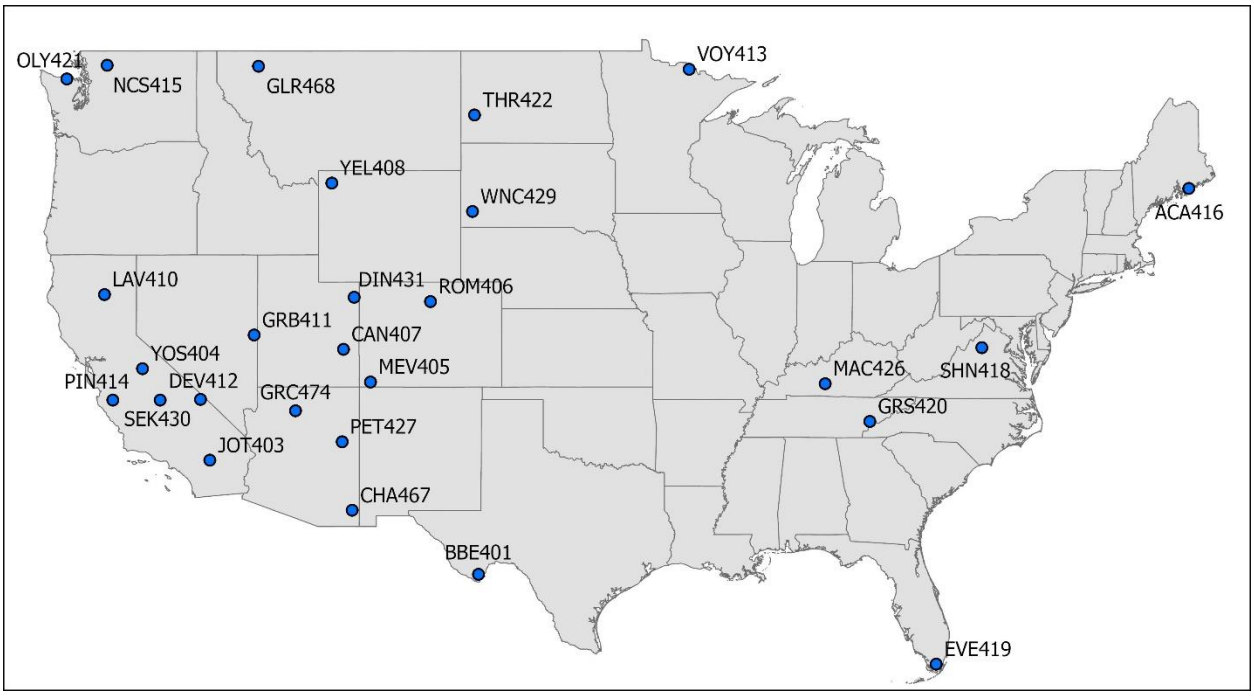

Figure 6-20. Locations of co-located CASTNET, NADP/NTN, and IMPROVE monitoring sites, denoted by CASTNET site identifier. The NADP/NTN and IMPROVE station identifiers are listed in Table 6-3.

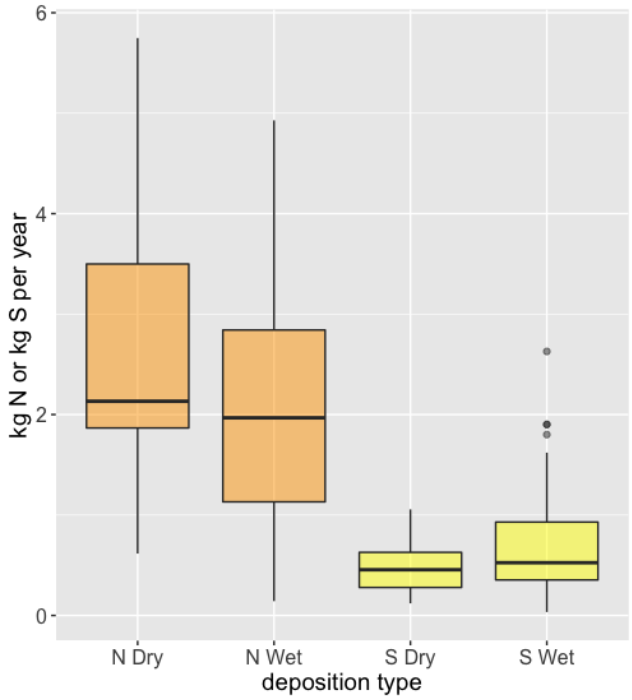

Figure 6-21. Annual average TDep-estimated dry and wet deposition of N and S (2017-2019) at Class I area NADP sites in Table 6-3. Boxes indicate interquartile range.

In the following subsections, the analyses focus on assessing relationships between: (1) simulated air concentrations and simulated total deposition using output from a chemical transport model (CMAQ) that reflects known physical and chemical processes, and (2) measured air concentrations (IMPROVE and CASTNET), measured wet deposition (NADP/NTN), and estimated total deposition (TDep). These sets of measured and predicted variables are compared using linear regression, which allows a more detailed assessment of the uncertainty and variability. There are several ways to assess how well one variable relates to the other, such as by calculating the correlation between variables (*r*), creating linear regressions for pairs of variables, and calculating the significance of those analyses (*p* value). The correlation coefficients used in this chapter are Spearman's Rank Correlation.[4] While the correlation coefficients are useful in evaluating the relative strength of a concentration-deposition association, it is also important to visually consider the relationship via a scatterplot. Such figures are also provided in this section.

6.2.2.1 Relationships in Chemical Transport Model Simulations

Since dry deposition flux is not routinely measured, models are often used to inform deposition estimates and to examine the relationship between air concentration and total deposition. The CMAQ is a numerical air quality model that relies on scientific first principles to simulate the concentration of airborne gases and particles and the deposition of these pollutants back to Earth's surface under user-prescribed scenarios. We utilize the results of a 21-year CMAQ simulation, as described in Zhang et al. (2018), to further analyze relationships between air concentrations and deposition of S- and N-related compounds as part of this review. One of the inherent advantages of evaluating the concentration to deposition relationship in a chemical transport model is that one is not limited by measurement technology (e.g., absence of widespread dry deposition data or challenges in measuring certain pollutant concentrations). However, an important caveat is that the model-estimated relationships will be affected by imperfect parameterizations as the model necessarily simplifies highly complex real-world processes in its simulations.

For model grid cells across the contiguous U.S., CMAQ-estimated annual average SO_X and N oxides concentrations, total S and N deposition, and the associated deposition-to-concentration ratios are presented in Figures 6-22 and 6-23. For SO_X (Figure 6-22), most of the U.S. generally exhibits deposition:concentration ratios of 1 to 5, especially in areas where local and regional sources of SO_2 are prevalent. However, as an air parcel moves further away from emissions sources, the more rapidly-depositing pollutants are removed, and pollutants are diluted by being mixed vertically in the atmosphere. In these locations, the deposition-to-concentration

[4] All linear regressions in this chapter were derived using R, version 4.3.1, and correlations were calculated using Spearman's rank correlation.

ratios for S oxides are higher (i.e., > 5). Such locations include parts of the northeastern U.S. and high elevation sites in the western U.S. These areas are generally further away from sources and ground-level air concentrations in these regions are relatively low. For N oxides the spatial patterns are similar, however the ratios are slightly lower over most of the U.S. (i.e., ratios range from 1 to 3, Figure 6-23). The spatial consistency in the simulated deposition-to-concentration ratios in the model, at least over the annual averaging time considered here, indicates some general association between local deposition rates and local ambient air concentrations of S and N oxides. However, this general rule has clear exceptions (e.g., high altitude sites) and there is some variability within the typical range of ratios.

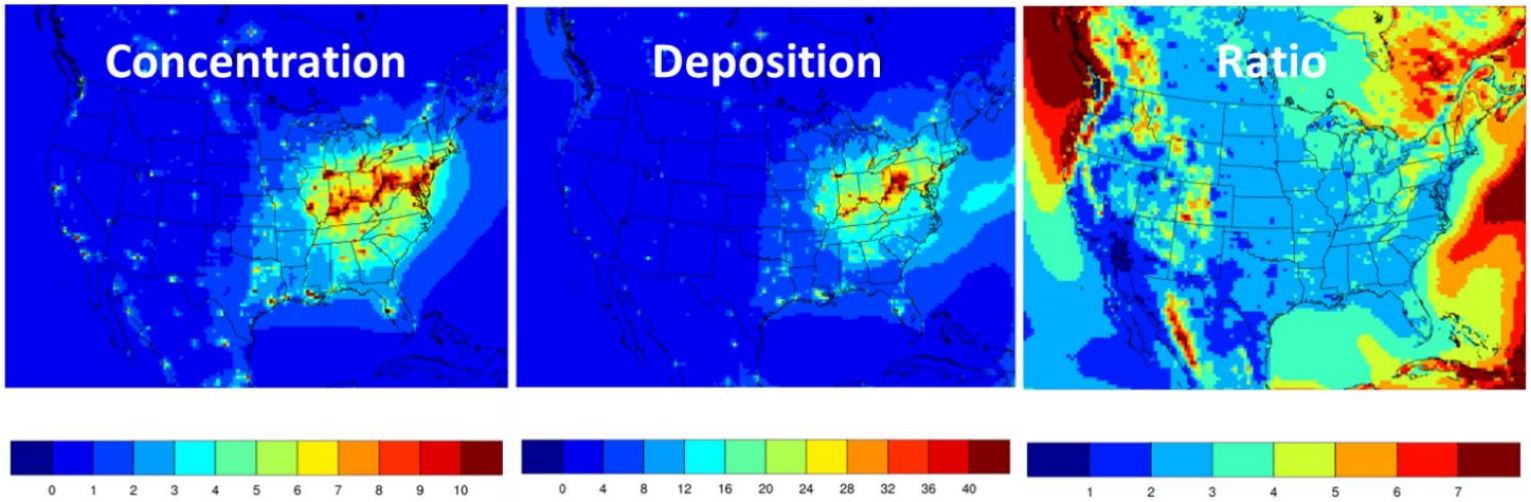

Figure 6-22. Annual average SO_X concentration, ppb, (left), total S deposition, kg/ha-yr, (middle), and associated deposition:concentration ratios (right), estimated from a 21-year (1990-2010) CMAQ simulation.

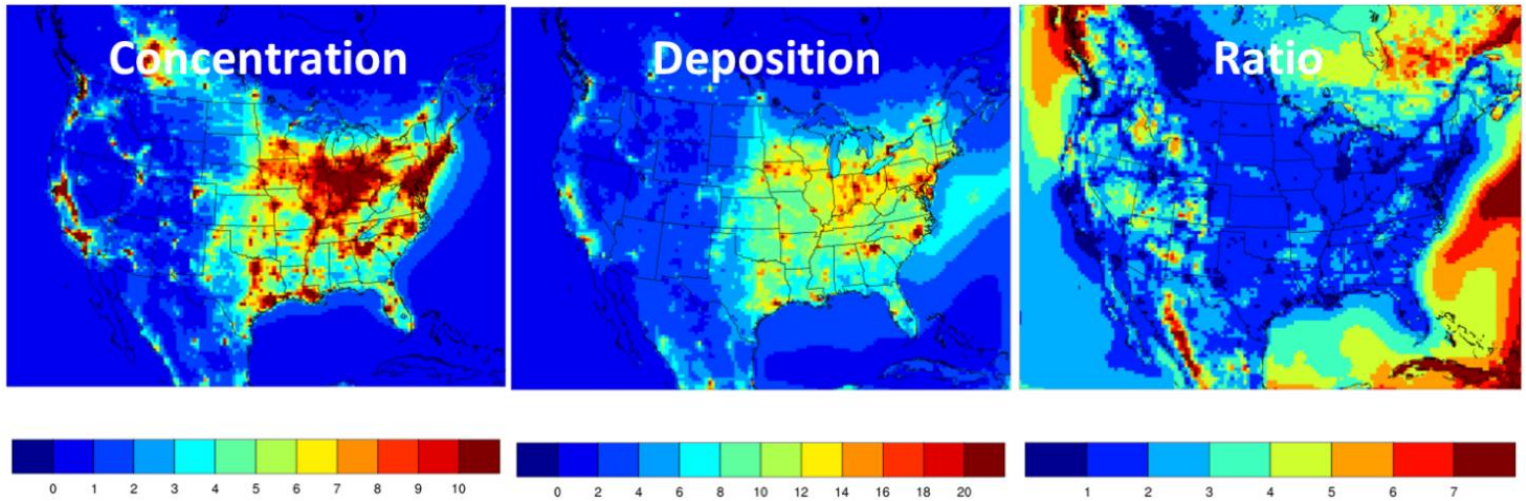

Figure 6-23. Annual average N oxides concentration, ppb (left), total N deposition, kg/ha-yr, (middle), and associated deposition:concentration ratios (right), as estimated from a 21-year (1990-2010) CMAQ simulation.

To further assess potentially influential chemical predictors of S and N deposition rates, we considered the CMAQ model results in more detail, evaluating data from the grid cells containing the 27 Class I area monitoring sites identified in Table 6-3. For Figures 6-24, 6-25, and 6-26, a histogram of each deposition or concentration variable is shown in a diagonal running from the top left to lower right. Below that diagonal are scatter plots and linear regressions for each pair of variables. Above that diagonal are the correlations between pairs of variables, with asterisks indicating p-value thresholds (*** = $p<0.001$). Each data point marks the annual average air concentration and annual total deposition for individual years at that location from the 21-year CMAQ simulation (1990-2010).

Figure 6-24 presents analyses of relationships between CMAQ estimates of annual average concentrations of SO_2 and S deposition in the same grid cells. These analyses indicate moderate correlations of SO_2 concentrations and total S deposition ($r = 0.57$, $p<0.001$). Figure 6-24 also illustrates the relationship between annual average $PM_{2.5}$ and S deposition in the same cells. A weaker correlation is seen for $PM_{2.5}$ concentrations with total S deposition ($r = 0.36$, $p<0.001$).

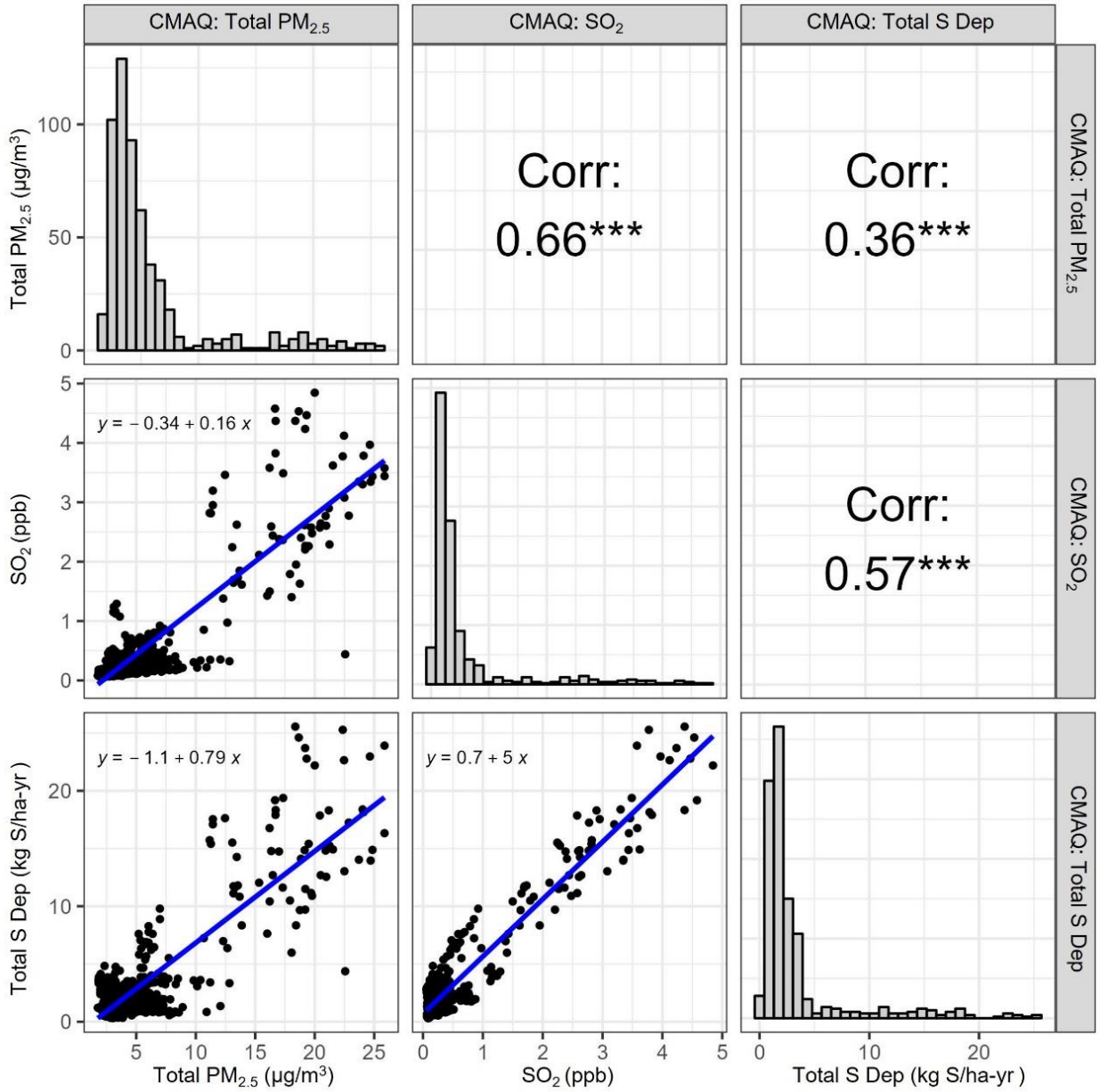

Figure 6-24. Scatter plot matrix of annual average CMAQ-simulated total S deposition versus annual average CMAQ-simulated concentrations of SO_2 and $PM_{2.5}$ for 27 grid cells in Class 1 areas from a 21-year simulation (1990-2010).

Using the same CMAQ simulations described in the above paragraph, Figure 6-25 illustrates the model relationships between total deposition of oxidized nitrogen (i.e., NO, NO_2, and NO_3^-) and air concentrations of NO_2 and particulate nitrate (NO_3^-) at the 27 Class 1 areas. Total oxidized nitrogen deposition has moderate correlations with both NO_2 ($r = 0.64$, $p<0.001$) and particulate nitrate ($r = 0.61$, $p<0.001$). The similarity in correlations among these variables is expected given chemical transformation of NO_2 to NO_3^- in the atmosphere. All the univariate

linear regressions generated with the CMAQ simulations in Figure 6-25 have positive correlations.

To better understand some of the patterns seen in Figure 6-25, we added an additional parameter in Figure 6-25 with the CMAQ-estimated NH_3 concentration. Concentrations of NH_3 are represented by color coding the data points. One notable feature of panels A, B, and C in Figure 6-25 are the existence of distinct groups of points in the data. In panel A, it can be observed that the particulate nitrate and NO_2 relationship varies as a function NH_3 concentrations, with a sharp divide at 1 $\mu g/m^3$ of particulate nitrate. In grid cells where the NH_3 concentrations are low, particulate nitrate is also low and largely independent of how much NO_2 exists. In grid cells where ammonia is higher (i.e., Shenandoah VA and Mammoth Cave KY) there is a strong relationship between particulate nitrate and NO_2. This contributes to the bi- and trifurcations in panels B and C. Panel B appears to show at least two, and possibly three groups of points with similar slopes. This suggests that site-specific correlations are likely higher than when all 27 areas are combined, and in particular, that NO_2 is strongly correlated with oxidized N deposition at the site-level. Panel C shows the relationship between particulate nitrate and total oxidized nitrogen deposition is strongest at sites associated with high NH_3 concentrations, and weaker at sites with lower NH_3.

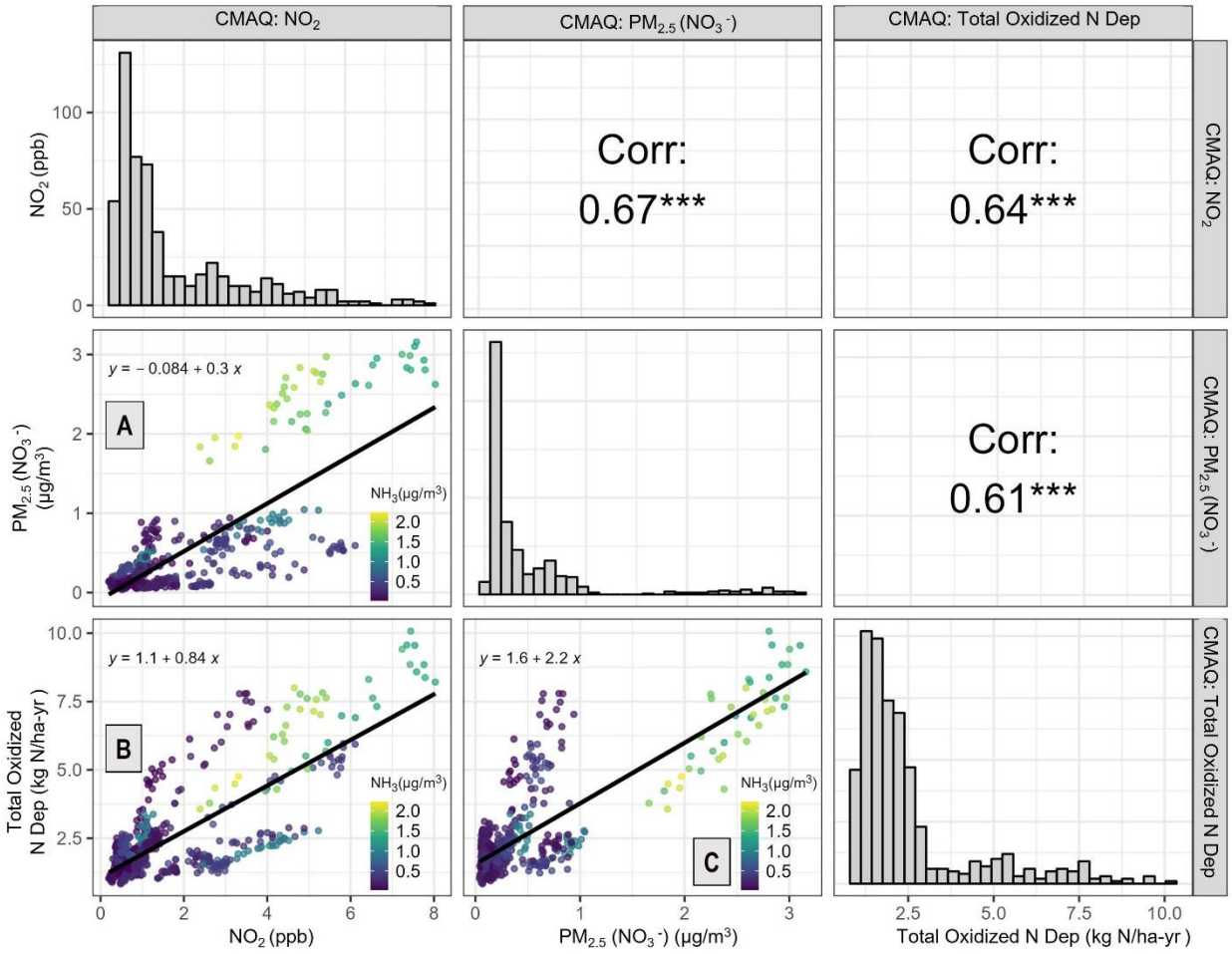

Figure 6-25. Scatter plot matrix of annual average CMAQ-simulated total oxidized nitrogen deposition versus annual average CMAQ-simulated concentrations of NO_2 and particulate nitrate for 27 Class 1 areas from 1990-2010. Colors in scatterplots indicate NH_3 concentrations.

Finally, Figure 6-26 indicates relationships between total reduced N deposition (i.e., NH_3 + NH_4^+) and NH_3 and ammonium in the same CMAQ simulations used for Figures 6-24 and 6-25. The correlation between total reduced N deposition and $PM_{2.5}$ (NH_4^+) is moderate ($r = 0.71$, $p<0.001$). The correlation between reduced N deposition and NH_3 is weaker ($r = 0.50$, $p<0.001$). All of the linear regressions generated with the CMAQ simulations in Figure 6-26 have positive correlations. As in Figure 6-25, there are bi-and trifurcations in the scatterplots (panels A, B, and C) and to better understand the causes of those different patterns, each data point in Figure 6-26 was color coded in terms of the model NO_2 concentration to investigate further. Panel B of Figure 6-26 illustrates that while the correlation between NH_4^+ and reduced N deposition is relatively higher than other concentration:deposition associations ($r = 0.71$), there is considerable scatter in that relationship when NO_2 concentrations are high (lighter colors). Conversely, in

Panel C of Figure 6-26, we see stronger associations between NH_3 and reduced N deposition when NO_2 is high.

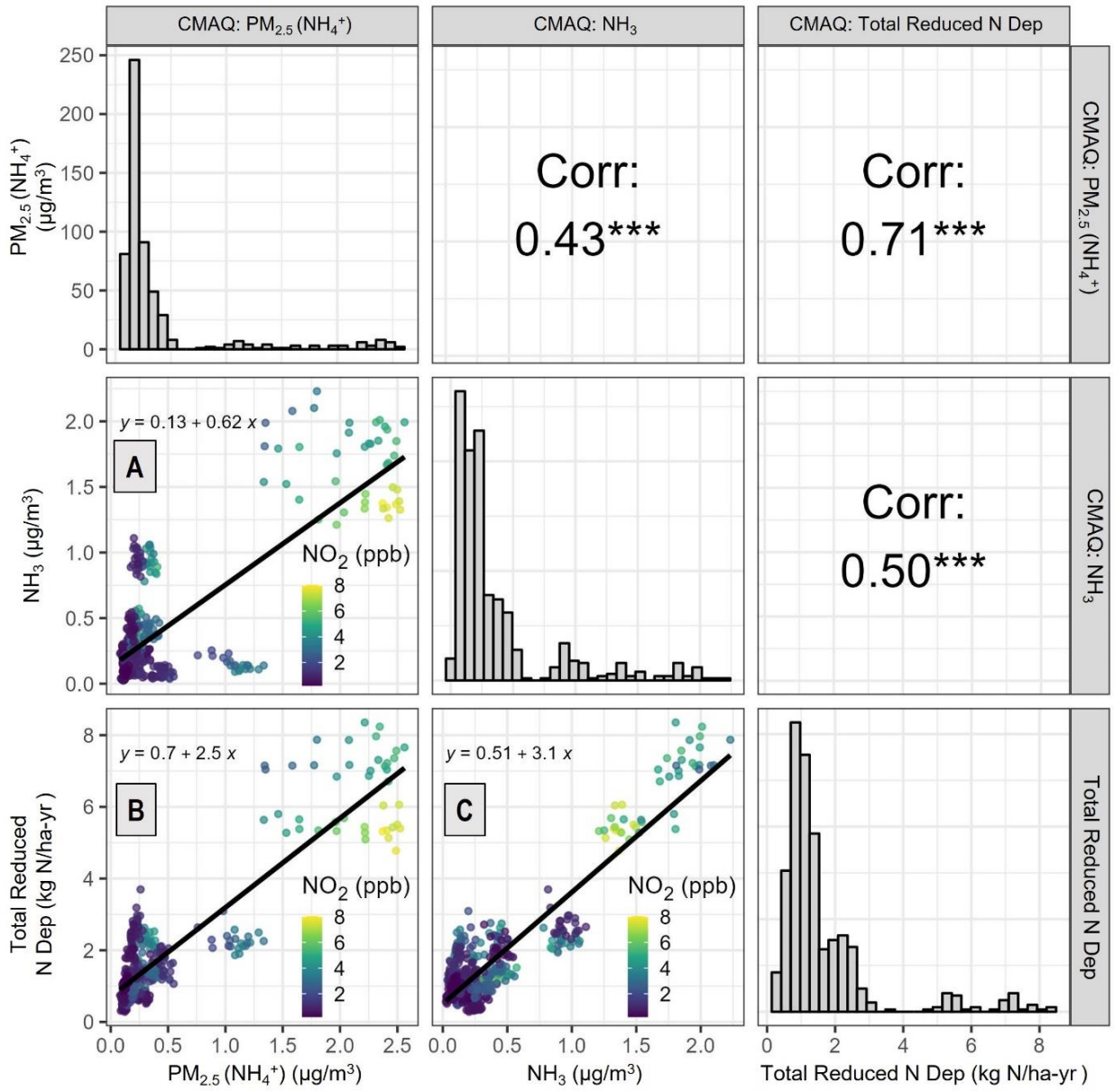

Figure 6-26. Scatter plot matrix of annual average CMAQ-simulated total reduced nitrogen deposition versus annual average CMAQ-simulated concentrations of $PM_{2.5}$ (NO_3^-) and NH_3 for 27 Class 1 areas from 1990-2010. Colors in scatterplots indicate NO_2 concentrations.

In summary, a CMAQ-based analysis of simulated air quality concentrations and deposition data over a 21-year simulation indicates that there is evidence of a moderate positive correlation at the 27 collocated sites between:

- Total S deposition and SO_2 concentrations at the sites
- Oxidized N deposition and NO_2 and particulate nitrate
- Reduced N deposition and ammonium and NH_3 concentrations.

Conversely, we see weaker association between $PM_{2.5}$ and S deposition in the CMAQ simulations. Further, there is evidence that the strength of the associations can vary by location and can be influenced by concentrations of other pollutants contributing to N or S deposition. Additionally, it is important to remember that the model simulations are highly parameterized and are developed to be simplified approximations of highly complex processes. In that regard, these CMAQ comparisons of concentration and deposition are best viewed as informative associations based on modeled physics.

6.2.2.2 Relationships between Air Quality and Wet Deposition Observations

This section evaluates how wet deposition measurements from the NADP monitoring network relate to ambient air measurements from the IMPROVE and CASTNET monitoring networks, at the sites listed in Table 6-3 above. Parameters considered from the NADP network include wet deposition of SO_4^{2-}, NO_3^- and NH_4^+, as well as the sum of NO_3^- and NH_4^+ (as wet deposition of N). For Figures 6-27 through 6-30, a histogram of each deposition or concentration variable is shown in a diagonal running from the top left to lower right. Below that diagonal are scatter plots for each pair of variables. Above that diagonal are the correlations between pairs of variables, with asterisks indicating p-value thresholds (*** = $p<0.001$).

Figure 6-27 shows the relationship between wet S deposition (NADP) and ambient air concentrations of SO_4^{+2} (IMPROVE) and of total S ambient air concentrations (SO_2 + SO_4^{+2}; CASTNET). Concentrations of SO_4^{+2} from IMPROVE have a strong positive relationship with CASTNET total S concentrations ($r = 0.76$, $p<0.001$). Both total S and SO_4^{+2} also indicate a moderate relationship with wet deposition of S measured at the same sites ($r = 0.52$, $p<0.001$ for CASTNET total S and $r = 0.59$, $p<0.001$ for IMPROVE SO_4^{+2}).

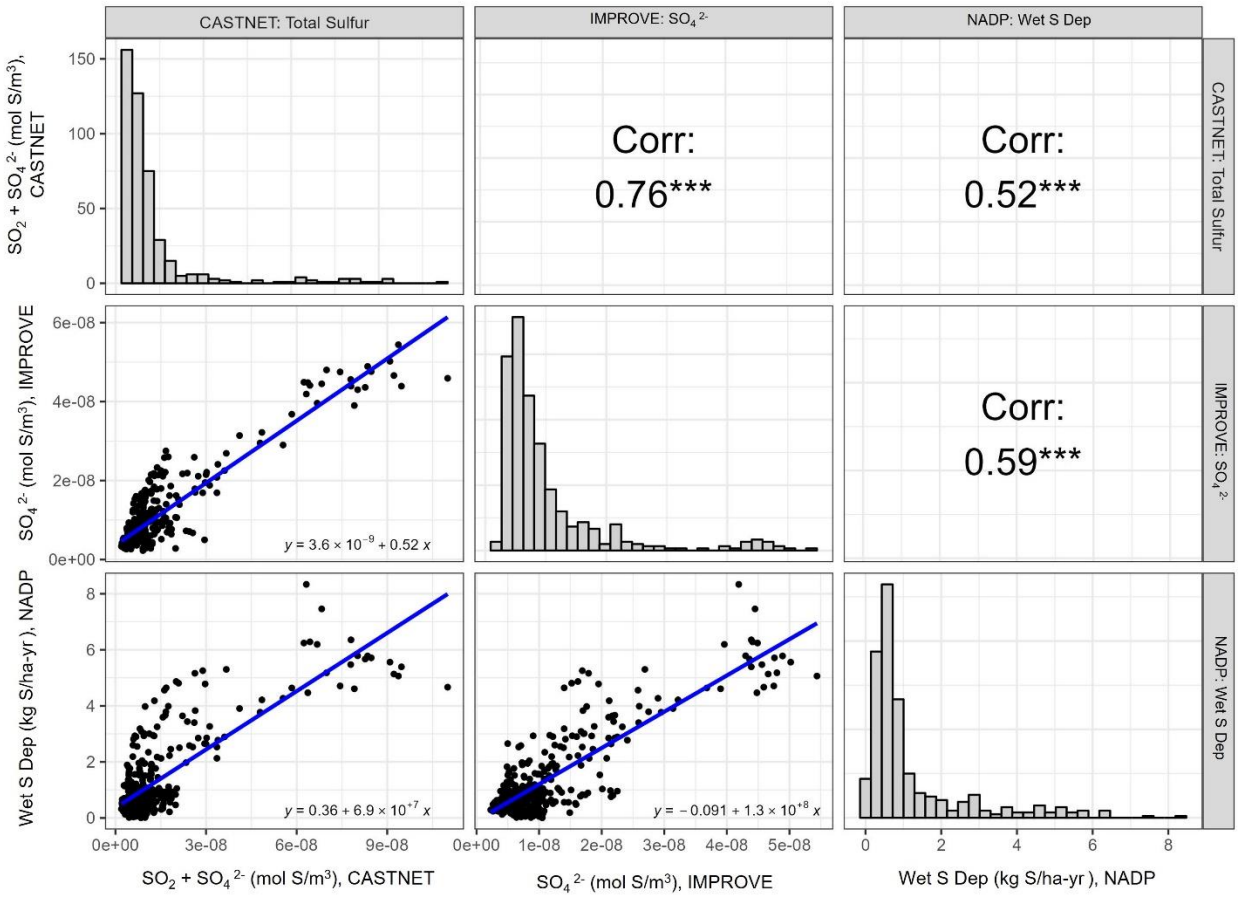

Figure 6-27. Scatter plot matrix of annual average wet S deposition (NADP) with annual average concentrations of SO_4^{2-} (IMPROVE) and total S (SO_2 + SO_4^{2-}, CASTNET) concentrations for 27 Class 1 areas (2000-2019).

For nitrogen, Figure 6-28 shows a strong relationship of CASTNET TNO_3 (HNO_3 and particulate NO_3^-) with IMPROVE NO_3^- ($r = 0.86$, $p<0.001$). This is not unexpected given that one parameter (NO_3^-) is a subset of the other (TNO_3). The relationship of N wet deposition with those parameters includes considerable scatter, with low correlations ($r = 0.32$, $p<0.001$ for NO_3^- alone [IMPROVE] and $r = 0.22$, $p<0.001$ for TNO_3). This may reflect the influence of reduced N compounds on wet N deposition, which may vary among the Class I sites.

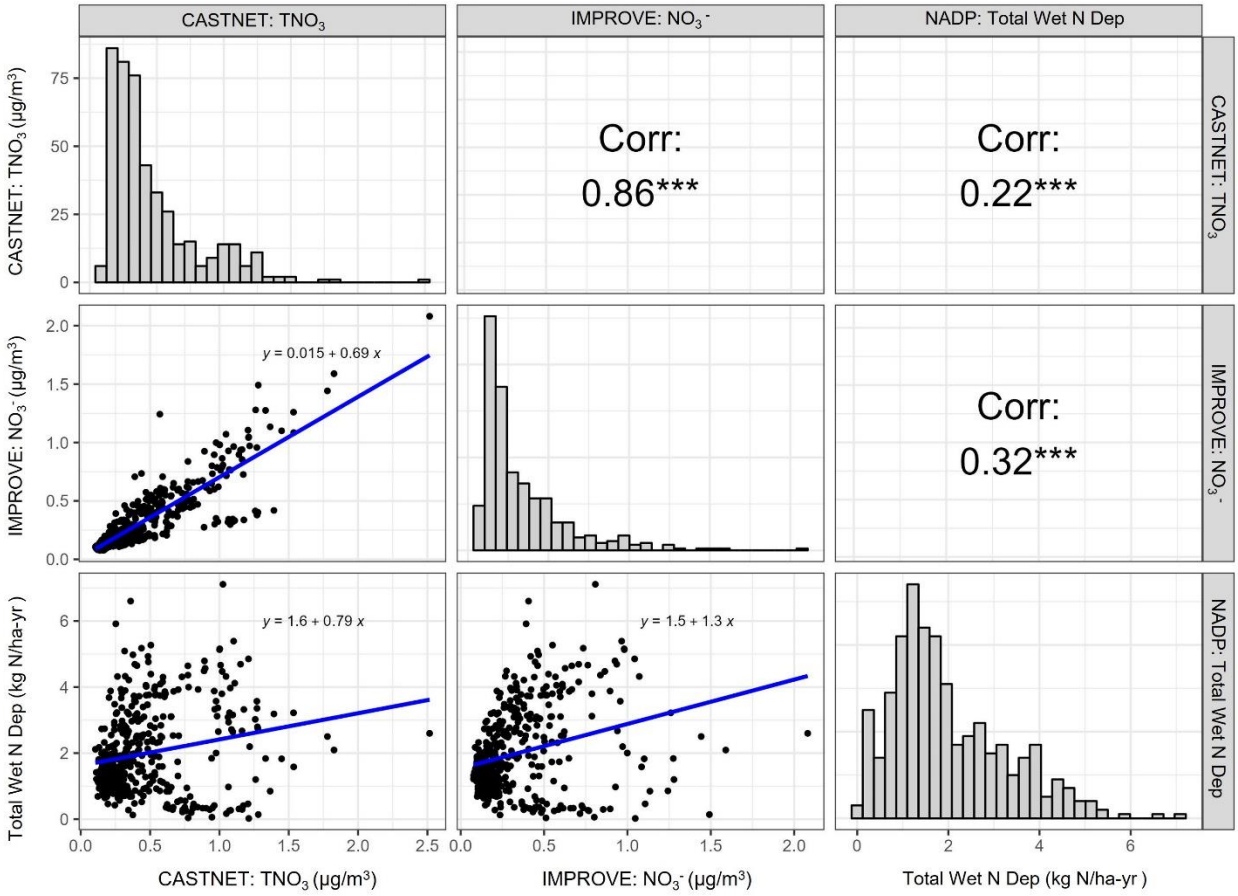

Figure 6-28. Scatter plot matrix of annual average wet N deposition (NADP) with annual average TNO_3 (CASTNET) and NO_3^- (IMPROVE) concentrations for 27 Class 1 areas (2000-2019).

Figure 6-29 shows relationships between wet deposition of N and wet deposition of each of its primary particulate components (NH_4^+ and NO_3^-) based on NADP monitoring data. Both wet deposition of NH_4^+ and NO_3^- are highly correlated with wet deposition of the sum of the two ($r = 0.96$, $p<0.001$ for NH_4^+ and $r = 0.93$, $p<0.001$ for NO_3^-). The correlation between NH_4^+ wet deposition and NO_3^- wet deposition is slightly weaker but still strong ($r = 0.79$, $p<0.001$).

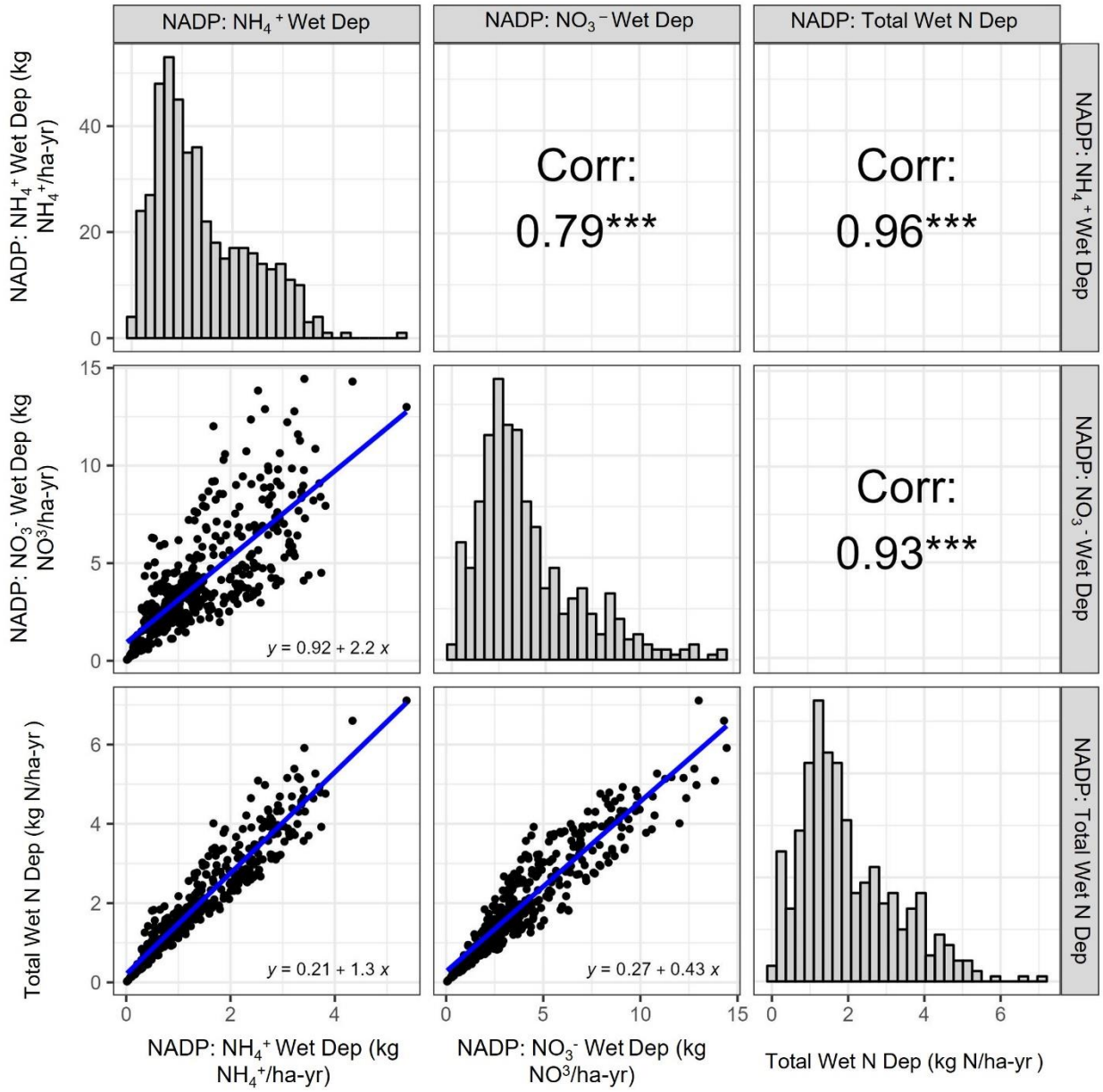

Figure 6-29. Scatter plot matrix of annual average wet N deposition (NADP) with annual average wet deposition of NH_4^+ and NO_3^- (NADP) deposition for 27 Class I areas (2000-2019).

Figure 6-30 presents wet deposition data for N and S (from NADP) and ambient air concentrations of $PM_{2.5}$ (IMPROVE) at the 27 Class I area sites. Somewhat weak but statistically significant correlations are observed for wet deposition of both N and S with $PM_{2.5}$ (IMPROVE) concentrations ($r = 0.38$, $p<0.001$ for wet S deposition and $r = 0.37$, $p<0.001$ for wet N deposition). The existence of a positive correlation likely reflects the presence of particulate S and N compounds in $PM_{2.5}$, and the variability in this relationship may reflect variation in $PM_{2.5}$ composition across the 27 Class I areas. The high correlation between wet deposition of nitrogen and of sulfur ($r = 0.84$, $p<0.001$) may be related to the role of precipitation rate in wet deposition.

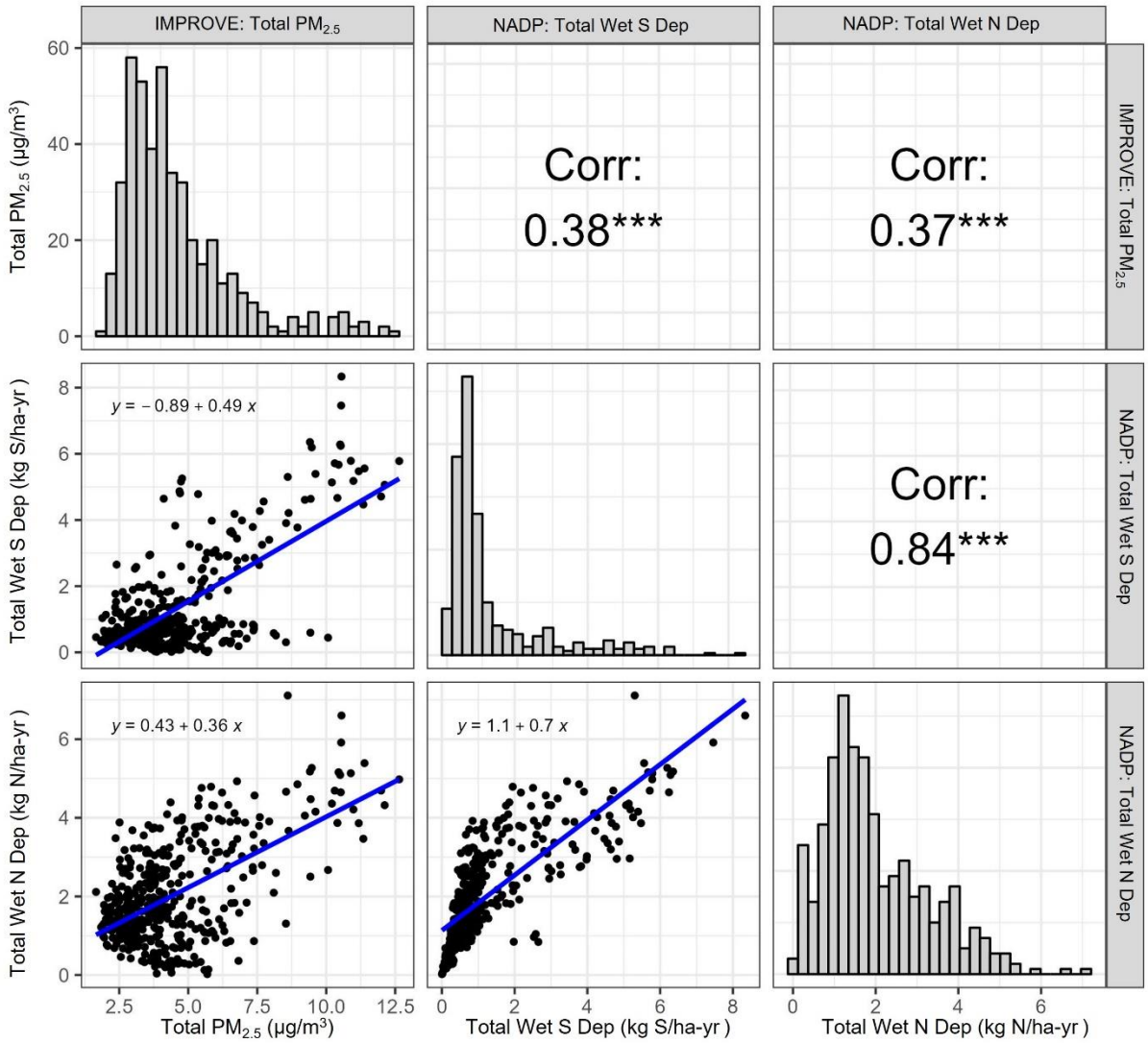

Figure 6-30. Scatter plot matrix of annual average wet deposition of N and S (NADP) with annual average $PM_{2.5}$ (IMPROVE) for 27 Class 1 areas (2000-2019).

6.2.2.3 Relationships between Observed Air Quality and TDep Estimates of Deposition

We next consider the extent of relationships between TDep-estimated total S deposition and $PM_{2.5}$ (total mass), particulate SO_4^{2-} and total S (SO_2 plus particulate SO_4^{2-}) using IMPROVE and CASTNET data at the 27 Class I area sites from 2000-2019 in Figure 6-31.[5] This figure indicates that air quality concentrations are lower at these sites in recent periods relative to the past. Additionally, as noted above for wet deposition, total S deposition appears to have only weak association with $PM_{2.5}$ (IMPROVE, $r = 0.33$, $p<0.05$), and the correlation of TDep estimated sulfur deposition and measured $PM_{2.5}$ is not as strong as that of TDep estimated sulfur deposition and SO_4^{2-} ($r = 0.55$, $p<0.05$). The strongest associations are seen for S deposition with total sulfur ($SO_4^{2-} + SO_2$) from CASTNET monitors ($r = 0.61$, $p<0.05$, Figure 6-31).

For total nitrogen deposition, concentrations of annual average $PM_{2.5}$ (IMPROVE), annual average NO_3^- (IMPROVE), and TNO_3 (CASTNET) are all associated with TDep-estimated total N deposition.[6] Of the ambient air concentrations from IMPROVE and CASTNET shown in Figure 6-32, NO_3^- had the strongest correlations with TDep estimates of total N deposition ($r = 0.63$, $p<0.05$). TDep estimates of total N deposition had the weakest correlation with $PM_{2.5}$ measurements from IMPROVE ($r = 0.53$, $p<0.05$). All three ambient air concentrations had positive correlations with TDep estimates of total N deposition. Linear regressions run on all three ambient air concentrations and their associated TDep N depositions were positive and significant.

[5] Deposition estimates in Figure 6-31 are based TDep version v2018.02, downloaded on March 7, 2021.

[6] Deposition estimates in Figures 6-32 and 6-33 are based TDep version v2018.02, downloaded on March 7, 2021

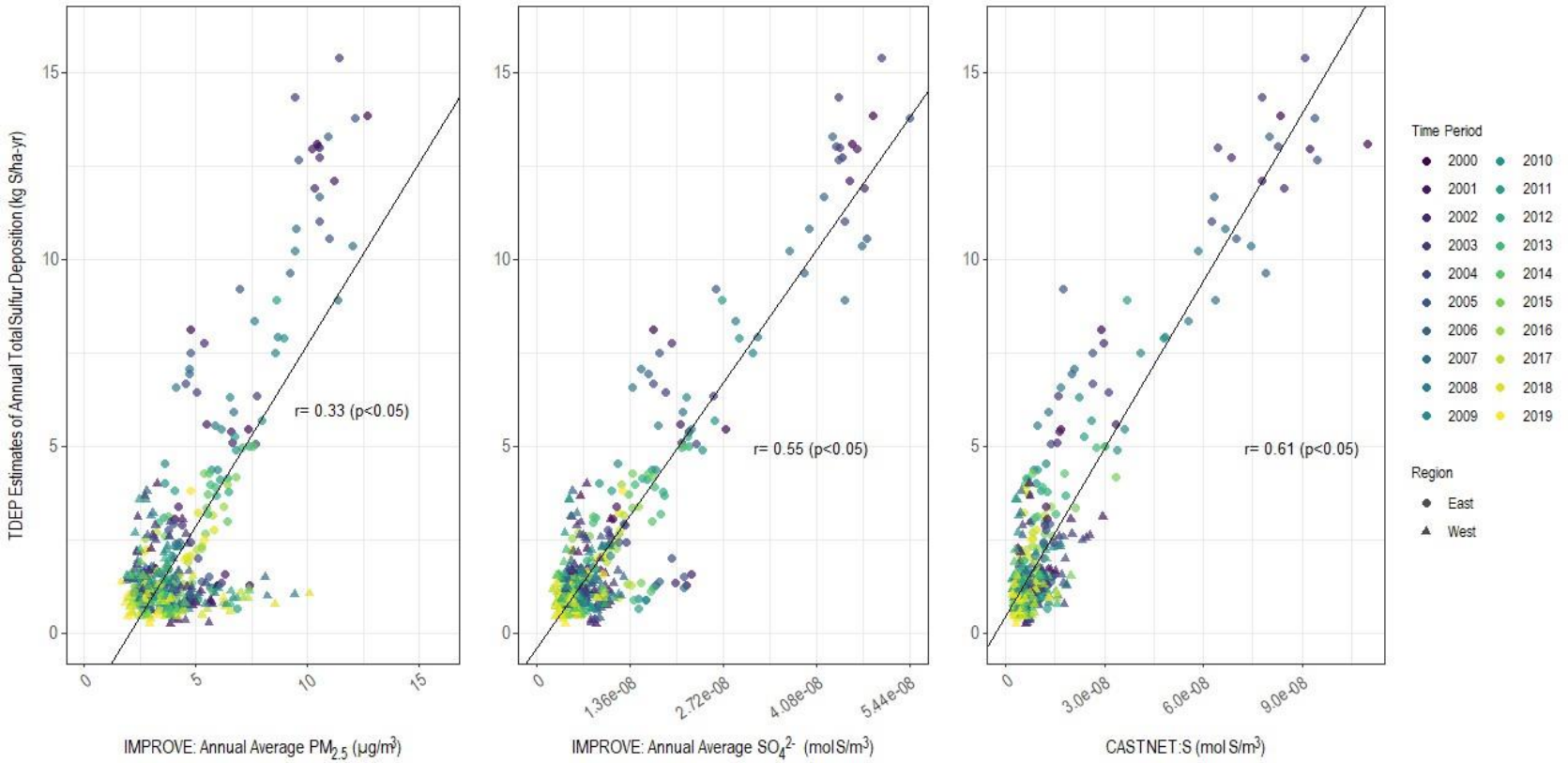

Figure 6-31. Total S deposition (TDep) *versus* annual average ambient air concentrations (2000–2019) of $PM_{2.5}$ (left; IMPROVE), SO_4^{2-} (center; IMPROVE) and total S (right; CASTNET) at 27 Class I area sites. Linear regressions are shown as black lines.

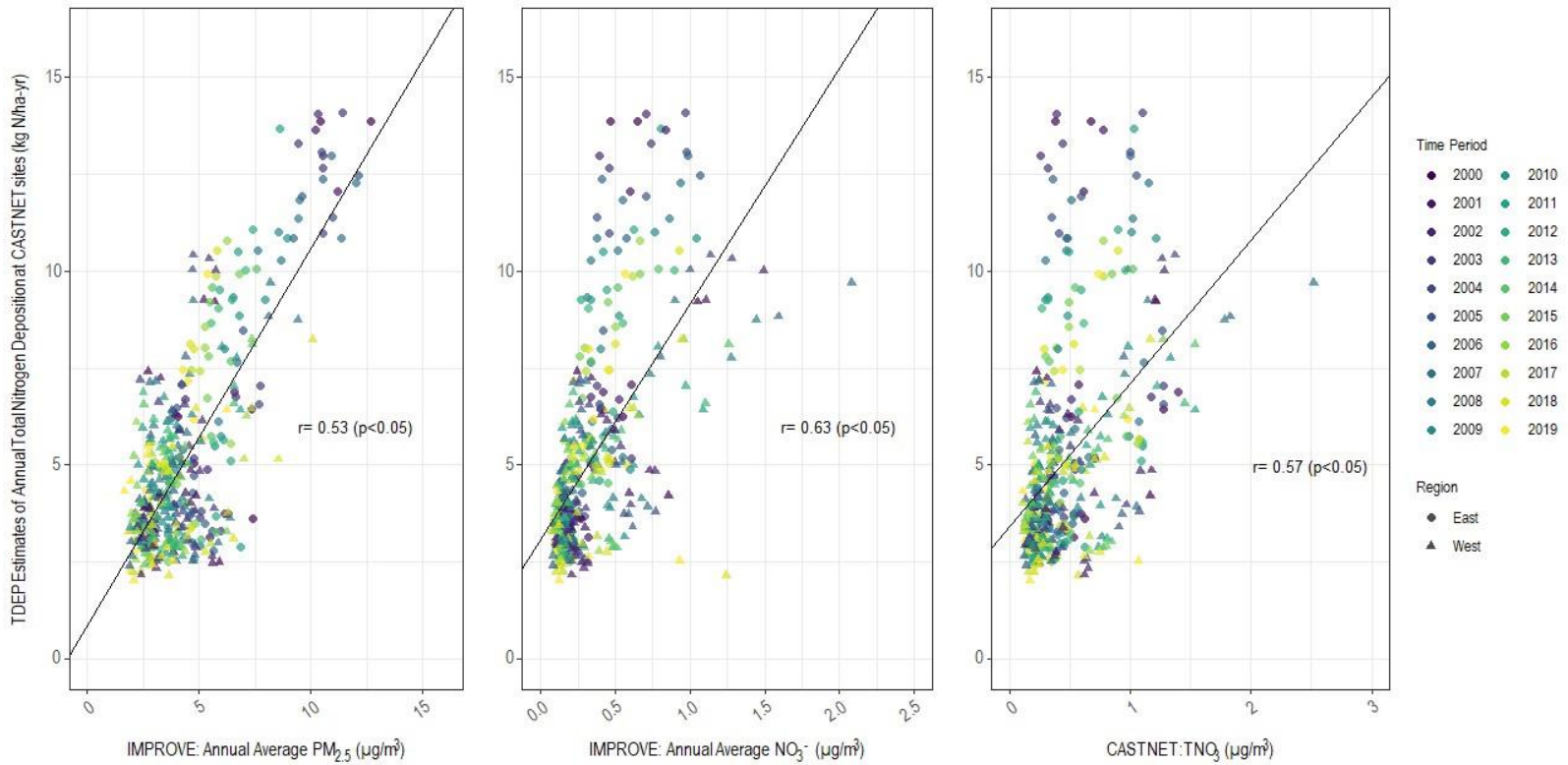

Figure 6-32. Total N deposition (TDep) *versus* annual average ambient air concentrations (2000-2019) of $PM_{2.5}$ (left; IMPROVE), annual average NO_3^- (center; IMPROVE), and TNO_3 (right; CASTNET) at 27 Class I area sites. Linear regressions are shown as black lines.

Estimated total N deposition (TDep) at the 27 Class I area sites are related to air concentrations of N species (IMPROVE and CASTNET) at those sites (Figure 6-33). Total N deposition estimates are moderately correlated with IMPROVE total N (NH_4^+ and NO_3^-) concentrations ($r = 0.62$, $p<0.05$), CASTNET total N (NH_4^+ and NO_3^-) concentrations ($r = 0.62$, $p<0.05$), and CASTNET NH_4^+ concentrations ($r = 0.62$, $p<0.05$). Note that IMPROVE ammonium is estimated assuming that the nitrate and sulfate are fully neutralized by ammonia. Although IMPROVE NH_4^+ is not directly measured, the total N deposition-concentration correlation using IMPROVE is similar to that of CASTNET.

Greater scatter is observed in the relationships between wet N deposition (NADP) and IMPROVE total N concentrations, measured CASTNET total N concentrations, and CASTNET NH_4^+ concentrations (Figure 6-34). Accordingly, the correlation coefficients are lower, ranging from 0.31 for wet N deposition (NADP) with CASTNET total N concentrations to 0.47 for wet N deposition (NADP) with CASTNET NH_4^+ concentrations (Figure 6-34).

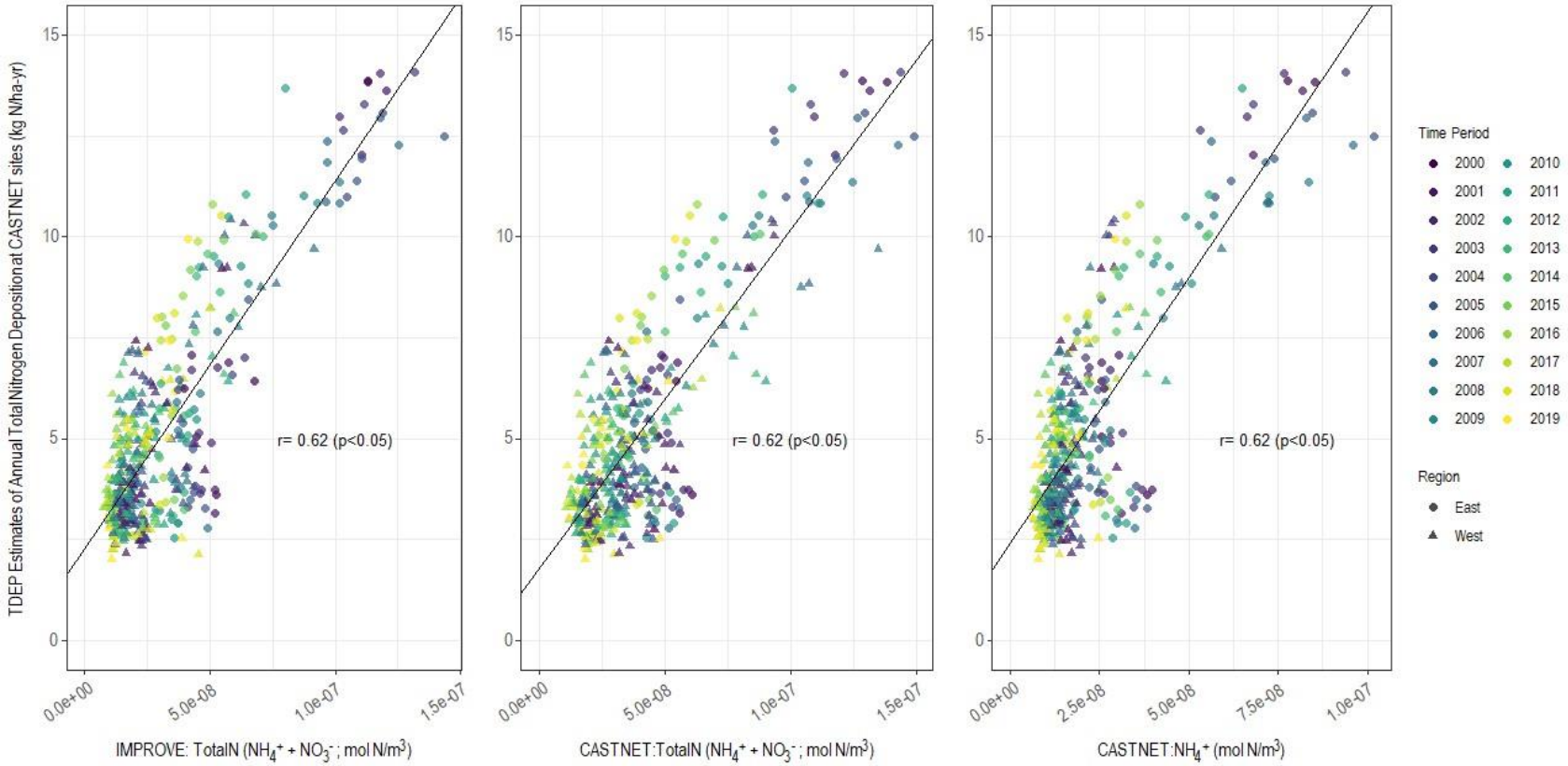

Figure 6-33. Total N deposition (TDep) *versus* annual average ambient air concentrations (2000-2019) of total particulate N (left; IMPROVE), total particulate N (center; CASTNET), and NH_4^+ (right; CASTNET) at 27 Class I area sites. Linear regressions are shown as black lines.

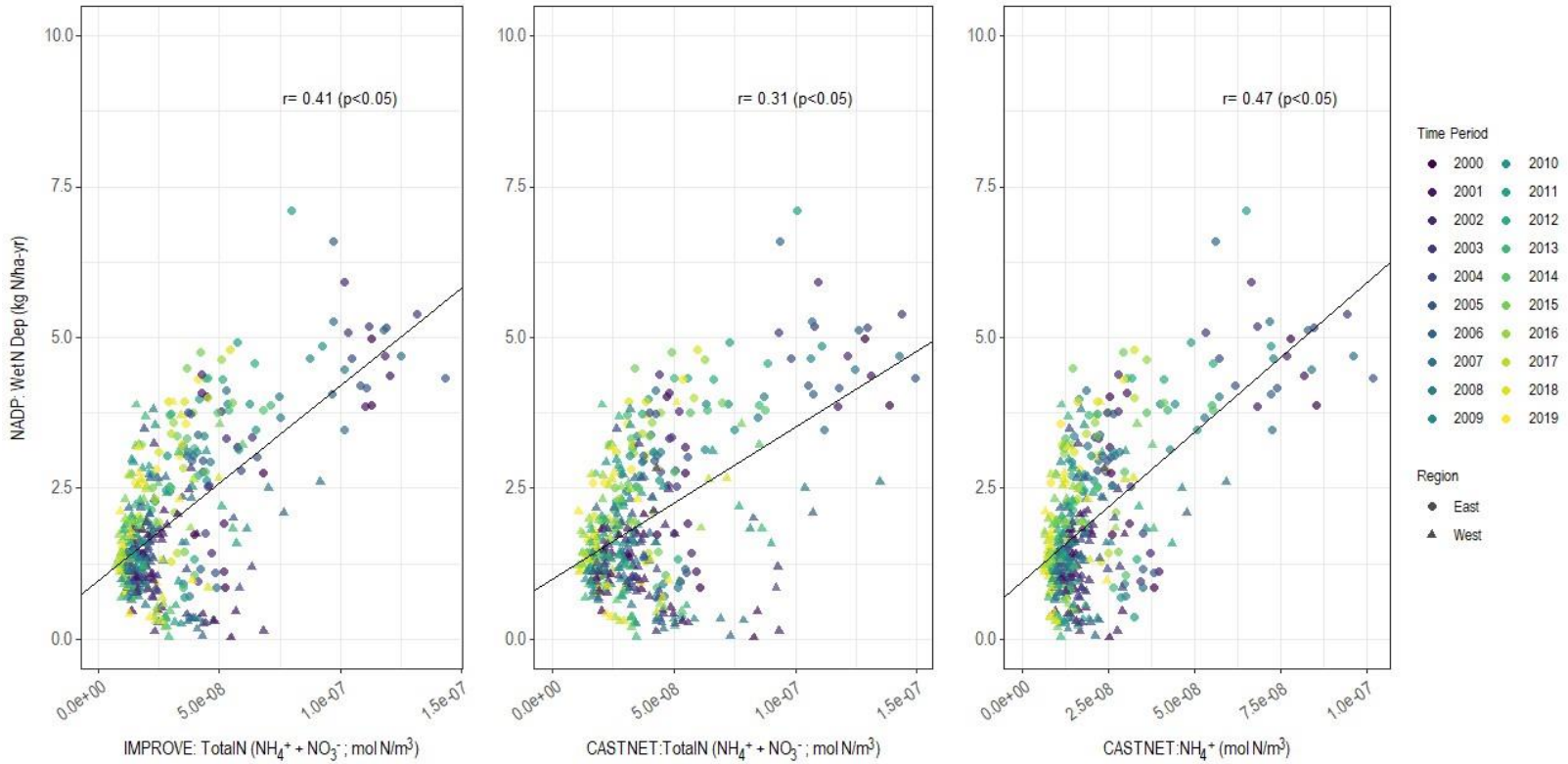

Figure 6-34. Wet N deposition (NADP) *versus* annual average ambient air concentrations (2000-2019) of total particulate N (right; IMPROVE), total particulate N (center; CASTNET), and NH_4^+ (right; CASTNET) at 27 Class I area sites. Linear regressions are shown as black lines.

6.2.2.4 Conclusions

The above analyses focus on characterizing relationships between various chemical species that are the air quality components of S and N deposition and of S and N air concentrations over longer time periods (e.g., annual observations or 21-year CMAQ simulations) in more rural locations by assessing various forms of available information collocated (measured and estimated) at 27 sites in Class I areas. Assessment of these various forms of information generally show consistency in the observed relationships. For air concentrations of S compounds (SO_4^{2-} or SO_2+SO_4^{2-}) and deposition of S, the analyses suggest that in more rural locations, such as those represented by these 27 Class I areas, S deposition is moderately associated with measurements of particulate SO_4^{2-} (r = 0.59, Figure 6-27) and the combination of SO_2+SO_4^{2-} (r = 0.52, Figure 6-27). There is a slightly weaker association between wet S deposition and $PM_{2.5}$ (r = 0.38, Figure 6-30) in these rural locations, marked by more variability. This variability and generally lower association likely relates to the fact that some percentage of the $PM_{2.5}$ mass is expected to be composed of compounds other than sulfate. These results suggest that total S deposition in rural areas is mostly resulting from deposition of sulfate and SO_2. This is consistent with our understanding of the chemical properties and physical transport of these compounds. For example, we know that fine particles, such as $PM_{2.5}$, have a much slower dry deposition velocity and remain in the atmosphere longer (Table 6-1). Thus, it is not surprising to see that sulfur can be transported as $PM_{2.5}$ in these rural locations. These results also indicate that among $PM_{2.5}$ (IMPROVE), SO_4^{2-} (IMPROVE), and total S (SO_2+SO_4^{2-}, CASTNET), total S (CASTNET) shows the strongest relationship with total sulfur deposition (Figure 6-31). For nitrogen, these results suggest that wet deposition of N in these rural areas has little association with air concentrations of TNO_3 (r = 0.22, Figure 6-28) while having a strong correlation with particulate nitrate (r = 0.86, Figure 6-28). Lower, somewhat moderate correlations are observed for total N deposition in these locations with $PM_{2.5}$ (IMPROVE, r = 0.53), NO_3^- (IMPROVE, r = 0.63), and TNO_3 (CASTNET, r = 0.57, Figure 6-32).

6.2.3 National SLAMS Network – Relationships Between Air Concentrations and Deposition

In this section, we consider ambient air concentrations and deposition estimates for the period 2001 to 2020[7] at the SLAMS monitors that employ FRM/FEM and collect data for NAAQS surveillance purposes. As with the analyses in the sections 6.2.1 and 6.2.2, the analyses

[7] Deposition estimates in Figures 6-35 to 6-39 and Tables 6-4 to 6-7 are based on TDep version: v2022.02. TDep data for Figures 6-35 and 6-37 through 6-39 were downloaded on September 7, 2022. TDep data for 6-36 were downloaded on August 30, 2023.

here focus on "local" concentrations and "local" deposition. This analysis will be most illustrative of concentration and deposition relationships where the deposition results primarily from local sources of the pollutants. Further, this analysis incorporates a national-scale consideration of criteria pollutant concentrations measured at the ambient air monitors used to judge attainment of the current secondary NAAQS for oxides of nitrogen, oxides of sulfur and $PM_{2.5}$. The locations for the SLAMS that were active in the 2019-2021 period are shown in Figures 2-11, 2-12 and 2-13 for NO_2, SO_2 and $PM_{2.5}$, respectively.

Figure 6-35 illustrates the relationship between SO_2 annual average concentrations (averaged over 3 years) across the U.S. and the S deposition at these locations.[8] When looking at the five time periods used in Chapter 5, there is a strong positive, significant association between annual SO_2 in the eastern[9] U.S. and the S deposition at those locations (Figure 6-35, $r = 0.79$, $p<0.05$, slope= 1.84). The figure suggests, however, that this association has become weaker over the most recent 3-year averages as SO_2 levels have decreased sharply across the eastern U.S. This is also reflected in the correlation coefficients for total sulfur deposition and SO_2 annual average concentrations at eastern SLAMS monitors (Table 6-4) for 2014-2016 ($r = 0.40$, $p<0.05$) and 2018-2020 ($r = 0.28$, $p<0.05$).

[8] There are two outlier SO_2 data points in the 2018-2020 period which have been removed from the plots and correlation calculations involving S deposition in this section. These data are driven by a location in southeastern MO where annual average SO_2 has exceeded 20 ppb in recent years. A preliminary analysis suggests that these SO_2 measurements reflect a relatively recent source that was not modeled in the CMAQ simulation that informed the TDep estimates of deposition. As there is no deposition monitor in the immediate vicinity of the source it is unlikely that the TDep estimates are capturing the impacts of this source. For that reason, we concluded it appropriate to exclude these data from evaluations of the concentration-deposition relationship.

[9] The East and West categorization of sites in this section is the same as that used in the aquatic acidification REA in section 5.1. That is, sites in ND, SD, CO, WY, MT, AZ, NM, UT, ID, CA, OR, WA (2009 REA, Appendix 1, p. 1-21) are designated West, and all other sites (which are in locations from the eastern U.S. out into the Great Plains) are designated East.

Table 6-4. Correlation coefficients for TDep-estimated S deposition and annual average SO_2 concentrations (averaged over three years) at SLAMS sites, by time period and region.

Sulfur Deposition and SO_2	SLAMS Total Deposition	Wet Deposition		Dry Deposition	
Annual DV-All Ecoregions	Correlation Coefficient (r)= 0.70*	*Annual DV-All Ecoregions*	r = 0.66*	*Annual DV- All Ecoregions*	r = 0.72*
Year	r	Year	r	Year	r
2001 - 2003	0.64*	2001 - 2003	0.67*	2001 - 2003	0.62*
2006 - 2008	0.72*	2006 - 2008	0.70*	2006 - 2008	0.66*
2010 - 2012	0.54*	2010 - 2012	0.55*	2010 - 2012	0.48*
2014 - 2016	0.37*	2014 - 2016	0.31*	2014 - 2016	0.40*
2018 - 2020	0.19*	2018 - 2020	0.06	2018 - 2020	0.27*
Annual DV-East Ecoregions	r = 0.79*	*Annual DV-East Ecoregions*	r = 0.79*	*Annual DV-East Ecoregions*	r = 0.78*
Year	r	Year		Year	r
2001 - 2003	0.58*	2001 - 2003	0.61*	2001 - 2003	0.57*
2006 - 2008	0.65*	2006 - 2008	0.62*	2006 - 2008	0.59*
2010 - 2012	0.52*	2010 - 2012	0.51*	2010 - 2012	0.47*
2014 - 2016	0.40*	2014 - 2016	0.36*	2014 - 2016	0.42*
2018 - 2020	0.28*	2018 - 2020	0.18*	2018 - 2020	0.37*
Annual DV-West Ecoregions	r = 0.29*	*Annual DV-West Ecoregions*	r = 0.10	*Annual DV-West Ecoregions*	r = 0.30*
Year	r	Year	r	Year	r
2001 - 2003	-0.03	2001 - 2003	-0.18	2001 - 2003	0.06
2006 - 2008	0.25*	2006 - 2008	-0.13	2006 - 2008	0.38*
2010 - 2012	0.03	2010 - 2012	0.02	2010 - 2012	0.16
2014 - 2016	0.33*	2014 - 2016	0.18	2014 - 2016	0.30*
2018 - 2020	0.20	2018 - 2020	0.27*	2018 - 2020	0.09
*p<0.05					
Correlations are Spearman's Rank correlation.					

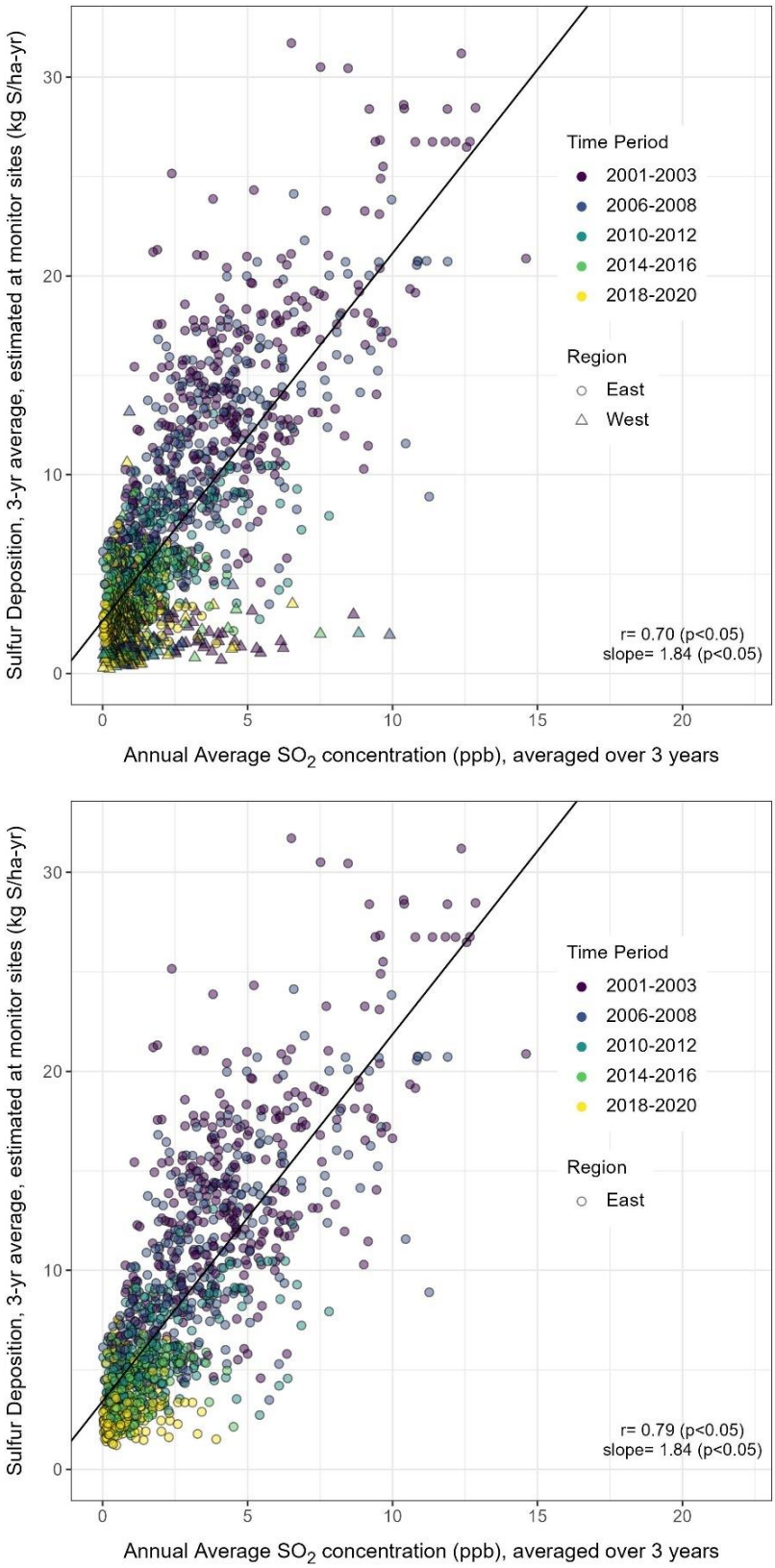

Figure 6-35. TDep estimated S deposition and annual average SO_2 concentrations (3-year average) at SLAMS across the CONUS (upper) and in the East (lower).

Figures 6-36 explores the relationship between annual average SO_2 concentrations and S deposition for dry, and wet deposition separately. As discussed in section 6.1.2, in the generally more arid areas of the West dry deposition tends to dominate, while wet deposition plays a larger role in the East. The correlation coefficients in Table 6-4 provide some indication of this as the correlation coefficient for wet S deposition and SO_2 in the eastern United States ($r = 0.79$, $p<0.05$) is much higher than the correlation coefficient in the western United States ($r = 0.10$, $p<0.05$). For the full dataset of sites across the CONUS, the correlation coefficient for wet S deposition ($r = 0.66$, $p<0.05$) is similar to that for dry S deposition ($r = 0.72$, $p<0.05$).

Figure 6-37 presents scatterplots for TDep-estimated S deposition and design values for the current secondary standard (annual second maximum 3-hour concentration), averaged over three years. Correlation coefficients are presented in Table 6-5. A moderate correlation is observed, although somewhat weaker than for annual average SO_2 concentrations ($r = 0.66$ compared to $r = 0.70$ for all sites and $r = 0.71$ compared to $r = 0.79$ at eastern sites).

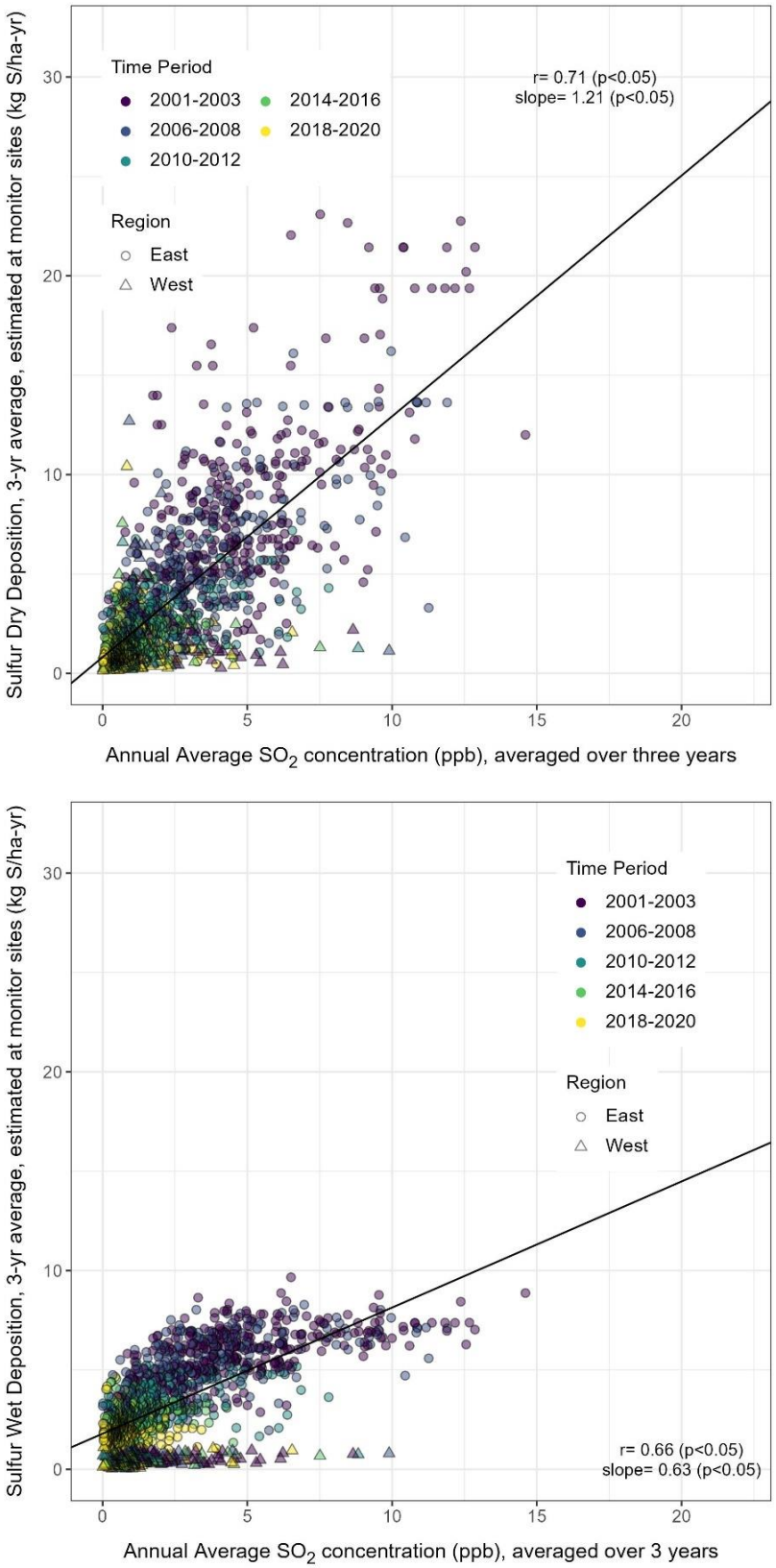

Figure 6-36. TDep-estimated dry S deposition (upper) and wet S deposition (lower) *versus* annual SO_2 concentrations (3-year average) at SLAMS in the CONUS.

Table 6-5. Correlation coefficients for TDEP-estimated S deposition and annual second highest 3-hr SO_2 concentration (averaged over three years), at SLAMS in the CONUS by region and time period.

Sulfur Deposition and SO_2 (3 hr Standard)	SLAMS Total Deposition				
3-hr DV-All Ecoregions	Correlation Coefficient (r) = 0.66*	*3-hr DV-East Ecoregions*	r = 0.71*	*3-hr DV-West Ecoregions*	r = 0.37*
Year	r	Year	r	Year	r
2001 - 2003	0.57*	2001 - 2003	0.52*	2001 - 2003	-0.005
2006 - 2008	0.60*	2006 - 2008	0.46*	2006 - 2008	0.44*
2010 - 2012	0.53*	2010 - 2012	0.55*	2010 - 2012	0.24
2014 - 2016	0.52*	2014 - 2016	0.58*	2014 - 2016	0.27
2018 - 2020	0.40*	2018 - 2020	0.42*	2018 - 2020	0.36*
*p< 0.05					
Correlations are Spearman's Rank correlation.					

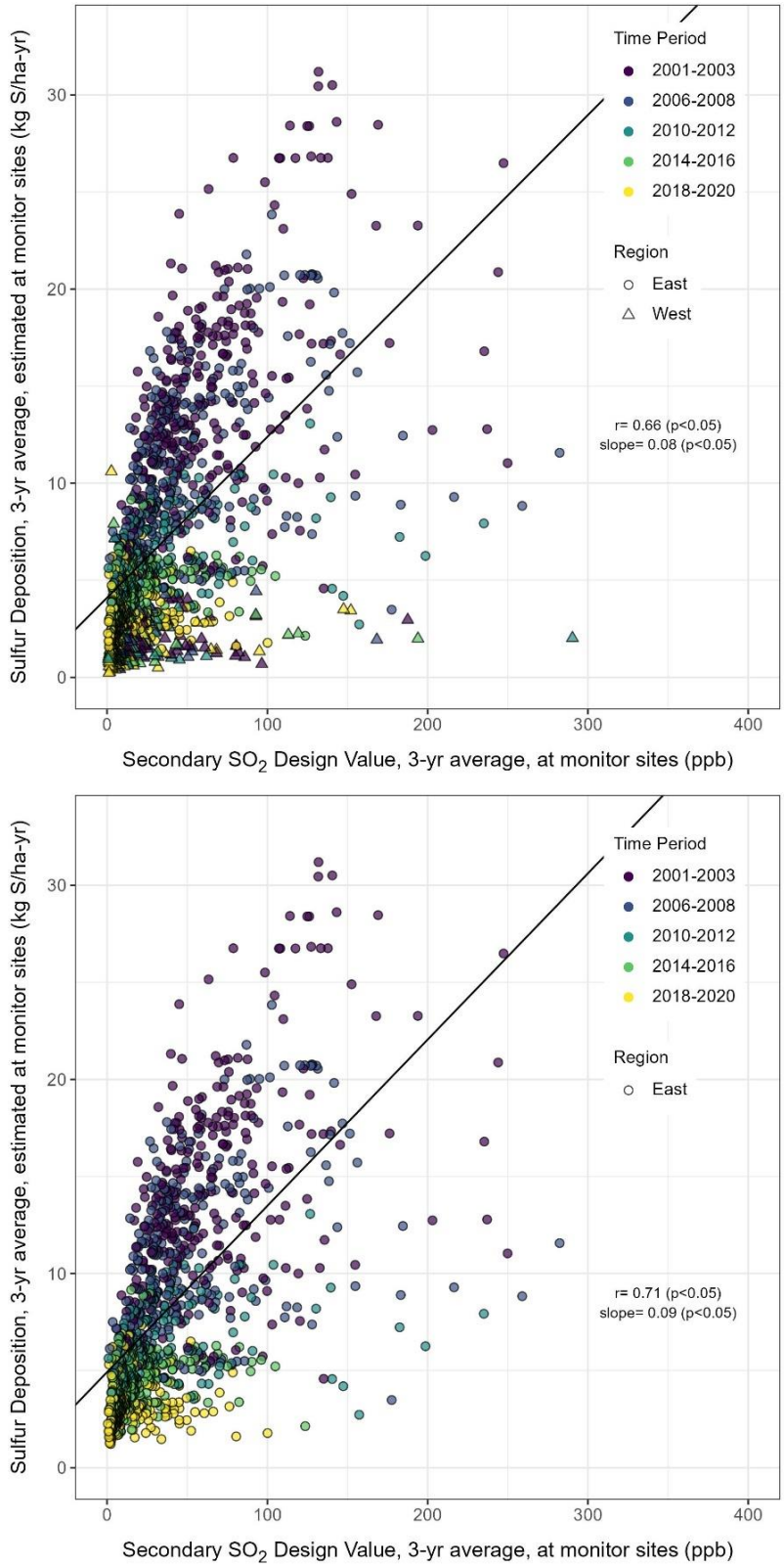

Figure 6-37. TDep estimated total S deposition and design values for the SO_2 secondary standard (annual second maximum 3-hour concentration), averaged over three years, at SLAMS across CONUS (upper) and in East (lower).

Figure 6-38 presents analyses of relationships between TDep N deposition estimates and NO_2 concentrations at SLAMS across the CONUS. Analysis of this dataset for the five time periods indicates a positive but weak association of nitrogen deposition with NO_2 ($r = 0.38$, $p<0.05$), with scatter in the relationship across both eastern and western monitor sites. Unlike for sulfur deposition and SO_2, the time period with the highest correlation coefficient for N deposition and NO_2 is 2014-2016, rather than the earliest time periods (Table 6-6). Also, unlike sulfur deposition *versus* SO_2 concentrations, there is a stronger correlation between N deposition and NO_2 at western SLAMS monitors ($r = 0.63$, $p<0.05$) than eastern sites ($r = 0.44$, $p<0.05$, Table 6-6). Indeed, all of the correlation coefficients are stronger in western sites over all time periods (Table 6-6).

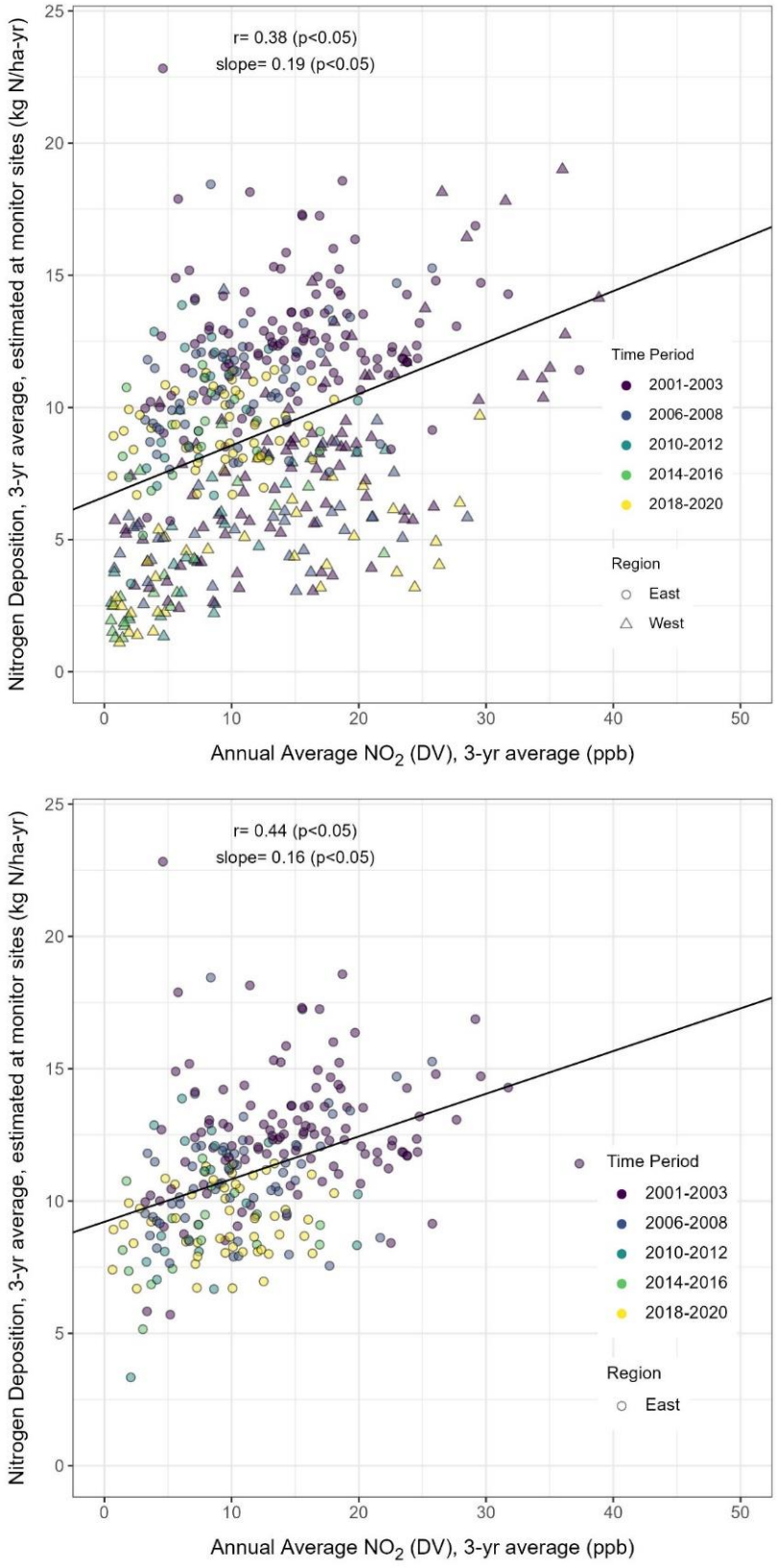

Figure 6-38. TDep-estimated N deposition and annual average NO_2 concentrations (3-year average) at SLAMS across the CONUS (upper), and in the East (lower).

Table 6-6. Correlation coefficients for N deposition (TDep) and annual average NO_2 concentrations (averaged over three years) at SLAMS in the CONUS by region and time period.

Nitrogen Deposition and NO_2	SLAMS Total Deposition				
Annual DV (averaged over three years) -All Ecoregions	Correlation Coefficient (r) = 0.38*	*Annual DV (averaged over three years)- East Ecoregions*	r = 0.44*	*Annual DV (averaged over three years) -West Ecoregions*	r = 0.63*
Year	r	Year	r	Year	r
2001 - 2003	0.31*	2001 – 2003	0.18*	2001 - 2003	0.63*
2006 - 2008	0.10	2006 – 2008	0.35	2006 - 2008	0.44*
2010 - 2012	0.36*	2010 – 2012	0.23	2010 - 2012	0.65*
2014 - 2016	0.60*	2014 – 2016	0.36	2014 - 2016	0.76*
2018 - 2020	0.13	2018 – 2020	0.10	2018 - 2020	0.64*
*p<0.05					
Correlations are Spearman's Rank Correlations.					

In addition to an assessment of N deposition and NO_2 ambient air concentrations, we assessed the relationship between N deposition and annual average $PM_{2.5}$ concentrations at SLAMS across the CONUS (Figure 6-39). Although there is substantial scatter, the correlation is moderate and statistically significant ($r = 0.57$, $p<0.05$). As with S deposition and SO_2 air concentrations, the correlation between N deposition and $PM_{2.5}$ concentrations is lower in the later years (Table 6-7). For example, across the CONUS, the correlation for N deposition and $PM_{2.5}$ concentrations is much higher for the 2001-2003 period ($r = 0.61$, $p<0.05$) than for the 2018-2020 period ($r = 0.20$, $p<0.05$), although both are significant (Table 6-7). This pattern of decreasing correlation coefficients with later years is also observed for the eastern and western subsets.

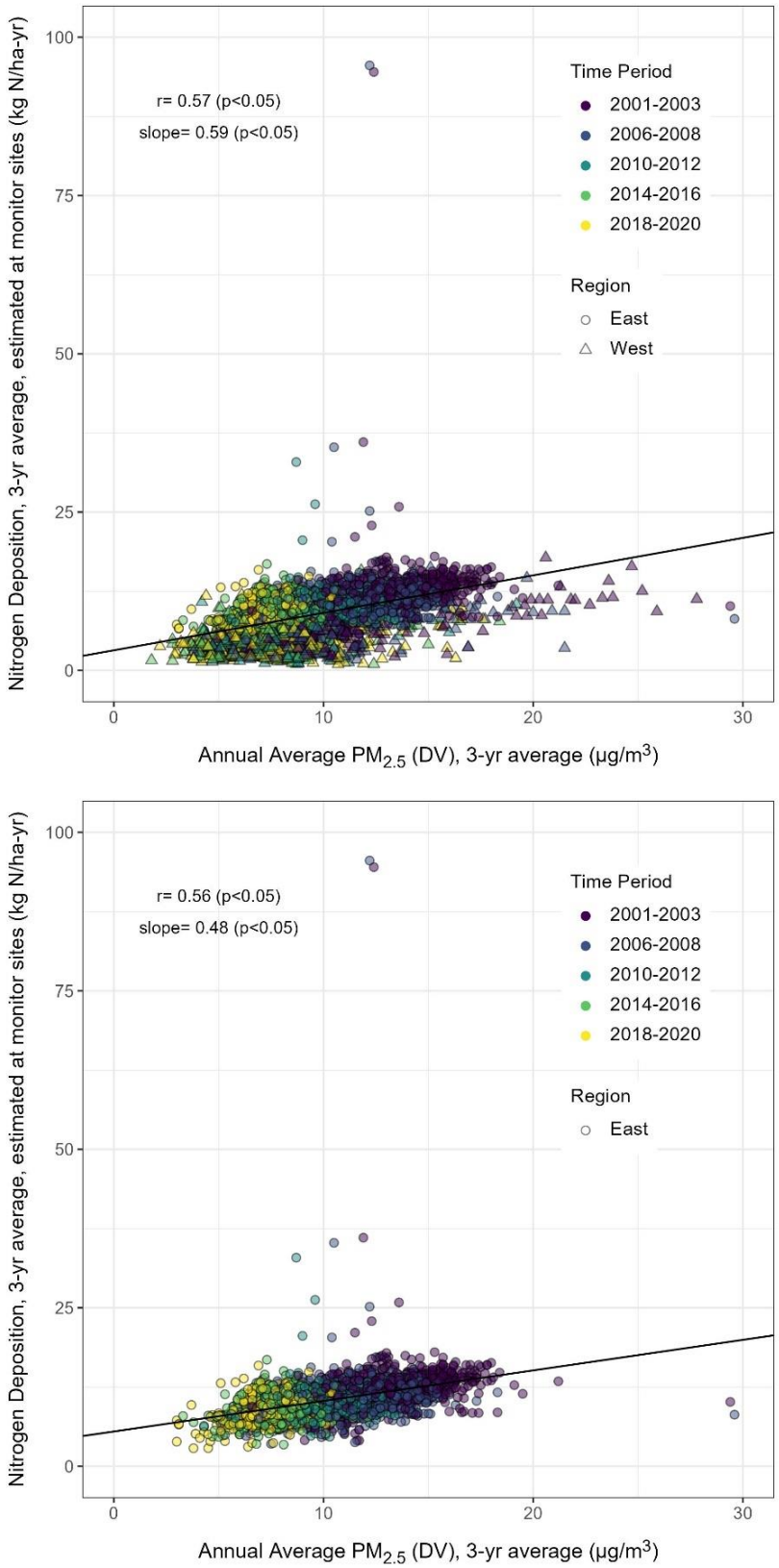

Figure 6-39. N deposition (TDep) and annual average $PM_{2.5}$ concentration (averaged over three years) at SLAMS across the CONUS (upper), and in East (lower).

Table 6-7. Correlation coefficients for TDep-estimated N deposition and annual average $PM_{2.5}$ concentrations (averaged across three years) at SLAMS in the CONUS.

Nitrogen Deposition and $PM_{2.5}$	SLAMS				
Annual DV-All Ecoregions	Correlation Coefficient (r) = 0.57*	*Annual DV-East Ecoregions*	r = 0.56*	*Annual DV-West Ecoregions*	r = 0.45*
Year	r	Year	r	Year	r
2001 - 2003	0.61*	2001 - 2003	0.46*	2001 - 2003	0.63*
2006 - 2008	0.47*	2006 - 2008	0.24*	2006 - 2008	0.50*
2010 - 2012	0.46*	2010 - 2012	0.24*	2010 - 2012	0.40*
2014 - 2016	0.38*	2014 - 2016	0.24*	2014 - 2016	0.24*
2018 - 2020	0.20*	2018 - 2020	0.33*	2018 - 2020	0.39*
*p<0.05					
Correlations are Spearman's Rank Correlations.					

In summary, the analyses described here expand on the Class 1 area analyses in section 6.2.2 above to consider deposition-concentration relationships at SLAMS regulatory monitors across the U.S., which are generally closer to sources than are the Class I monitors. At SLAMS locations, S deposition is strongly correlated with SO_2 concentrations, particularly in eastern sites and during the earliest period (2001-2003). This association is weaker in later periods and at western sites. Overall, the correlations are weaker for N deposition with NO_2 concentrations than those for S deposition and SO_2. Additionally, in contrast to what is seen for S and SO_2, correlations between N deposition and NO_2 concentrations are strongest at sites in the West. Nitrogen deposition and $PM_{2.5}$ concentrations have similar, low to moderate, correlation coefficients at sites in the East and West, with much weaker correlations in earlier time periods.

6.2.4 National-scale Sites of Influence Analyses

One limitation of the collocated analyses (Class 1 areas and at SLAMS monitors; "local" concentrations vs. "local" deposition) presented above is their inability to account for the role of upwind emissions, transport and chemical transformation in deposition. This section presents the results from a trajectory-based methodology that first identifies "sites of influence" that have the potential to contribute to deposition in a downwind location. Then, as a second step, considers the relationships between "upwind" air quality concentrations and "downwind" deposition in impacted ecoregions.

6.2.4.1 Methodology

We used the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) air parcel trajectory model to examine the potential transport of pollutants from source to receptor (see Appendix 6A for more detailed information). We generated forward trajectories from all

NO_2, SO_2, and $PM_{2.5}$ ambient air monitor locations with valid air quality data (i.e., from the SLAMS network described in Chapter 2). This was done to estimate how pollution observed at certain locations (referred to here as "sites of influence") could potentially be transported to downwind ecoregions. In a source-receptor framework, this analysis is considering the monitored values to be the "source" and downwind ecoregions as the "receptor." By identifying which air quality monitors are potentially representative of the air quality that contributes to deposition in a particular ecoregion, one can potentially better understand the relationship between upwind ambient air concentrations and downwind deposition rates.

After identifying the upwind geographic areas from which emissions potentially contribute to N and S deposition in each ecoregion,[10] we aggregated air quality concentrations within each ecoregion's set of sites of influence to estimate a ***weighted-average*** air quality metric, where the value of each site was weighted by how often the forward HYSPLIT trajectory crossed into the ecoregion (i.e., sites with more frequent trajectory intersections with the ecoregion are weighted higher). In addition to the weighted-average metric, we also extracted the ***area-wide maximum*** monitored concentration across the area contributing to deposition in each ecoregion. Both the weighted-averages and area-wide maximum air quality metrics were estimated for each ecoregion and for three separate pollutants: NO_2, SO_2, and $PM_{2.5}$.[11] For SO_2, we estimated two sets of metrics, one based on an annual average and one based on the 2nd high 3-hour max within the year. For NO_2 and $PM_{2.5}$, the data are based on annual average concentrations. These data are intended to provide a perspective of air quality levels in the upwind regions that potentially contribute to downwind deposition levels. For ease of reference, we have established the term Ecoregion Air Quality Metric, or EAQM, as shorthand for these metrics. The two types of metrics are referred to as EAQM-max and EAQM-weighted. All EAQM estimates were calculated for five 3-year periods: 2001-2003, 2006-2008, 2010-2012, 2014-2016, and 2018-2020 (i.e., further averaging the annual data).

To better understand the differences between these two types of air quality metrics, consider the following simplified hypothetical example for annual SO_2. The trajectory analysis suggests that there are four upwind monitoring sites where emissions contributing to the concentrations at those locations could also be contributing to S or N deposition in a specific downwind ecoregion. Other sites can also impact the downwind ecoregion but they do so less frequently (i.e., below some identified threshold) and therefore do not get included in the EAQM calculation.

[10] As in Chapter 5 above, we focused on level III ecoregions.

[11] We focused on the metric for the annual $PM_{2.5}$ standard because this averaging timescale is more relevant to assessing accumulated deposition than a standard with a form set to reduce peak concentrations (i.e., $PM_{2.5}$ 24-hour standard with its 98th percentile form).

- Site A – contributes 2% of the total ecoregion "hits;" 3-year average annual SO_2 = 10 ppb
- Site B – contributes 1% of the total ecoregion "hits;" 3-year average annual SO_2 = 8 ppb
- Site C – contributes 0.5% of the total ecoregion "hits;" 3-year average annual SO_2 = 12 ppb
- Site D – contributes 0.5% of the total ecoregion "hits;" 3-year average annual SO_2 = 10 ppb.

The EAQM-weighted metric for SO_2 for this ecoregion-year would be: [(2*10) + (1*8) + (0.5*12) + (0.5*10)] / [2 + 1 + 0.5 + 0.5] = 9.75 ppb. The EAQM-max metric in this example would be 12 ppb (from Site C). The EAQM-max metric offers insight into the highest design value associated with a particular deposition level, while the EAQM-weighted metric is useful in assessing how well upwind air quality is correlated with estimated S and N deposition. Used together, the assessment of these two metrics is intended to help further inform conclusions regarding the association between upwind regional air quality concentrations and downwind S and N deposition.[12]

6.2.4.2 Results

Starting with SO_2, we observe a strong positive correlation between SO_2 EAQM-weighted values across upwind sites of influence and S deposition in impacted ecoregions across the eastern U.S. As shown in Figure 6-40, higher S deposition values are associated with higher weighted-average SO_2 at upwind sites of influence ($r = 0.85$, slope = 2.22, $p<0.05$). This association holds across all five time periods, although there is more scatter in the 2018-2020 period than the others. As expected, EAQM-weighted annual SO_2 (averaged over 3 years) has decreased with time as have the S deposition amounts across the eastern U.S. The figure reaffirms the decreasing trends in ambient air SO_2 concentrations and S deposition discussed elsewhere in the PA. Prior to the 2010-2012 period, it was not uncommon for ecoregions to experience median S deposition exceeding 5 kg/ha-yr. However, since the 2014-2016 period, no regions have experienced median S deposition above 5 kg/ha-yr (Figure 6-40). Turning attention to the western U.S. (Figure 6A-63), the data suggest that the relationship between upwind SO_2 and downwind S deposition is less certain ($r = 0.19$, slope = 0.14, $p<0.05$). Annual SO_2 EAQM levels have decreased across the periods, but the S deposition in western ecoregions has been

[12] In these analyses S and N deposition are ecoregion medians derived from grid-cell estimates based on TDEP version: v2020.02, downloaded on September 7, 2022. These estimates are also presented in Figures 6-53 and 6-55.

relatively low (i.e., less than 2.5 kg/ha-yr) and exhibits smaller changes than what was observed in the EAQM-based air quality concentrations.

The EAQM-weighted data tend to be slightly better correlated with deposition than the EAQM-max data (Table 6-8). Figure 6-41 compares EAQM-max annual SO_2 values across the identified sites of influence against the downwind ecoregion S deposition for the eastern U.S.[13] There is some suggestion across the 20 years of data of a relationship between the concentration and deposition terms ($r = 0.65$, slope = 0.95, $p<0.05$), but it is largely driven by the older time periods when S deposition was higher. Figure 6-41 shows that there is no significant relationship between EAQM-max annual SO_2 at upwind sites of influence and S deposition in the western U.S. However, it should be noted that S deposition is relatively low in the western U.S. ecoregions.

[13] There are several outlier points in this comparison where the EAQM-max annual average SO_2 value exceeds 20 ppb in the 2018-2020 period. These points have been removed from this plot. These data are driven by a monitor in southeastern MO where annual average SO_2 has exceeded 20 ppb in recent years. Any downwind ecoregion that is linked to this upwind monitor will have an EAQM-max with this value. A preliminary analysis suggests that these observed SO_2 data are due to a new source that was not modeled in the CMAQ simulation that informed the TDep estimates of deposition. As there is no deposition monitor in the immediate vicinity of the source it is unlikely that the TDep estimates are capturing the impacts of this source. For that reason, the EPA concluded it was appropriate not to consider these data in our evaluation of the concentration-deposition relationship.

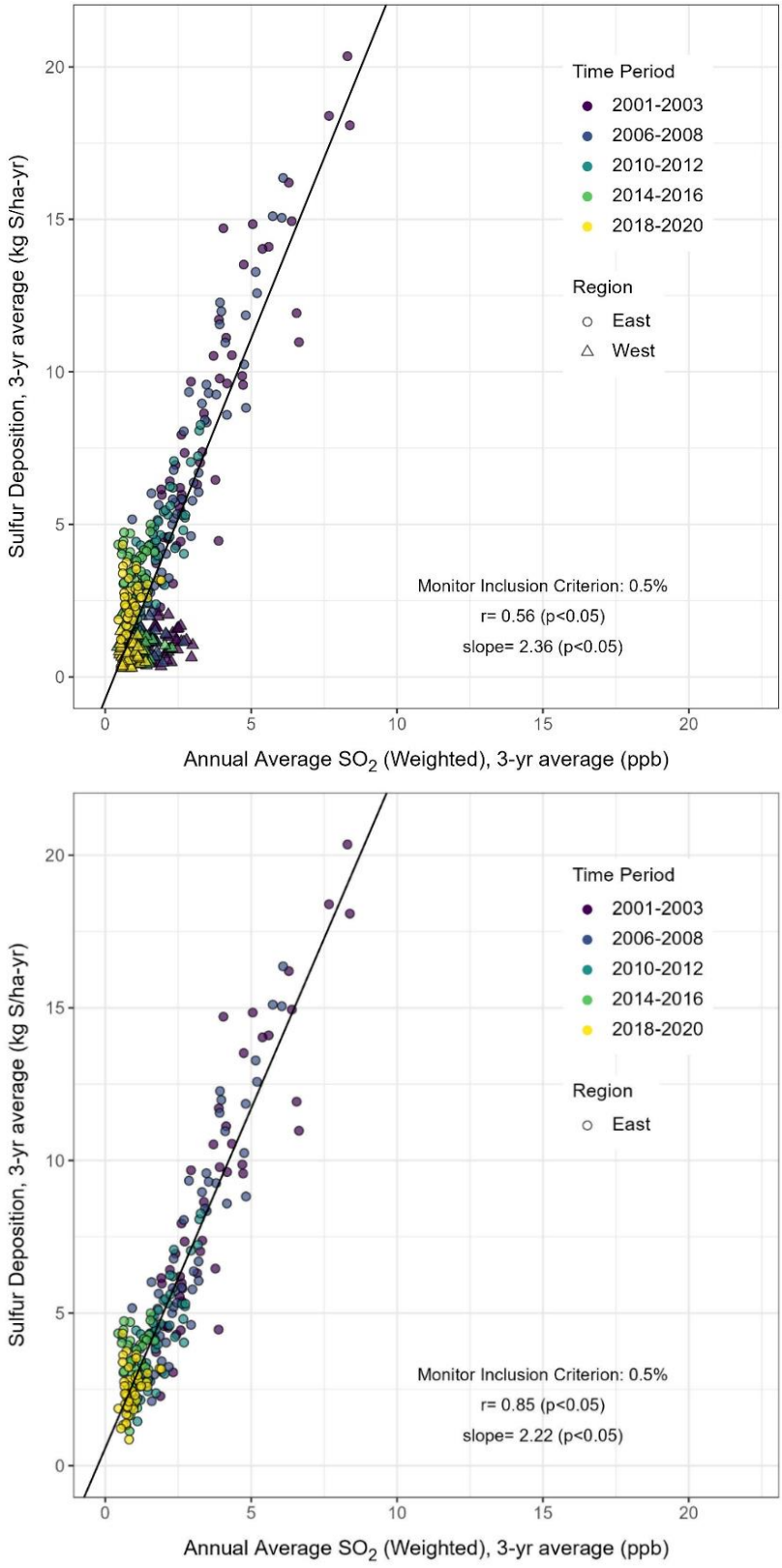

Figure 6-40. TDep-estimated median S deposition in all ecoregions (upper) and eastern ecoregions (lower) *versus* upwind annual SO_2 EAQM-weighted values.

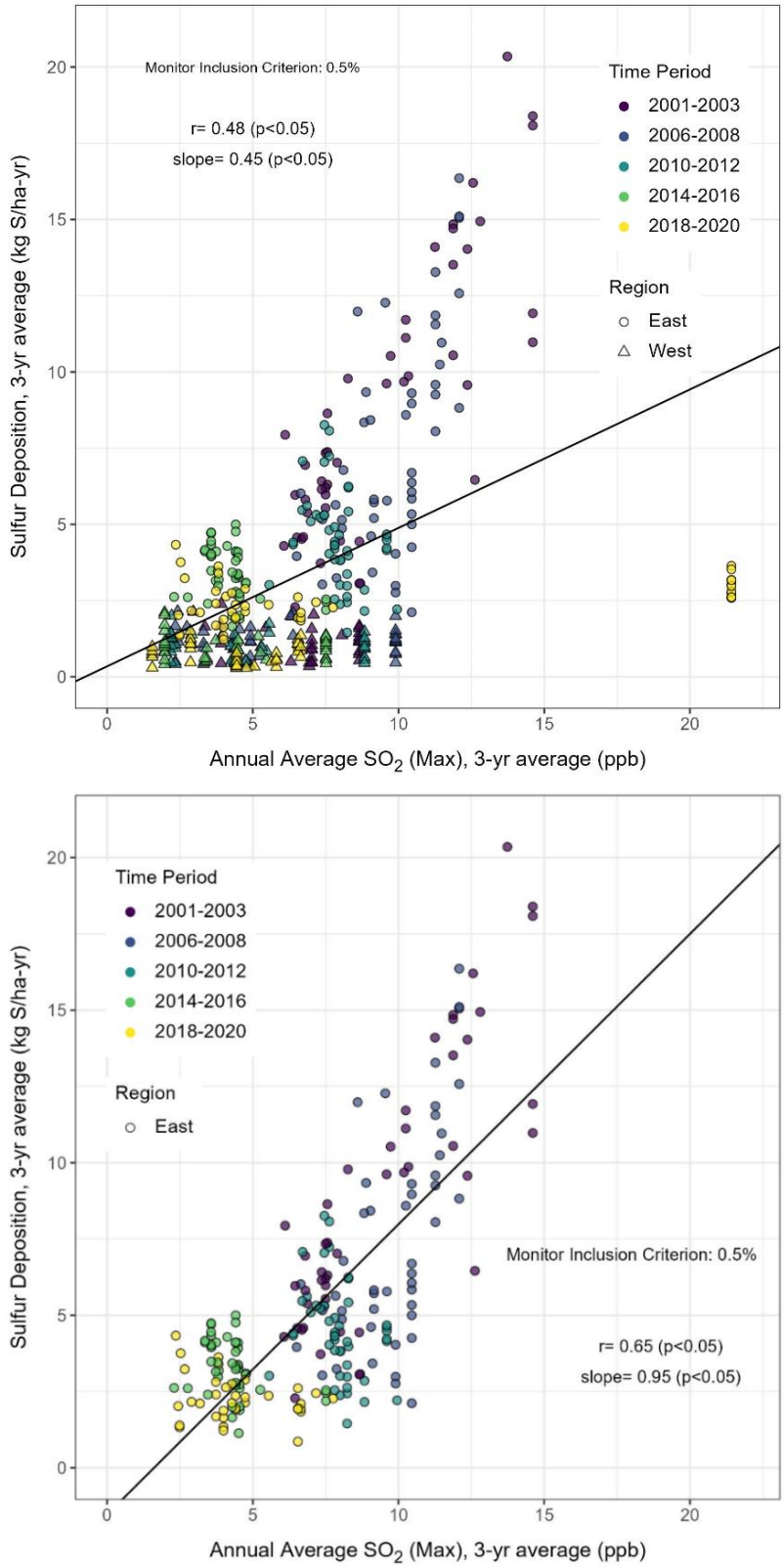

Figure 6-41. TDep-estimated median S deposition in all ecoregions (upper) and eastern ecoregions (lower) *versus* upwind annual SO_2 EAQM-max values.

Table 6-8. Correlation coefficients of TDep-estimated S deposition and annual SO_2 EAQMs by time period and region.

Sulfur Deposition and SO_2					
Annual Max-All Ecoregions- Monitor Inclusion Criteria: 0.5%	Correlation Coefficient (r) = 0.49*	*Annual Max-East Ecoregions- Monitor Inclusion Criteria: 0.5%*	r = 0.65*	*Annual Max-West Ecoregions- Monitor Inclusion Criteria: 0.5%*	r = 0.04
Year	r	Year	r	Year	r
2001 – 2003	0.62*	2001 – 2003	0.78*	2001 – 2003	-0.11
2006 – 2008	0.69*	2006 – 2008	0.59*	2006 – 2008	0.12
2010 – 2012	0.28*	2010 – 2012	-0.43*	2010 – 2012	-0.07
2014 – 2016	-0.05	2014 – 2016	-0.44*	2014 – 2016	-0.06
2018 – 2020	0.10	2018 – 2020	-0.13	2018 – 2020	0.03
Weighted Annual Average-All Ecoregions- Monitor Inclusion Criteria: 0.5%	r = 0.56*	*Weighted Annual Average-East Ecoregions- Monitor Inclusion Criteria: 0.5%*	r = 0.85*	*Weighted Annual Average-West Ecoregions- Monitor Inclusion Criteria: 0.5%*	r = 0.19*
Year	r	Year	r	Year	r
2001 – 2003	0.77*	2001 – 2003	0.89*	2001 – 2003	0.04
2006 – 2008	0.81*	2006 – 2008	0.9*	2006 – 2008	-0.07
2010 – 2012	0.71*	2010 – 2012	0.75*	2010 – 2012	-0.12
2014 – 2016	0.16	2014 – 2016	0.19	2014 – 2016	-0.14
2018 – 2020	0.22*	2018 – 2020	0.30*	2018 – 2020	0.04
*p< 0.05					

Considering the current secondary SO_2 NAAQS has an averaging time of 3 hours and a level of 0.5 ppm (500 ppb) that is not to be exceeded more than once per year, we next evaluate the concentration-deposition relationship for the 2nd highest 3-hour SO_2 EAQM values (weighted and max, again averaged over 3 years). Figure 6-42 suggests that there is strong association between S deposition and the weighted 3-hour EAQM ($r = 0.83$, slope = 0.16, $p<0.05$) across eastern U.S. ecoregions where higher values of downwind S deposition are associated with higher values of the weighted EAQM and roughly equivalent to the strong association reported for the annual SO_2 (Figure 6-40). However, as shown in Figure 6-43, there is a weaker association ($r = 0.42$, slope = 0.02, $p<0.05$) between EAQM-max and downwind deposition across eastern U.S. ecoregions for the 3-hour form of the standard. This is not a surprising result given that deposition is accumulated over several years, with pollution contributed by multiple locations, that may not be captured by simply looking at a short-term metric (2nd highest, 3-hour). This suggests that any revised SO_2 standard designed to protect against deposition-related effects would benefit from a longer averaging time. There is little significant association between the EAQM value and S deposition in the western U.S. for the current secondary SO_2 standard (Table 6-9).

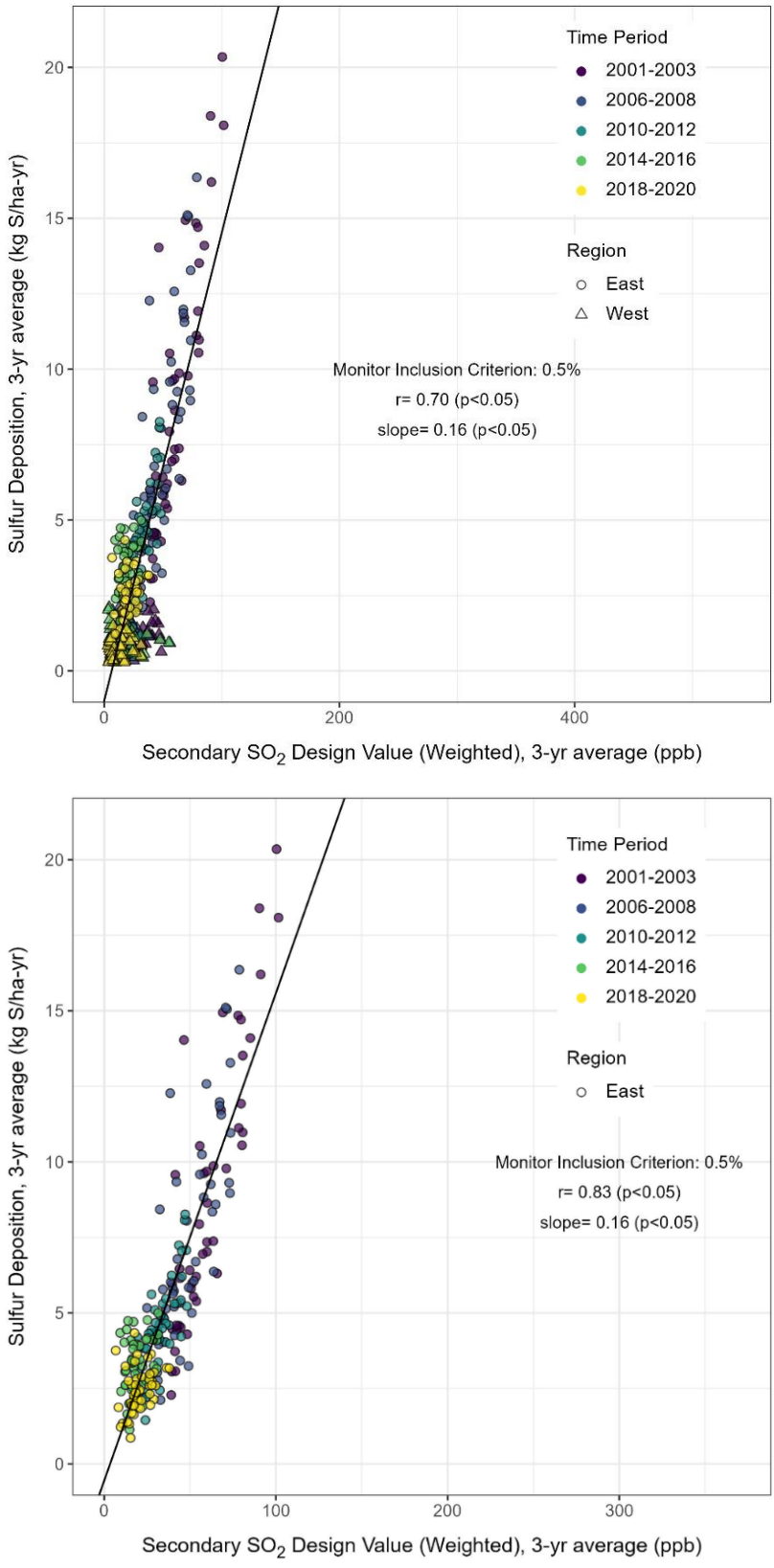

Figure 6-42. TDep-estimated median S deposition in all ecoregions (upper) and eastern ecoregions (lower) *versus* upwind 3-hour SO_2 EAQM-weighted values.

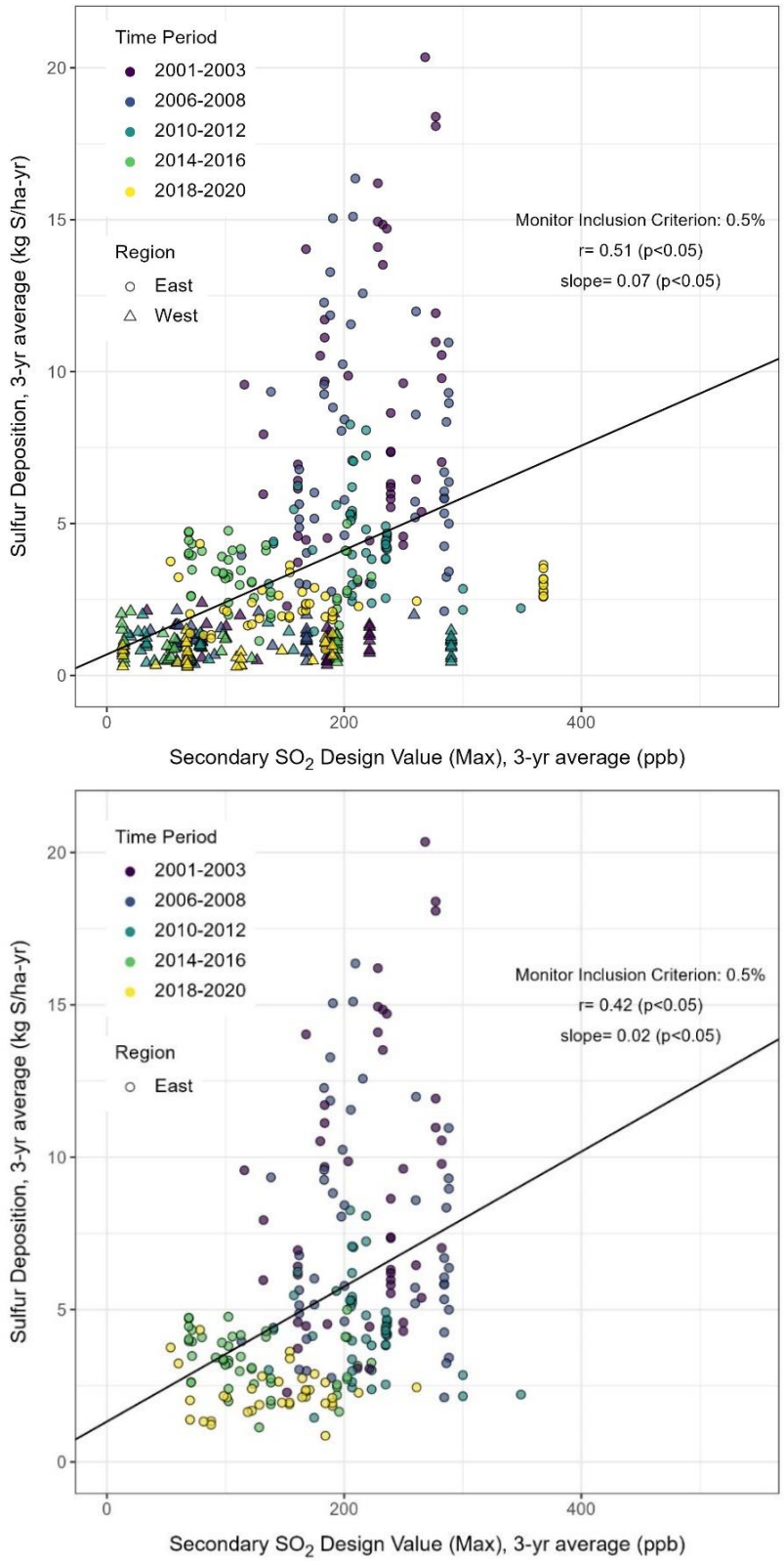

Figure 6-43. TDep-estimated median S deposition in all ecoregions (upper) and eastern ecoregions (lower) *versus* upwind 3-hour SO_2 EAQM-max values.

Table 6-9. Correlation coefficients of TDep-estimated ecoregion median S deposition and 3-hr SO_2 EAQM values at upwind site of influence by time period and region.

Sulfur Deposition and SO_2 (3-hour Standard)					
3-hr Max-All Ecoregions- Monitor Inclusion Criteria: 0.5%	Correlation Coefficient (r) = 0.51*	*3-hr Max-East Ecoregions- Monitor Inclusion Criteria: 0.5%*	r = 0.52*	*3-hr Max-West Ecoregions- Monitor Inclusion Criteria: 0.5%*	r = 0.07
Year	r	Year	r	Year	r
2001 - 2003	0.49*	2001 - 2003	0.49*	2001 - 2003	-0.06
2006 - 2008	0.69*	2006 - 2008	0.69*	2006 - 2008	0.18
2010 - 2012	0.25*	2010 - 2012	0.25*	2010 - 2012	-0.09
2014 - 2016	0.23*	2014 - 2016	0.23*	2014 - 2016	-0.09
2018 - 2020	0.54*	2018 - 2020	0.40*	2018 - 2020	0.10
3-hr Weighted Average-All Ecoregions- Monitor Inclusion Criteria: 0.5%	r = 0.70*	*3-hr Weighted Average-East Ecoregions- Monitor Inclusion Criteria: 0.5%*	r = 0.83*	*3-hr Weighted Average-West Ecoregions- Monitor Inclusion Criteria: 0.5%*	r = 0.20*
Year	r	Year	r	Year	r
2001 - 2003	0.86*	2001 - 2003	0.86*	2001 - 2003	0.15
2006 - 2008	0.89*	2006 - 2008	0.78*	2006 - 2008	0.31
2010 - 2012	0.77*	2010 - 2012	0.76*	2010 - 2012	0.06
2014 - 2016	0.38*	2014 - 2016	0.24	2014 - 2016	-0.16
2018 - 2020	0.54*	2018 - 2020	0.41*	2018 - 2020	0.15
*p< 0.05					

Similar analyses were completed assessing the relationship between upwind EAQM values in the form of the current secondary NO_2 standard (annual mean), averaged over three years, against downwind N deposition. Based on the results of section 6.2.1, one would expect it to be less likely that the upwind annual NO_2 EAQM values would be strongly correlated with N deposition due to the multiple pathways for N deposition and including ammonia-related sources. This is borne out as shown in Figure 6-44 for NO_2 EAQM-weighted values and ecoregion median N deposition in the East. The data indicate that the ecoregions with higher N deposition are associated with higher annual average NO_2 EAQM values in the older time periods. However, the correlation is much weaker than for annual average SO_2 EAQM for the eastern ecoregions ($r = 0.48$ versus $r = 0.85$ for the weighted metrics), and no association is observed between the upwind NO_2 concentrations and downwind N deposition in the more current periods or in the western ecoregions (Table 6-10 and Figure 6-44). The pattern of findings are generally similar for the NO_2 EAQM-max metric (Table 6-10 and Figure 6-45).

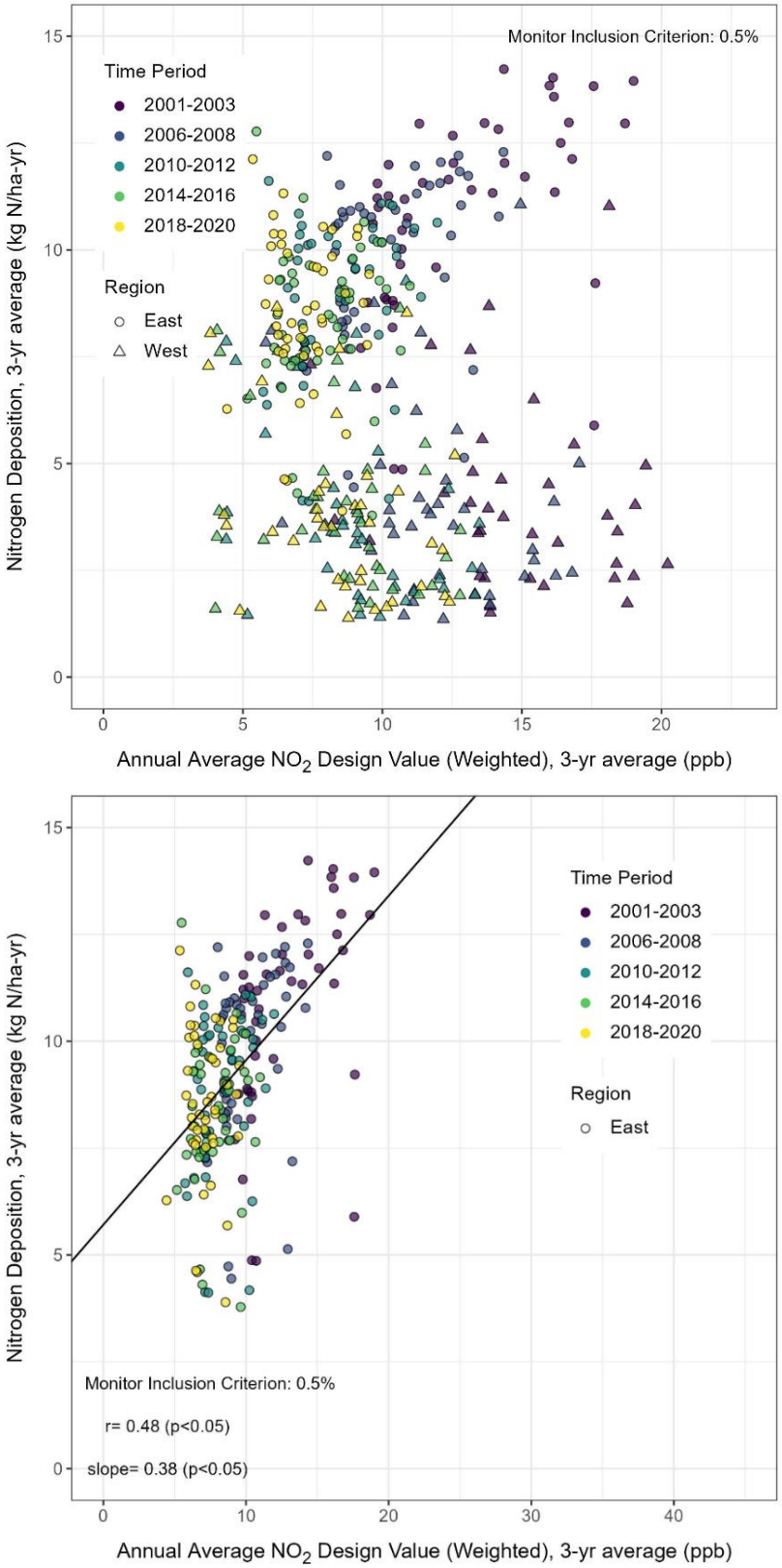

Figure 6-44. TDep-estimated median N deposition in all ecoregions (upper) and eastern ecoregions (lower) versus upwind annual NO_2 EAQM-weighted values.

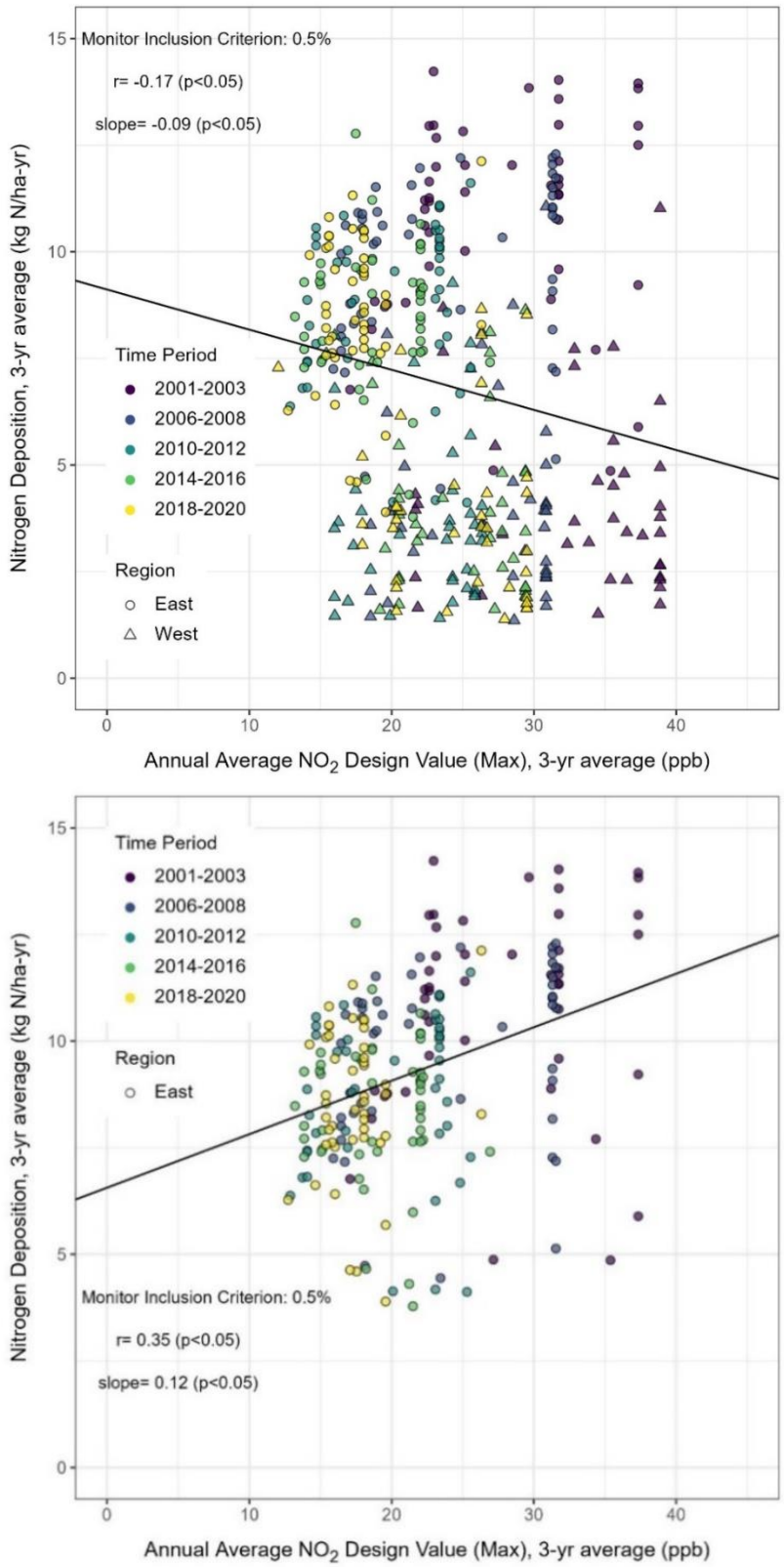

Figure 6-45. TDep-estimated median N deposition in all ecoregions (upper) and eastern ecoregions (lower) *versus* upwind annual NO_2 EAQM-max values.

Table 6-10. Correlation coefficients of ecoregion N deposition and upwind NO_2 annual EAQM values by time period and region.

Nitrogen Deposition and NO_2					
Annual Max-All Ecoregions- Monitor Inclusion Criteria: 0.5%	Correlation Coefficient (r) = -0.17*	*Annual Max-East Ecoregions- Monitor Inclusion Criteria: 0.5%*	r = 0.35*	*Annual Max-West Ecoregions- Monitor Inclusion Criteria: 0.5%*	r = -0.04
Year	r	Year	r	Year	r
2001 - 2003	-0.31*	2001 - 2003	0.24*	2001 - 2003	-0.12
2006 - 2008	0.05	2006 - 2008	0.35*	2006 - 2008	-0.05
2010 - 2012	-0.26*	2010 - 2012	0.15	2010 - 2012	0.02
2014 - 2016	-0.41*	2014 - 2016	0.03	2014 - 2016	-0.19
2018 - 2020	-0.58*	2018 - 2020	0.02	2018 - 2020	-0.25
Weighted Annual Average-All Ecoregions- Monitor Inclusion Criteria: 0.5%	r = -0.06	*Weighted Annual Average-East Ecoregions- Monitor Inclusion Criteria: 0.5%*	r = 0.48*	*Weighted Annual Average-West Ecoregions- Monitor Inclusion Criteria: 0.5%*	r = -0.17*
Year	r	Year	r	Year	r
2001 - 2003	-0.1	2001 - 2003	0.61*	2001 - 2003	-0.22
2006 - 2008	-0.21	2006 - 2008	0.39*	2006 - 2008	-0.23
2010 - 2012	-0.14	2010 - 2012	0.32*	2010 - 2012	-0.34*
2014 - 2016	-0.20	2014 - 2016	0.21	2014 - 2016	-0.28
2018 - 2020	-0.37*	2018 - 2020	-0.03	2018 - 2020	-0.26
*p< 0.05					
Correlations are Spearman's Rank Correlation					

Regarding upwind $PM_{2.5}$ EAQM values and N deposition in downwind ecoregions, as the composition of $PM_{2.5}$ over much of the U.S. is dominated by species that will not contribute to N deposition (e.g., organic carbon, elemental carbon), a strong relationship between $PM_{2.5}$ and N deposition is not expected, even though the ammonium component of $PM_{2.5}$ can contribute. Figures 6-46 and 6-47 show the results for the EAQM-weighted and EAQM-max, respectively. The $PM_{2.5}$ EAQM values have decreased over the past two decades and that the association between concentrations and N deposition is not significant over the western U.S. where deposition values are generally lower (Table 6-11). However, there is a moderate association between EAQM-weighted annual $PM_{2.5}$ and N deposition in eastern ecoregions (Figure 6-46; $r = 0.62$, slope = 0.63, $p<0.05$). As was the case for the S deposition and the SO_2 EAQMs, the EAQM-weighted values for annual $PM_{2.5}$ tend to be slightly better correlated with N deposition than the EAQM-max values ($r = 0.53$, slope = 0.44, $p<0.05$), but it is largely driven by the older time periods when N deposition was higher (Figure 6-47, Table 6-11).

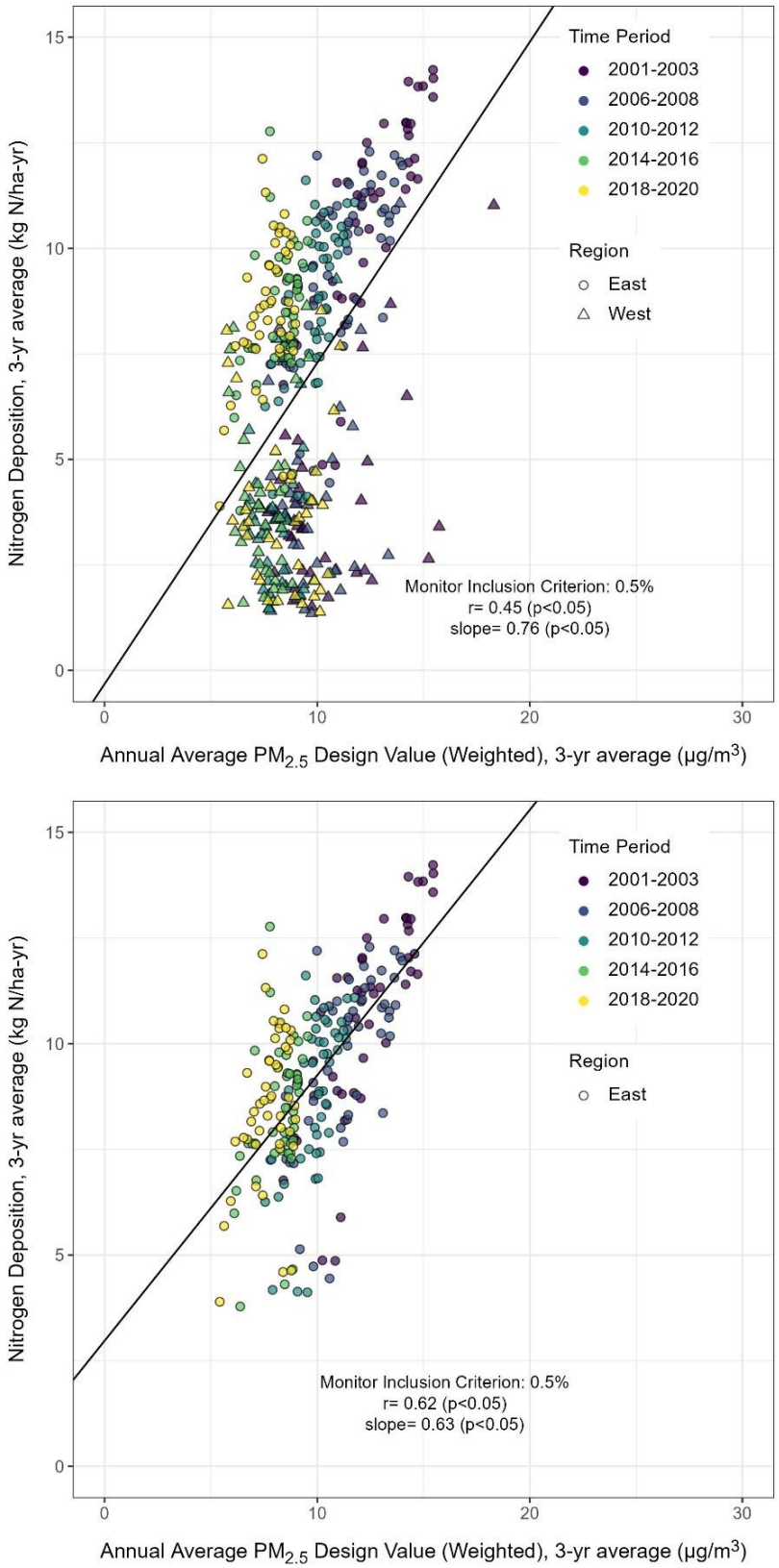

Figure 6-46. TDep-estimated median N deposition in all ecoregions (upper) and eastern ecoregions (lower) *versus* upwind annual $PM_{2.5}$ EAQM-weighted values.

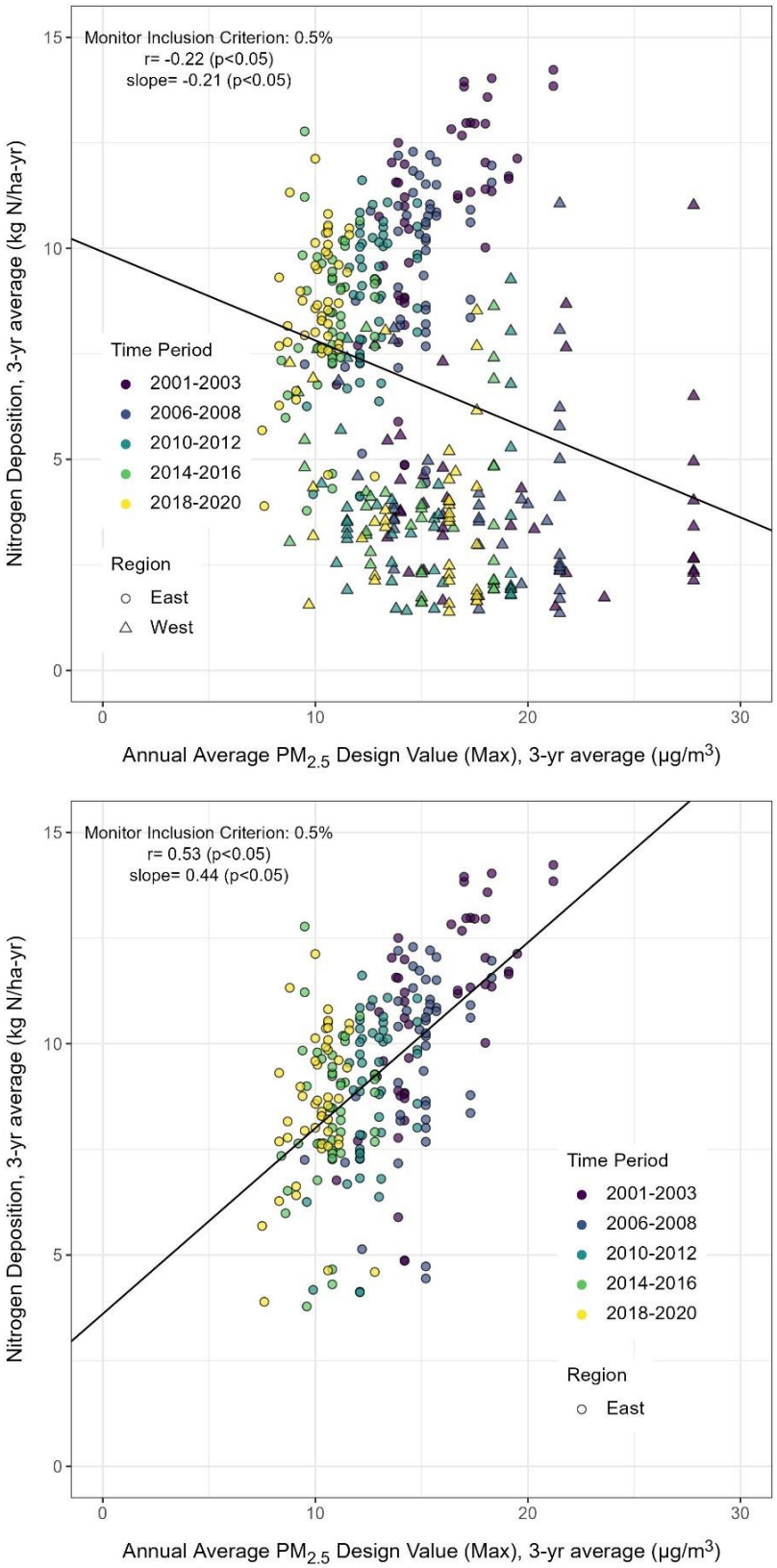

Figure 6-47. TDep-estimated median N deposition in all ecoregions (upper) and eastern ecoregions (lower) versus upwind annual $PM_{2.5}$ EAQM-max values.

Table 6-11. Correlation coefficients of $PM_{2.5}$ EAQM values with TDep-estimated median N deposition in downwind ecoregions.

Nitrogen Deposition and $PM_{2.5}$					
Annual Max-All Ecoregions- Monitor Inclusion Criteria: 0.5%	Correlation Coefficient (r) = -0.22*	*Annual Max-East Ecoregions- Monitor Inclusion Criteria: 0.5%*	r = 0.53*	*Annual Max-West Ecoregions- Monitor Inclusion Criteria: 0.5%*	r = -0.12
Year	r	Year	r	Year	r
2001 – 2003	-0.12	2001 – 2003	0.64*	2001 – 2003	-0.18
2006 – 2008	-0.30*	2006 – 2008	0.34*	2006 – 2008	-0.22
2010 – 2012	-0.14	2010 – 2012	0.46*	2010 – 2012	-0.13
2014 – 2016	-0.46*	2014 – 2016	0.27	2014 – 2016	-0.24
2018 – 2020	-0.49*	2018 – 2020	0.26	2018 – 2020	-0.07
Weighted Annual Average-All Ecoregions- Monitor Inclusion Criteria: 0.5%	r = 0.45*	*Weighted Annual Average-East Ecoregions- Monitor Inclusion Criteria: 0.5%*	r = 0.62*	*Weighted Annual Average-West Ecoregions- Monitor Inclusion Criteria: 0.5%*	r = 0.02
Year	r	Year	r	Year	r
2001 – 2003	0.65*	2001 – 2003	0.85*	2001 – 2003	-0.03
2006 – 2008	0.64*	2006 – 2008	0.67*	2006 – 2008	-0.14
2010 – 2012	0.75*	2010 – 2012	0.60*	2010 – 2012	0.09
2014 – 2016	0.45*	2014 – 2016	0.42*	2014 – 2016	-0.16
2018 – 2020	-0.09	2018 – 2020	0.27	2018 – 2020	-0.02
*p< 0.05					
Correlations are Spearman's Rank Correlations					

Regarding $PM_{2.5}$ EAQM values and median S deposition in downwind ecoregions, the correlations for both the max and weighted metrics for the full datasets (all time periods and both regions) were nearly identical with those for N deposition (Table 6-12). The correlations for both metrics with deposition in the eastern ecoregions were appreciably stronger for S than for N deposition (r = 0.83 and r = 0.90 *versus* r = 0.53 and r = 0.62). Little correlation was observed in the western ecoregions for either N or S deposition (Figures 6-48 and 6-49, Table 6-12).

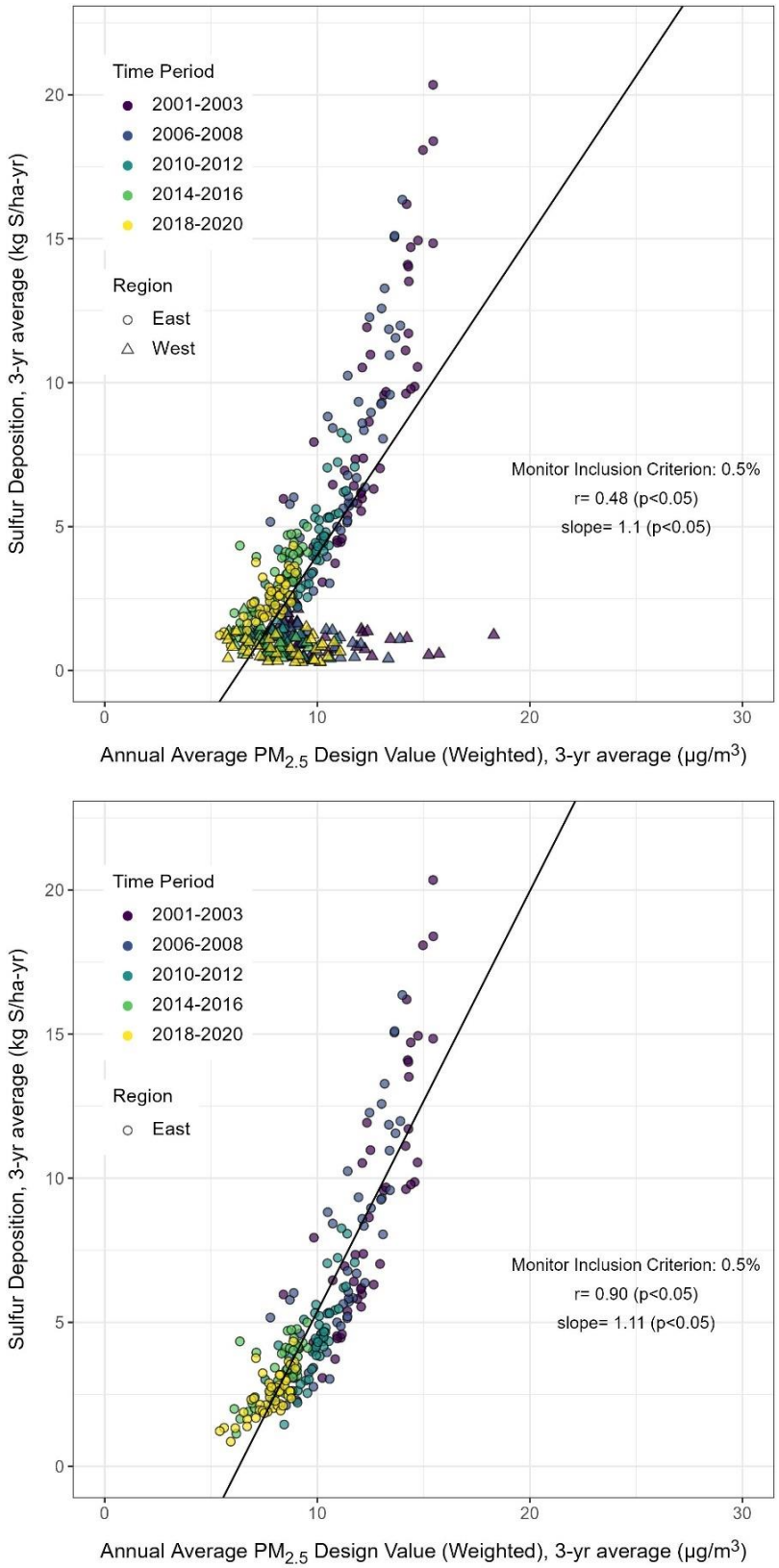

Figure 6-48. TDep-estimated median S deposition in all ecoregions (upper) and eastern ecoregions (lower) *versus* upwind annual $PM_{2.5}$ EAQM-weighted values.

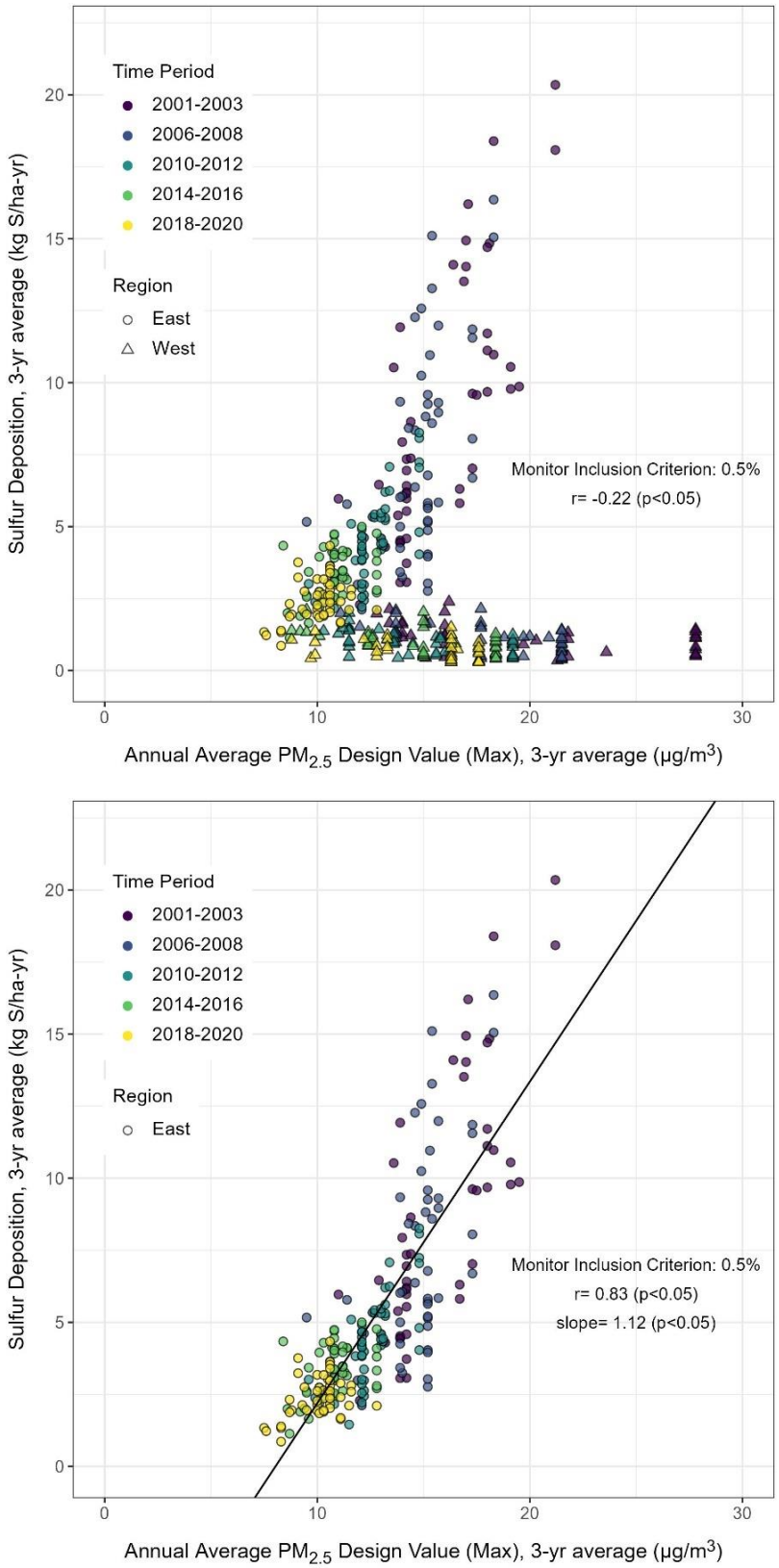

Figure 6-49. TDep-estimated median S deposition in all ecoregions (upper) and eastern ecoregions (lower) *versus* upwind annual $PM_{2.5}$ EAQM-max values.

Table 6-12. Correlation coefficients of TDep-estimated median S deposition and upwind $PM_{2.5}$ EAQM values.

Sulfur Deposition and $PM_{2.5}$					
Annual Max-All Ecoregions- Monitor Inclusion Criteria: 0.5%	Correlation Coefficient (r) = -0.22*	*Annual Max-East Ecoregions- Monitor Inclusion Criteria: 0.5%*	r = 0.83*	*Annual Max-West Ecoregions- Monitor Inclusion Criteria: 0.5%*	r = -0.33*
Year	r	Year	r	Year	r
2001 – 2003	-0.21	2001 – 2003	0.73*	2001 – 2003	-0.48*
2006 – 2008	-0.37*	2006 – 2008	0.53*	2006 – 2008	-0.61*
2010 – 2012	-0.22*	2010 – 2012	0.70*	2010 – 2012	-0.56*
2014 – 2016	-0.53*	2014 – 2016	0.43*	2014 – 2016	-0.53*
2018 – 2020	-0.54*	2018 – 2020	0.53*	2018 – 2020	-0.37*
Weighted Annual Average-All Ecoregions- Monitor Inclusion Criteria: 0.5%	r = 0.48*	*Weighted Annual Average-East Ecoregions- Monitor Inclusion Criteria: 0.5%*	r = 0.90*	*Weighted Annual Average-West Ecoregions- Monitor Inclusion Criteria: 0.5%*	r = -0.22
Year	r	Year	r	Year	r
2001 – 2003	0.55*	2001 – 2003	0.88*	2001 – 2003	-0.44*
2006 – 2008	0.56*	2006 – 2008	0.86*	2006 – 2008	-0.62*
2010 – 2012	0.70*	2010 – 2012	0.84*	2010 – 2012	-0.40*
2014 – 2016	0.43*	2014 – 2016	0.65*	2014 – 2016	-0.55*
2018 – 2020	-0.07	2018 – 2020	0.69*	2018 – 2020	-0.25
*p< 0.05					
Correlations are Spearman's Rank Correlations.					

6.2.4.3 Conclusions

For SO_2 downwind ecoregion S deposition, we examined both the 3-hour and annual average metrics. The results indicate that both metrics are correlated with S deposition, with the strongest correlations observed in the earliest time periods and for S deposition in the eastern ecoregions. As would be expected given its derivation, there is somewhat higher correlation for the EAQM-weighted metric. The figures for SO_2 also indicate median ecoregion S deposition to be above approximately 10 kg/ha-yr in the first two time periods assessed and generally approximately 5 kg/ha-yr in the most recent period. However, the SO_2 figures also indicate that there can be high measured SO_2 concentrations associated with low S deposition (i.e., < 5 kg S/ha-yr), in both the eastern and western U.S., and that there is generally more scatter in the data at lower deposition values. Both of these observations could be driven by uncertainties in the TDep calculations and/or uncertainties in our EAQM assessment methodology.

For NO_2, the correlations between the annual NO_2 EAQM values and N deposition are not nearly as strong as they are between metrics for SO_2 concentrations and S deposition. This could be because oxidized nitrogen only contributes to part of the total N deposition estimate (Figure 6-18), and as discussed in sections 2.5.3 and 6.2.1, the contribution of reduced nitrogen

to total N deposition has grown over the last few decades (*e.g.*, Li et al., 2016). The NO_2 EAQM analysis, like others in this section, suggest that, based on patterns of and trends in N species in recent air quality, NO_2 may not be a good indicator for total N deposition and consideration of related effects.

For $PM_{2.5}$, the results show some limited correlation between the measurements of annual average $PM_{2.5}$ and estimates of N deposition, particularly in the east and in the earlier time periods. The findings of association may relate to measurements at $PM_{2.5}$ monitors where both oxidized and reduced forms of N (i.e., NO_3 and NH_4^+) are copious. However, $PM_{2.5}$ mass is also comprised of many components unrelated to N or N deposition. Further, the scatterplot of annual $PM_{2.5}$ EAQM-max and N deposition shows that the range of ecoregion deposition values associated with individual annual average $PM_{2.5}$ concentrations is broad. For example, for annual average $PM_{2.5}$ concentrations (averaged over three years) from approximately 15 to 10 $\mu g/m^3$, downwind N deposition ranges from less than 5 to somewhat above 12 kg/ha-yr (Figure 6-47). The analyses for $PM_{2.5}$ and ecoregion S deposition indicate a somewhat tighter relationship. Except for a few datapoints prior to 2010, S deposition was below 10 kg/ha-yr when upwind annual average $PM_{2.5}$ concentrations (averaged over three years) were at or below 15 $\mu g/m^3$ (Figure 6-49).

Regarding the EAQM approach, we take note of certain assumptions and limitations that are discussed in detail in Appendix 6A. We emphasize here that the EAQM-based relationships between concentrations and deposition in downwind ecoregions are not intended to represent predictive associations that can determine what the downwind deposition will be as a function of upwind air quality. In fact, the scatter in the data (e.g., same concentrations can lead to different deposition levels, same deposition levels can result from different upwind pollutant concentrations) argues just the opposite. The findings of the EAQM analysis suggest that among the three criteria pollutants, SO_X will have the closest relationships between concentrations and eventual S deposition, particularly for a concentration metric with a longer-term averaging time, such as a 3-year average of annual average hourly data.

6.3 LIMITATIONS AND UNCERTAINTIES

A summary of key limitations and associated uncertainties of the data and analyses described in this chapter is provided below. This summary is based on the characterization of uncertainties presented in section 6.3.1 and is followed by sensitivity analyses in section 6.3.2 that provide additional support to the characterization of uncertainty associated with the trajectory-based analyses discussed in section 6.2.4 above. The mainly qualitative approach to uncertainty characterization that is presented in section 6.3.1 and used for air quality, exposure

and risk assessments performed in other NAAQS reviews,[14] is also informed by quantitative sensitivity analyses, as described by WHO (2008).

The linkage between air concentration and deposition can vary based on site-specific conditions, including the chemical form of nitrogen and sulfur, frequency of precipitation, and micrometeorological factors relevant to the dry deposition velocity. The analyses above attempt to provide insight into these relationships and variability for multiple measured air quality metrics. As with any assessment, there are uncertainties and limitations associated with the work, most of which are discussed above in the context of each the analyses. In this section, we summarize some of the overarching uncertainties and limitations.

In section 6.2.2, multiple forms of data were analyzed using co-located information in a subset of Class I areas. While there are uncertainties in each of the different sets of modeled and measured data analyzed, the fact that the assessment saw consistent results across these different forms of data reduces the concern with these potential data-related issues. The biggest limitation of the assessment in section 6.2.2 is the limited geographical coverage of the Class I areas that were included. Although these areas are in different parts of the country and were chosen based on the availability of co-located air quality (i.e., IMPROVE, CASTNET) and NADP/NTN monitors, most were located in the western U.S., where terrain, emissions and air quality chemistry can look different from other parts of the country. This analysis may neglect or underestimate the role of large ammonia emission sources in the Midwest and large nitrogen oxide emission sources in the eastern U.S. Additionally, these selected Class I areas include greater representation of the West (20 of the 27 sites are in the ecoregions designated West) than is the case for the locations that were quantitatively assessed in Chapter 5 for potential aquatic acidification effects (e.g., only 8 of the 25 ecoregions are in the West).

In section 6.2.4, an analysis using the HYSPLIT model was included to assess the linkage between TDep estimates of N and S deposition and measured air quality concentrations of NO_2, SO_2 and $PM_{2.5}$. There are uncertainties in the HYSPLIT application itself, including the use of one year of meteorological data to estimate multiple years of transport. Additionally, this analysis included judgments on the percentage of trajectory impacts warranting inclusion in an ecosystem's sites of influence. It is unclear how much and in what way these uncertainties and assumptions might impact the results. Although increasing the geographic scope of the sites of influence could lead to higher maximum values, there are also uncertainties in the TDep estimates, which are discussed in more detail in section 2.5. There is also uncertainty as to whether only SO_2, NO_2 and $PM_{2.5}$ concentrations at the monitor site influence the designated

[14] This approach to uncertainty characterization has been utilized in welfare and health REAs for reviews of the ozone, NO_2, SO_2, and carbon monoxide NAAQS (e.g., U.S. EPA 2014, 2018).

receiving ecoregion deposition. An additional uncertainty that should also be considered is the application of HYSPLIT to somewhat large areas of the country (i.e., ecoregions), which may have substantial spatial variability in deposition levels.

6.3.1 Characterization of Uncertainty

Briefly, with this approach, we have identified key aspects of the assessment approach that may contribute to uncertainty in the conclusions and provided the rationale for their inclusion (Table 6-13). Then, we characterized the *magnitude* and *direction* of the influence on the assessment for each of these identified sources of uncertainty. Consistent with the WHO (2008) guidance, we scaled the overall impact of the uncertainty by considering the degree of uncertainty as implied by the relationship between the source of uncertainty and interpretations drawn from the air quality analyses. A qualitative characterization of *low*, *moderate*, and *high* was assigned to the magnitude of influence and knowledge base uncertainty descriptors, using quantitative observations relating to understanding the uncertainty, where possible. Where the magnitude of uncertainty was rated *low*, it was judged that large changes within the source of uncertainty would have only a small effect on the assessment results (e.g., upwards to a factor of two). A designation of *medium* implies that a change within the source of uncertainty would likely have a moderate (or proportional) effect on the results (e.g., a factor of two or more). A characterization of *high* implies that a change in the source would have a large effect on results (e.g., an order of magnitude). We also included the direction of influence, whether the source of uncertainty was judged to potentially over-estimate ("*over*"), under-estimate ("*under*"), or have an *unknown* impact on the analyses designed to assess relationships between air quality concentrations and deposition (Table 6-13).

Table 6-13. Characterization of key uncertainties in analyses that relate air quality to deposition.

Sources of Uncertainty		Uncertainty Characterization			
		Influence of Uncertainty on Analyses		Knowledge-base Uncertainty	Comments
Category	Element	Direction	Magnitude		
Ambient Air Concentrations at IMPROVE Monitors	Ambient Air Concentration Measurements	Unknown	Low	High	IMPROVE monitoring of $PM_{2.5}$ uses a gravimetric assessment of aerosol mass collected on a Teflon filter, which may have biases outside of the sampling relative humidity range (30-40%). Sulfate and nitrate mass are calculated by assuming that either is fully neutralized by ammonia. The bias in this estimate would be affected by the actual composition of sulfate or nitrate (e.g., in organic vs. inorganic forms). A comparison of the IMPROVE gravimetric $PM_{2.5}$ and reconstructed mass methods suggests that they generally co-vary on a seasonal to annual basis (R^2 = 0.93-0.96; Malm et al., 2011), such that we do not anticipate that uncertainties in either measurement method will alter the conclusions drawn from our assessment of the correlation between deposition and IMPROVE concentration measurements.
	Air Quality System (AQS) Database Quality	Unknown	Unknown	High	See above
	Spatial Representation	Low	Medium	Medium	Overall, IMPROVE sites are in national parks and Federal Class 1 locations, which are generally remote and relatively pristine ecosystems. There is a higher density in the west versus eastern USA, and there is a dearth of monitors in the midwest. More analysis would be needed to assess the extent to which our collocated assessment using IMPROVE is extendable to areas with fewer collocated monitors and differing environmental conditions (e.g., urban). IMPROVE monitors at remote locations help reduce uncertainty in HYSPLIT estimated concentrations.
	Temporal Representation	Low	Low	Medium	IMPROVE collects samples on a 24-h basis every three days. There may be some uncertainty associated with non-continuous sampling.
Ambient Air Concentrations at CASTNET Monitors (part of NADP)	Air Concentration Measurements	Unknown	Low	High	The precision of CASTNET measured ammonium, nitric acid and nitrate are estimated as 3.0%, 5.5% and 7.8%, respectively (Sickles and Shadwick, 2002). The volatility of ammonium nitrate can contribute biases in nitrate (low bias) and nitric acid (high bias), while the total nitrate concentration (NO_3^- + HNO_3) is conserved (Lavery et al., 2009; Zhang et al., 2009). Although volatility-related bias in ammonium concentrations was not the focus of these studies, this bias should be lower where ammonium is generally associated with sulfate (Walker et al., 2019).
	Spatial Representation	Low	Medium	High	CASTNET monitors are located in remote or rural areas. CASTNET-measured concentrations may be representative of pollution levels that affect sensitive or pristine ecosystems. CASTNET follows the legacy of acid rain, so that most sites are located in the eastern USA. There is more uncertainty in the western and midwest USA due to a relative sparsity in measurements.
	Temporal Representation	Low	Low	Medium	CASTNET measures the total mass of HNO_3, SO_2 and PM on a weekly basis. This level of temporal resolution should be sufficient for inferring annual, cumulative impacts from ecosystem exposure.

Sources of Uncertainty		Uncertainty Characterization			
		Influence of Uncertainty on Analyses		Knowledge-base Uncertainty	Comments
Category	Element	Direction	Magnitude		
Wet Deposition Measurements (NADP)	Sample Collection Methods	Unknown	Low	High	Collocated collectors suggest that the median absolute error of NTN precipitation measurements of ammonium and nitrate is 11% and 5.0%, respectively (Wetherbee et al., 2005). Precipitation sampled ammonium is biased low by approximately 10% (Gilliland et al., 2002; Walker et al., 2012).
	Chemical Analysis	Unknown	Low	High	Since 2018, NADP chemical analysis has been conducted by the Wisconsin State Laboratory of Hygiene. Previous assessment of inter-lab measurements a significant difference in sulfate, although with a small median difference of 0.048 mg/L. There were not significant differences in the measurement of ammonium or nitrate (Wetherbee et al., 2010).
	Precipitation measurements	Unknown	Low	High	Differences across precipitation monitors varied by 4.1 to 8% at several NTN study sites between 2007-2009. The precision among precipitation measurements was found to be between 0.6 to 2.2%. Although these differences were statistically significant, the magnitude of biases was small enough to be considered negligible (Wetherbee et al., 2010).
	Spatial Representation	Low	Medium	High	Wet deposition is currently measured at approximately 250 sites in the NADP/NTN network. NTN monitors are mainly located away from urban areas and pollution sources.
	Temporal Representation	Low	Low	Medium	Wet deposition is currently measured on a weekly average by the NADP/NTN network.
TDep – Continuous estimate of deposition	Spatial interpolation to estimate 4km wet deposition	Unknown	Medium	Low	Estimates of wet deposition are interpolated using inverse distance weighting (NADP, https://nadp.slh.wisc.edu/networks/national-trends-network/). While there are other methods for spatial interpolation, to our knowledge potential differences among them have not been tested for wet deposition. Areas having a relatively lower density of monitor sites may have greater uncertainty than other areas.
	CMAQ estimates of gas phase NO_2, SO_2, and NH_3 and particulate SO_4^{2-}, NO_3^- and NH_4^+	Unknown	Medium	Medium	CMAQ air concentration biases may be substantial, varying by season and region. These biases will affect the interpolated estimates of deposition from TDep, with a larger extent of influence further from monitor locations.
	Bias correction with CASTNET measurements of gas phase SO_2 and NO_y, and particulate SO_4^{2-}, NO_3^- and NH_4^+	Unknown	Low	Low	There are several potential approaches for bias correction that have not been evaluated as part of our analysis. However, we anticipate that the selection of bias correction method will have a smaller influence than the measurement uncertainty.
	Effective dry deposition velocity estimates	Unknown	High	Low	Affected by meteorology, surface conditions (e.g., complex terrain), elevation and land cover. Although NH_3 fluxes are bi-directional, there is not yet a way to represent this feature more dynamically in TDep.

Sources of Uncertainty		Uncertainty Characterization			
		Influence of Uncertainty on Analyses		Knowledge-base Uncertainty	Comments
Category	Element	Direction	Magnitude		
	Dry deposition interpolation to estimate dry deposition at 4km grid	Unknown	Medium	Low	Additional measurements would be needed to evaluate TDep interpolation. The interpolation may obscure fine resolution variability distant from monitors.
Assignment of monitors to ecoregion zones of influence with HYSPLIT modeling	Use of a single year (2016) in HYSPLIT to be representative of long term meteorological patterns	Unknown	Medium	Low	HYSPLIT analyses require meteorological data to identify the trajectories of air parcel transport from a source to a receptor. A single year was chosen to keep the analyses manageable and for consistency with prior EPA trajectory analysis. 2016 was selected for consistency with prior EPA trajectory analyses and because it appears to represent typical meteorological conditions. However, the use of only one year of meteorological data adds uncertainty to the identification of potential upwind sites of influence as the true frequency of wind directions over the 20-year study period (2000-2020) may differ from what was determined based on 2016 alone.
	Resolution of HYSPLIT meteorological inputs	Low	Low	High	Sensitivity analyses compared how associations between pollutants and deposition differed when the trajectories were based off of 12-km data instead of 32-km data (see Appendix 6A). The results suggest the impact of meteorological input resolution is small.
	Duration of HYSPLIT trajectories (120 hours)	Low	Low	High	Sensitivity analyses compared how associations between pollutants and deposition differed when the trajectories that identified the upwind sites of influence were developed using 120-hour trajectories, as opposed to 48-hour trajectories (see Appendix 6A). The results suggest the impact of trajectory duration is small.
	Extent to which monitors represent areas with air quality of interest	Unknown	Medium	Low	The air quality data on which the EAQM calculations are based are from the SLAMS network. The presumption is that U.S. air quality monitoring networks for NO_2, SO_2, and $PM_{2.5}$ are robust enough to enable one to use these data to establish a meaningful representation of the air quality that may contribute to downwind deposition. It is beneficial that NO_2, SO_2, and $PM_{2.5}$ monitors are often located near sources or in highly-populated areas (e.g., source-oriented monitoring, near-road monitoring). However, there are some background-oriented sites included in this analysis which may influence conclusions. Additionally, there are likely some source of pollutants that eventually impact deposition which are not captured. This is most likely to impact the EAQM-max metric.
	Monitor inclusion criteria	Unknown	High	Moderate	Sensitivity analyses assessed how different choices about which upwind monitors should be considered as potential sites of influence (and therefore part of the EAQM calculation) impacted both the spatial extent of the upwind influence and the eventual assessments of the strength of relationships between pollutant metrics and deposition. The lower the threshold for inclusion resulted in a larger areal extent of sites of influence. This, in turn, affected the strength of several associations (as measured by R2 and slope).

Sources of Uncertainty		Uncertainty Characterization			
		Influence of Uncertainty on Analyses		Knowledge-base Uncertainty	Comments
Category	Element	Direction	Magnitude		
	Variation in ecoregion size and shape, as well as topographic, geologic and other features	Unknown	Medium	Moderate	Smaller ecoregions are more likely to have fewer sites of influence, as most of the trajectory hits come from monitors within the ecoregion itself, making it less likely that sites outside the ecoregion will reach the 0.5% criteria for fraction of total trajectory hits.

6.3.2 Sensitivity Analyses Related to Aspects of Trajectory-Based Assessment

As described in more detail in Appendix 6A, we conducted sensitivity testing on three aspects of the analytical methodology used to calculate EAQM values. Specifically, we examined two durations for the forward parcel trajectories (48 hours and 120 hours), two different meteorological input data sets (NARR-32 and NAM-12) with differing resolution, and three different monitor inclusion criteria ranging from 1% of total hits in an ecoregion to 0.1% of total hits in an ecoregion. Each of these methodological changes, when moving from the original analysis to the final analysis, had the effect of allowing more distant upwind sites to be included in the EAQM calculations of air quality across potential sites of influence.

Figure 6-50 shows the association between annual SO_2 EAQM values and S deposition across the 84 ecoregions and 5 time periods, based on a 48-hour duration for the trajectory analysis, the NARR-32 inputs, and a monitor inclusion criterion of 1%. Figure 6-51 shows the association between annual SO_2 EAQM values and S deposition across the 84 ecoregions and 5 time periods, based on 120-hour duration for the trajectory analysis, the NAM-12 input data, and a minimum hit rate of 0.5% for monitoring site inclusion criterion. In both analyses, similar themes emerge. It is clear from both figures that the SO_2 EAQM and TDep-estimated S deposition association is strongest in the eastern U.S., and essentially non-existent in western U.S. locations. In both cases, we can conclude that the relationship between upwind air quality and downwind deposition was stronger in the earlier periods than it is in the most recent, 2018-2020, period. It is also noted that the R-value increases slightly with the inclusion of more distant sites, from 0.45 to 0.56. Figures 6A-23 and 6A-24 in Appendix 6A present the same types of plots for ecoregion S deposition and SO_2 EAQM-max but for data limited to the eastern ecoregions limit the comparisons to sites in the eastern U.S. and the associations are equally strong in both iterations of the methodology ($r = 0.85$, slope ~ 2.2, $p<0.05$). In sum, we concluded that the overall strength of association between upwind air quality and downwind deposition are not strongly affected by the choice of trajectory length, meteorological inputs, or monitor inclusion criteria.

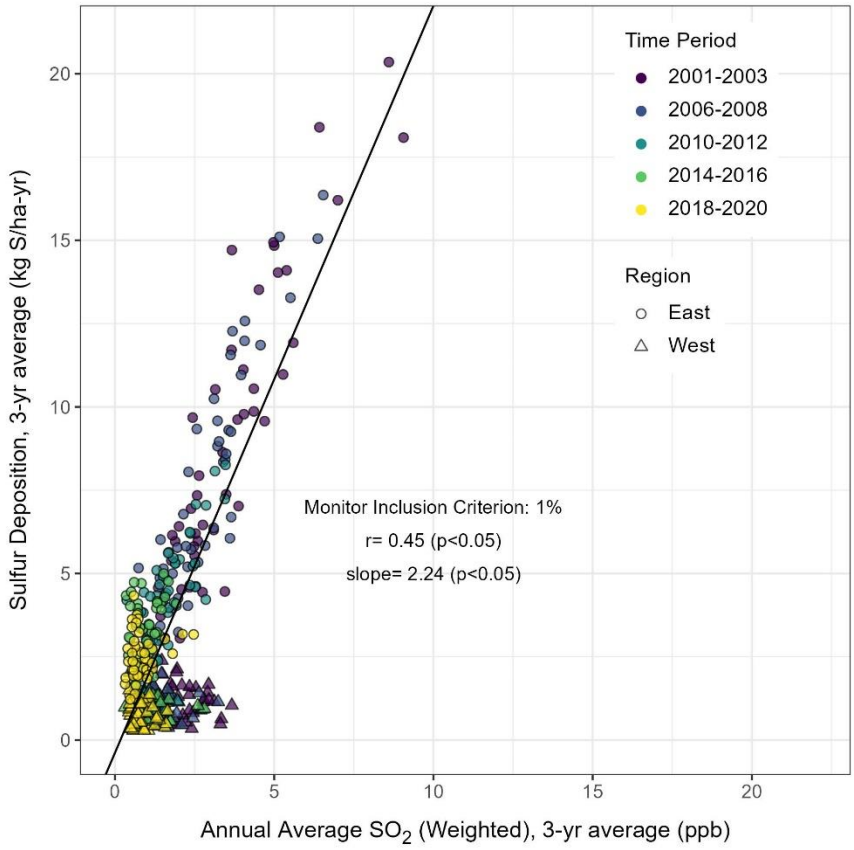

Figure 6-50. Plot of annual SO_2 EAQM values against TDep total S deposition across 84 ecoregions. The individual pairs are color-coded by 3-year periods and the symbols differentiate between sites in the eastern U.S. and western U.S. This figure is based on EAQM data using 48-hour trajectories, the NARR-32 meteorological data, and a monitor inclusion criterion of 1%.

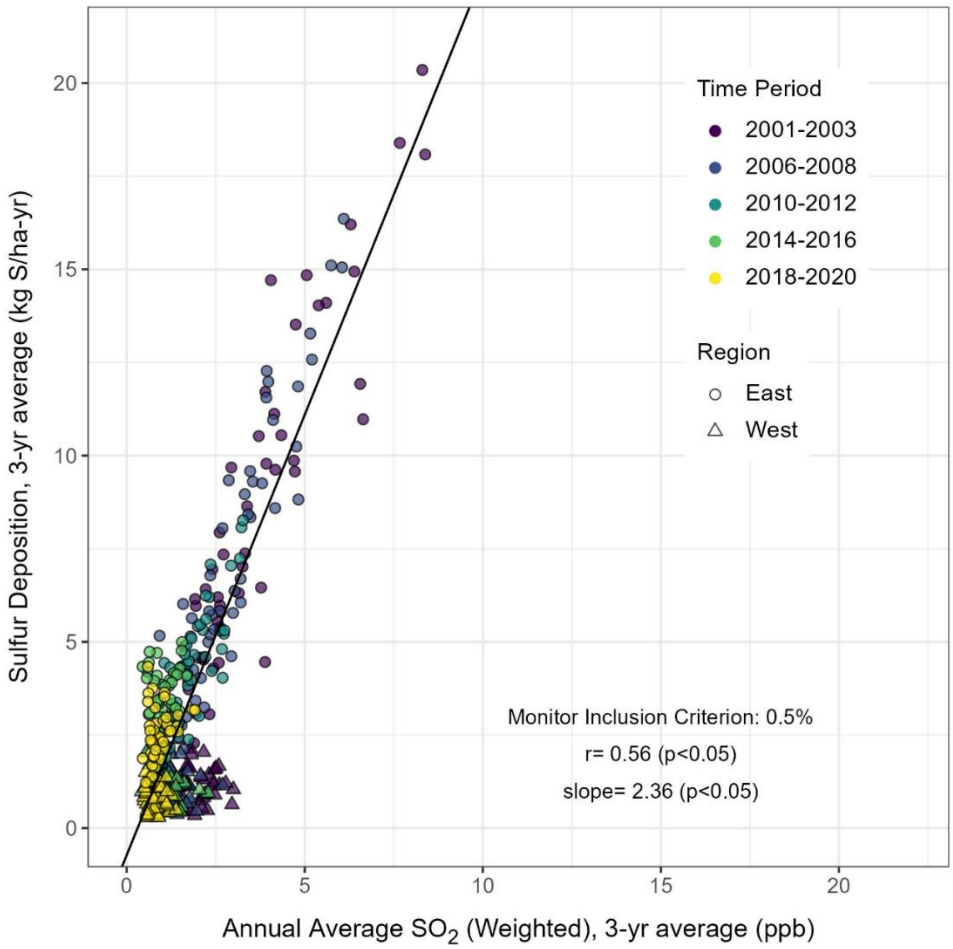

Figure 6-51. Plot of annual SO_2 EAQM values against TDep total S deposition across 84 ecoregions. The individual pairs are color-coded by 3-year periods and the symbols differentiate between sites in the eastern U.S. and western U.S. This figure is based on EAQM data using 120-hour trajectories, the NAM-12 meteorological data, and a monitor inclusion criterion of 0.5%.

6.4 KEY OBSERVATIONS

Based on the information above, this section discusses how well various air quality metrics relate to S and N deposition. We used five separate approaches to evaluate these relationships. The first approach consisted of a simple comparison of AQ and deposition trends over the past two decades, as a type of "real-world experiment," to determine how these two terms have correlated over recent periods. The strength of this approach is that it relies entirely on monitoring data and the observed trends. A limitation of this is that while one can observe correlation between the downward trends in emissions, air quality concentrations, and deposition in nitrogen and sulfur, more analyses are needed to determine that the trends in emissions and SO_2 and NO_X concentrations caused the decrease in deposition.

The second approach assessed how air quality concentrations and resultant deposition levels are related within a chemical-transport model (CMAQ) both nationally and then at certain Class 1 areas. The advantage of this particular approach is that it allowed comparison of air quality and deposition without some of the monitoring limitations that constrain other types of concentration to deposition relationships (e.g., could be assessed at every model grid cell, model estimates dry, wet, and total deposition which allows for more detailed comparisons). These comparisons have the disadvantage of being subject to model input errors or imperfect model parameterizations.

The third approach focused exclusively on a subset of monitoring sites where detailed air quality data (CASTNET, IMPROVE) were collocated with wet deposition measurements (NADP). Comparisons at these 27 Class 1 sites allowed for an evaluation of the association between wet deposition of N and S against concentrations of multiple gaseous and particulate N- and S-containing chemicals and $PM_{2.5}$. The strength of this analysis is that it is entirely based on monitoring data (i.e., no contribution from air quality modeling as in analyses involving TDep estimates). The primary limitations of this approach are (1) that the collocated measurements are only available in certain locations (mostly in the western U.S.) and thus any associations may not be representative of national conditions; and (2) that the deposition data do not include dry deposition.

The fourth approach looked at the associations between measured air quality concentrations (SO_2, NO_2, and $PM_{2.5}$) and TDep estimates of deposition at all sites that measure those pollutants across the U.S. This allows for a robust comparison of local concentrations and local deposition across the U.S. This analysis is particularly relevant given that the current standards (both primary and secondary) are judged using design value metrics based on measurements at the current SO_2, NO_2 and $PM_{2.5}$ SLAMS monitors. Many of these monitors are in the areas of higher pollutant concentrations, and many are sited near sources of SO_X and N oxides emissions. For example, as discussed in section 2.3, many SO_2 monitors are sited near large point sources of SO_2 (e.g., electric generating units) and for NO_2, larger urban areas are required to site NO_2 monitors near larger roadways with a focus on mobile source emissions. One limitation of this approach is that it does not account for deposition associated with the transport of pollutants emitted some distance upwind.

The fifth approach assesses relationships between a composite air quality metric (EAQM) and TDep estimates of deposition within downwind ecoregions. This approach provides a way to account for the air quality data at upwind locations with the potential to influence downwind deposition. One limitation of this approach is that it is challenging to identify the upwind sites with the potential to influence downwind deposition. The fourth and fifth approaches may be

affected by biases in the model simulations and uncertainties in the interpolation method used to create the TDep product.

No single approach to assessing the relationship between concentrations and deposition is perfect. Each of them is informative to our understanding. The conclusions discussed below are based on an evaluation of the results from all five approaches, with higher weighting assigned to the fourth, fifth, and first approaches, in that order.

6.4.1 SO_2 Metrics

As introduced in Chapter 2, and discussed earlier in this chapter, S tends to deposit as SO_2 close to sources of SO_2 emissions but as SO_4^{2-} in areas further away, including more rural areas of the country. In the western U.S., where S tends to be low, S may deposit more equally from SO_2 and SO_4^{2-}. Section 6.2 considers the current form and averaging time of the SO_2 secondary NAAQS (i.e., the 2nd highest 3-hour daily maximum for a year) in the deposition to air quality analyses. Additionally, given that the deposition-related impacts examined in this review are associated with deposition over some longer period of time (e.g., growing season, year, multi-year), this chapter also assesses an SO_2 air quality metric in the form of an annual average. Additionally, noting the many factors that can lead to variability in the deposition, including frequency of precipitation, and micrometeorological factors relevant to the dry deposition velocity, the analyses focus on a 3-year average for each of the air quality and deposition metrics and include multiple years of data to better assess more typical relationships.

Starting with the annual SO_2 metric and the fourth approach, we observe that the comparisons of annual SO_2 concentrations (averaged over 3 years) and deposition estimates at the SLAMS for the same time periods indicate that the two entities are strongly correlated (r = 0.70). This is especially true for the earlier periods of the record (e.g., 2001-2003) and across the eastern U.S. (Figures 6-35). While there are exceptions, there is a general association of SLAMS with higher annual average SO_2 concentrations with higher local S deposition estimates. The EAQM analyses in the fifth approach then extend the conclusion that annual SO_2 is also likely a good indicator for regional S deposition levels. The EAQM-weighted comparisons of annual average SO_2 (averaged over 3 years) in the eastern U.S. exhibit a high degree of correlation (r = 0.85 and 0.65, Figures 6-40 and 6-41). Finally, per the first approach, we note that the observed declines in national levels of S deposition over the past two decades has occurred during a period in which emissions of SO_2 have also declined sharply (Figures 6-52).

Figure 6-52 displays the trend in SO_2 emissions (averaged over 3 years) nationally for five time periods from 2001 through 2020. Figure 6-53 displays the distributions of median S deposition estimates for the 84 ecoregions in the CONUS for the same five time periods. The two parameters (annual average SO_2 emissions and S deposition) have exhibited consistent

decreases across the 20-year period, suggesting again that SO_2 may be a good indicator for a secondary standard associated with S deposition.

The decline in ambient air SO_2 concentrations observed at SLAMS monitors (Figure 6-7) is not as sharp as the decline in SO_2 emissions or in ecoregion median S deposition. This is likely due to the direct relationship of emissions with atmospheric loading of S compounds, which is then directly related to S deposition. The ambient air monitoring dataset is not limited to only monitors with consistent monitoring across the time period examined, and the monitor locations are not uniformly distributed across the U.S. and/or are not necessarily sited adjacent to all significant sources operating in all of the time periods examined. Even so, the ambient air concentration declines are consistent over the period and exhibit correlations with the declining trend in S deposition.

The different approaches for examining relationships between SO_2 concentrations and S deposition indicate associations between the two variables, locally, regionally, and nationally. We also find that the CMAQ comparisons (second approach) and the Class I areas analyses (third approach) also indicate annual average SO_2 concentrations to generally be associated with S deposition. We note, however, that many of the relationships between SO_2 concentrations and S deposition values become much weaker when S deposition levels are less than 5 kg/ha-yr (e.g., across the western U.S., more recent S deposition levels in the eastern U.S.). There is also substantial scatter at these lower deposition values, calling into question the ability to identify a specific SO_2 concentration and metric that might be consistently associated with deposition below approximately 5 kg/ha-yr.

In addition to the annual average SO_2 metric, the current 3-hour SO_2 metric also appears to relate to S deposition. The correlations for S deposition with this metric and the annual metric vary across all the approaches, with one or the other having a somewhat higher correlation than the other. Overall, the metrics demonstrate similar strength in correlation, with r values for the full datasets ranging from about 0.5 to 0.7. Thus, as the focus in this review is on annual deposition across sensitive regions, we conclude that the SO_2 annual average, averaged over three years, would likely be the better metric for consideration of policy options to address S deposition-related effects.

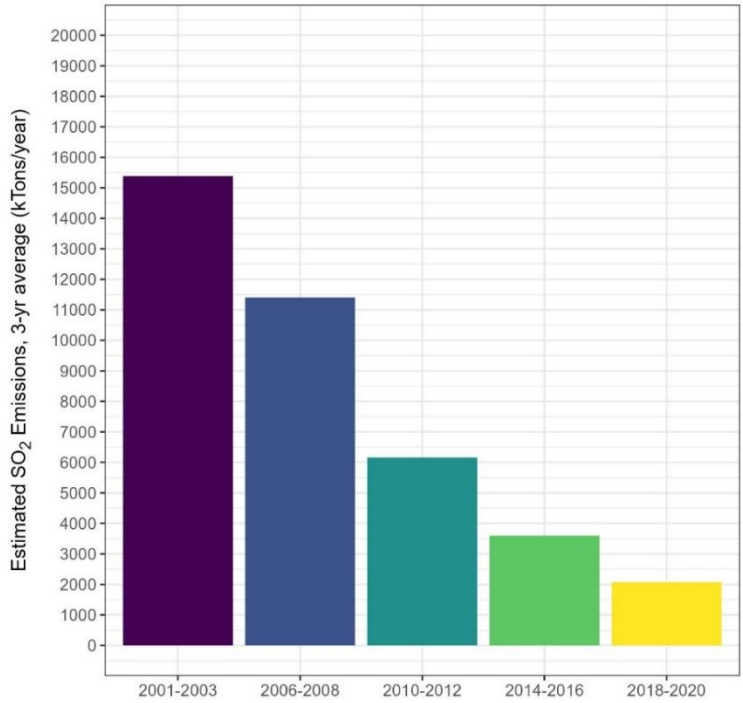

Figure 6-52. Estimated annual SO_2 emissions, nationally (NEI), averaged over three years, from 2001-2020.

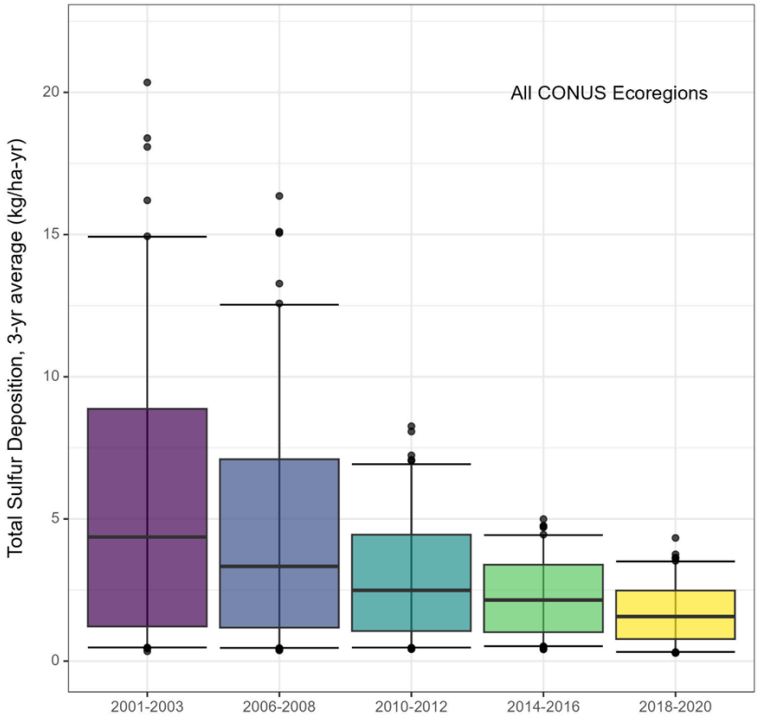

Figure 6-53. TDep estimates of ecoregion median S deposition. Whiskers mark 5th and 95th percentiles; estimates above 95th percentiles are black dots.

6.4.2 NO_2 and $PM_{2.5}$ Metrics

Both NO_2 and certain components of $PM_{2.5}$ can contribute to N deposition. As was the case for SO_2 and S deposition, there are multiple pathways for N deposition (dry and wet), and multiple scales of N deposition (local and regional). However, there are some additional complications in the consideration of how air quality concentrations (i.e., NO_2 and $PM_{2.5}$ mass) are associated with eventual N deposition. First, not all N deposition is caused by the criteria pollutants. As discussed in Chapter 2, ammonia emissions also lead to N deposition, especially through dry deposition at local scales. Second, only certain components of $PM_{2.5}$ mass contribute to N deposition (i.e., nitrate and ammonium). As a result of these two complications, there is reason to expect that the association between NO_2 concentrations and N deposition, and $PM_{2.5}$ concentrations and N deposition will be less robust than what we observed for SO_2.

Considering NO_2, we note that the current form and averaging time of the NO_2 secondary NAAQS is the annual average NO_2 concentration. As in the assessments of SO_2 metrics, these analyses focus on a 3-year average of NO_2 and N deposition and include multiple years of data to better assess more typical relationships. At the SLAMS, there was only weak association ($r = 0.38$) between NO_2 concentrations, and the N deposition levels at those locations (Table 6-6, Figure 6-38). The associations were stronger in the western U.S., which are generally less affected by ammonia. The comparisons of collocated NO_2 and N deposition at the 27 Class 1 sites (mostly western U.S.) confirmed this conclusion. The regional EAQM comparisons confirm that the associations between NO_2 and N deposition are much smaller than what was observed for SO_2 and S deposition, but the regional signals are different than what was observed with the local SLAMS comparisons, i.e., some weak positive association ($r = 0.48$ and $r = 0.35$, Figures 6-44 and 6-45) in the eastern U.S., but no association in the western U.S. When considering national trends over the past 20 years, we note that sharp declines in NO_2 emissions and concentrations are linked in time with sharp declines in oxidized N deposition (Table 6-2), but the same is not true when considering total atmospheric N deposition. For the five time periods, figure 6-54 displays the distributions of annual average NO_2 concentrations (averaged over 3 years) at SLAMS monitors with valid data. Figure 6-55 displays the distributions of median N deposition amounts at the 84 ecoregions across the same time periods. In the earliest two periods (2001-2003, 2006-2008) both parameters exhibited decreases. However, since 2010, NO_2 concentrations have continued to drop while N deposition has remained steady. In sum, the evidence suggests that NO_2 would be a weak indicator of total atmospheric N deposition, especially in areas where ammonia is prevalent.

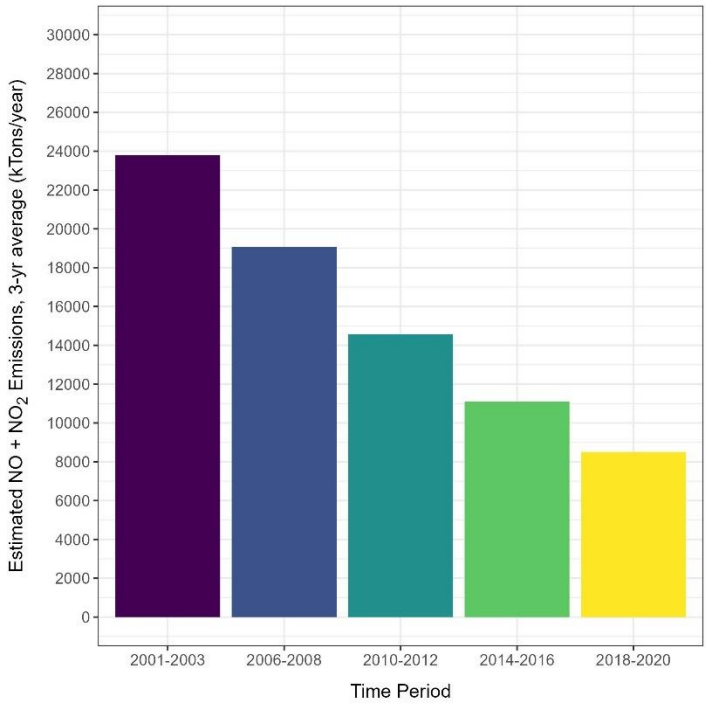

Figure 6-54. Estimated annual NO_2 emissions, nationally (NEI), averaged over three years, from 2001-2020.

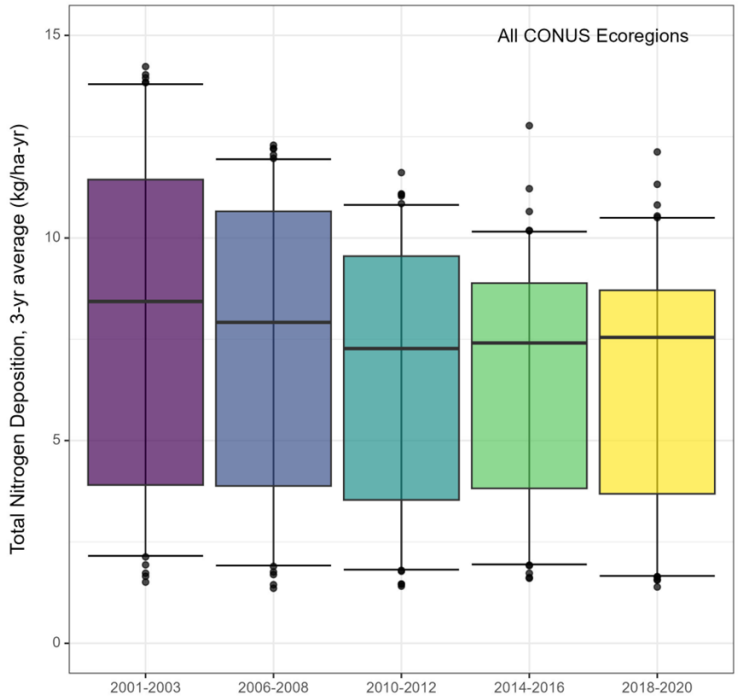

Figure 6-55. Box and whisker plot of TDep estimates of median total N deposition in all CONUS ecoregions (2001-2020). Whiskers show the 5th and 95th percentiles, with data points outside the 5th and 95th percentiles shown as black dots.

Given this finding that NO_2 would be a poor indicator of total atmospheric N deposition and our understanding about these relationships, we also evaluated $PM_{2.5}$ annual average, averaged over three years, recognizing that it captures particulate ammonium. Analyses at the SLAMS suggest some moderate correlation (r = 0.57) between the two parameters (Figure 6-39). The shallowness of this association and the variation (or scatter) in both parameters, however, indicates that $PM_{2.5}$ would not be expected to provide a useful or effective indicator for a policy option for limiting N deposition.

Consistent with the SO_2 and NO_2 comparisons against deposition, the associations between $PM_{2.5}$ and N deposition were stronger in the earlier time periods. The more regionally focused EAQM results confirm this moderate correlation (r = 0.62), but only at the eastern U.S. sites (Figure 6-46 and 6-47), with near-zero correlation in the western U.S. Again however, it is important to recognize that any $PM_{2.5}$ to N deposition associations will be affected by the fact that some parts of the $PM_{2.5}$ total mass do not contribute to N deposition (e.g., organic carbon, elemental carbon). Figure 6-56 shows the fraction of total $PM_{2.5}$ that is attributable to the sum of particulate nitrate and ammonium at CSN sites for the 2020-2022 average. The median across sites is less than 20%site and the highest fraction is in Riverside County, CA where the value is 30%. Further, this fraction has declined since the 2006-2008 period, the first for which these data are available. In sum, the evidence suggests that $PM_{2.5}$ would be a weak indicator of total atmospheric N deposition, especially in areas where other components of the $PM_{2.5}$ total are dominant.

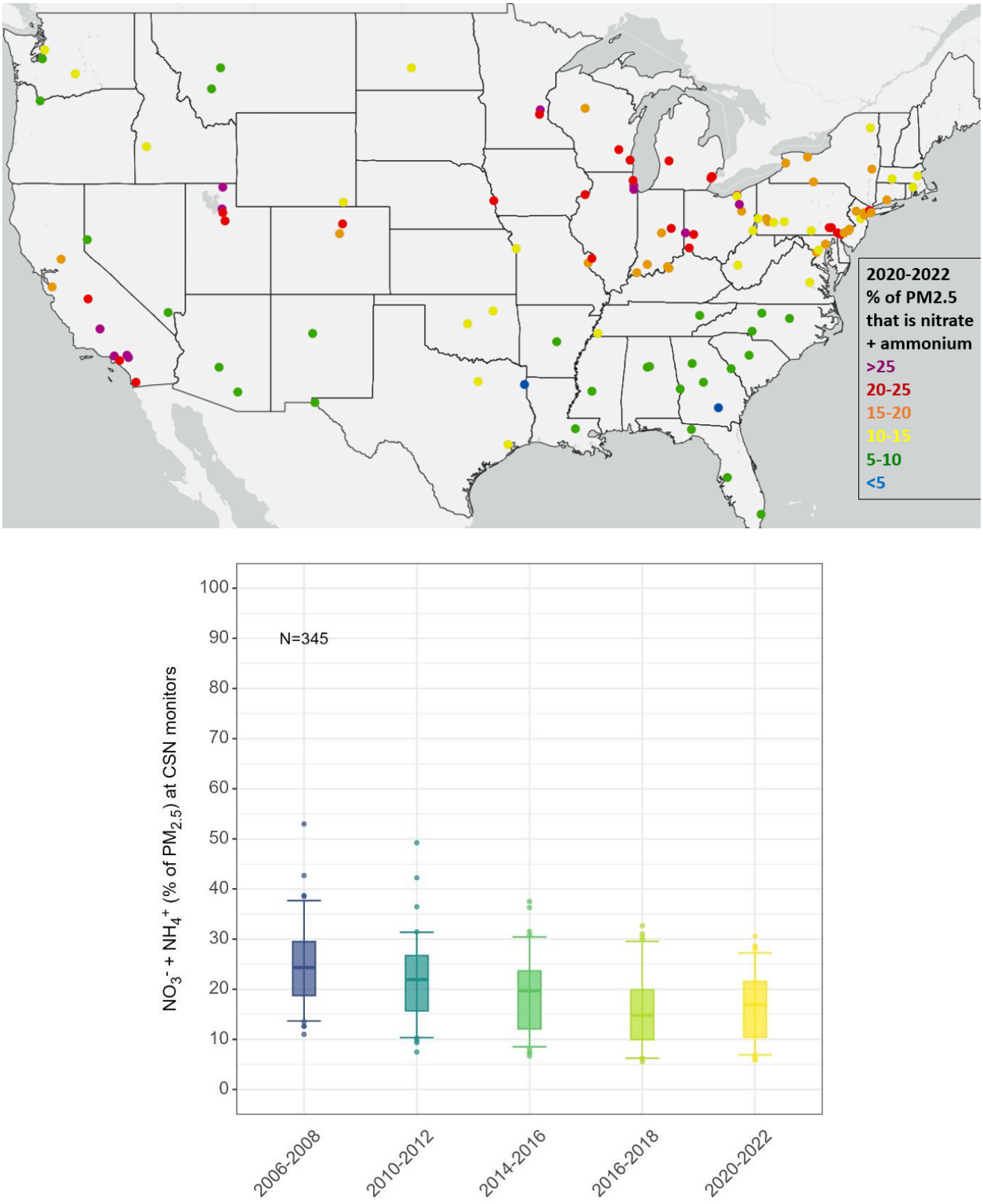

Figure 6-56. Fraction of total $PM_{2.5}$ at CSN sites that is either NO_3^- or NH_4^+ in 2020-2022 (upper) and across five time periods at consistently sampled sites (lower).

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7 REVIEW OF THE STANDARDS

In considering what the currently available evidence and exposure/risk information indicate with regard to the current secondary SO_2, NO_2 and PM standards, the initial overarching question we address is:

- **Do the currently available scientific evidence and air quality and exposure analyses support or call into question the adequacy of the protection afforded by the current secondary standards?**

To assist us in interpreting the currently available scientific evidence and quantitative information, including results of recent and past quantitative analyses to address this question, we have focused on a series of more specific questions. In considering the scientific and technical information, we consider both the information previously available and information newly available in this review which has been critically analyzed and characterized in the current ISA, the 2008 ISA for the oxides of N and S, the 2009 ISA for PM, and prior AQCDs for all three criteria pollutants. In so doing, an important consideration is whether the information newly available in this review alters the EPA's overall conclusions from the last reviews regarding ecological effects associated with oxides of N and S and with PM in ambient air. We also consider the currently available quantitative information regarding environmental exposures (characterized by the pertinent metric) likely to be associated with the air quality metric representing the current standards. Additionally, we consider the significance of these exposures with regard to the potential for ecological effects, their potential severity and any associated public welfare implications.

Within this chapter, sections 7.1 and 7.2 discuss the evidence and exposure-based questions regarding policy-relevant aspects of the currently available information on welfare effects, public welfare implications, the current standards and as appropriate, consideration of potential alternatives. Section 7.1 addresses the questions in the context of effects other than those related to ecosystem deposition of S and N compounds and, in similar fashion, section 7.2 addresses policy-relevant questions in the context of deposition-related effects. Advice received from the CASAC on the standards is summarized in section 7.3. Staff conclusions derived from the evaluations presented in this PA are described in section 7.4. Section 7.5 identifies key uncertainties and areas for future research.

7.1 EVIDENCE AND EXPOSURE/RISK BASED CONSIDERATIONS FOR EFFECTS OTHER THAN ECOSYSTEM DEPOSITION-RELATED EFFECTS OF S AND N

In considering the currently available evidence and quantitative information pertaining to ecological effects of oxides of N and S and PM in ambient air other than those associated with ecosystem deposition of S and N, we focus on addressing several questions (listed below). Included in this consideration is what this information indicates regarding effects, and associated public welfare implications, that might be expected to occur under air quality meeting the existing standards.

- **To what extent has the newly available information altered our scientific understanding of the ecological effects of oxides of S and N and PM in ambient air?**
- **To what extent does the currently available information indicate the potential for exposures associated with ecological effects under air quality meeting the existing standards? If so, might such effects be of sufficient magnitude, severity, extent and/or frequency such that they might reasonably be judged to be adverse to public welfare?**
- **To what extent have important uncertainties identified in past reviews been reduced and/or have new uncertainties emerged?**

Framed by these questions, we consider the evidence and quantitative information for the three criteria pollutants in the subsections below.

7.1.1 Sulfur Oxides

As summarized in section 4.1 above, the previously available evidence base describes the direct effects of SO_X in ambient air on vegetation, and very little of the currently available information is newly available in this review. Among the SO_X — which can include SO, SO_2, SO_3, and S_2O — only SO_2 is present in the lower troposphere at concentrations relevant for environmental considerations (ISA, Appendix 2, section 2.1). Sulfate is the prominent S oxide present in the particulate phase. The available evidence, largely comprising studies focused on SO_2, documents the effects of SO_2 on vegetation, including foliar injury, depressed photosynthesis and reduced growth or yield (ISA, Appendix 3, section 3.2). The newer studies continue to support the determination also reached in the last review that the evidence is sufficient to infer a causal relationship between gas-phase SO_2 and injury to vegetation (ISA, Appendix 3, section 3.6.1).

The SO_2 effects evidence derives from a combination of laboratory studies and observational studies. In general, effects on plants occur at SO_2 exposures higher than a 3-hour average concentration of 0.5 ppm. For example, a recent laboratory study reports some transient effects on lichen photosynthesis for short exposures, with more long-lasting effects only

observed for exposures of nearly 1 ppm SO_2, as summarized in section 5.4.1 above. With regard to the sensitive effect of foliar injury, the current ISA states there to be "no clear evidence of acute foliar injury below the level of the current standard" (ISA, p. IS-37). Further, the "limited new research since 2008 adds more evidence that SO_2 can have acute negative effects on vegetation but does not change conclusions from the 2008 ISA regarding … the SO_2 levels producing these effects" (ISA, p. IS-37).

Uncertainties associated with the current information are generally similar to those existing at the time of the last review. In large part these uncertainties relate to limitations of experimental studies in reflecting the natural environment and limitations of observational studies in untangling effects of SO_2 from those related to other pollutants that may have influenced the analyzed effects. Regardless of these uncertainties, we note that the evidence from either type of study indicates exposures associated with effects to generally be associated with air concentrations and durations which would not be expected to occur when the current standard is met.

7.1.2 Nitrogen Oxides

The currently available information on direct effects of N oxides in ambient air, which generally concerns effects on plants and lichens (as summarized in section 4.1 above), is comprised predominantly of studies of NO_2 and HNO_3, and also of PAN. The very few studies newly available in this review do not alter our prior understanding of effects of these N oxides, which include visible foliar injury and effects on photosynthesis and growth at exposures considered high relative to current levels in ambient air (ISA, Appendix 3, section 3.3). Thus, as in the last review, the ISA again concludes that the body of evidence is sufficient to infer a causal relationship between gas-phase NO, NO_2, and PAN and injury to vegetation (ISA, section IS.4.2).

Regarding another N oxide compound, HNO_3, the previously available evidence included experimental studies of leaf cuticle damage in tree seedlings, a finding confirmed in a more recent study, as well as effects on lichens, as summarized in section 5.4.2 above. Effects of HNO_3 may be related to vapor exposures and gaseous uptake or, given the very high deposition velocity of HNO_3, to direct contact via deposition on surfaces (ISA, Appendix 2, section 2.5.2.1 and Appendix 3, section 3.4). Among other studies, the evidence includes studies of effects related to historic conditions in the Los Angeles basin, although no such studies are available for other areas of the U.S. A more recent 2008 reassessment of an area in the Los Angeles basin in which there was a significant decline in lichen species in the late 1970s found that lichen communities have not recovered from the damage evident in the 1970s, as described in section 5.4.2 above (ISA, Appendix 3, section 3.4). The newer studies continue to support the findings

of the 2008 ISA, such that as in the last review, the ISA again concludes "the body of evidence is sufficient to infer a causal relationship between gas-phase HNO_3 and changes to vegetation" (ISA, Appendix 3, p. 3-17).

With regard to the exposure concentrations, we note that the ISA concludes that for NO_2 "[w]ith few exceptions, visible injury has not been reported at concentrations below 0.20 ppm, and these exceptions occurred when the cumulative duration of exposures extended to 100 hours or longer" (ISA, Appendix 3, p. 3-8). Effects on plant photosynthesis and growth have resulted from multiday exposures of six or more hours per day to NO_2 concentrations above 0.1 ppm, with a newly available study documenting effects at exposures of 4 ppm NO_2, effects that the ISA finds to be "consistent with past studies of plants with relatively high NO_2 exposure" (ISA, Appendix 3, p. 3-12). Regarding PAN, there is "little evidence in recent years to suggest that PAN poses a significant risk to vegetation in the U.S." (ISA, Appendix 3, p. 3-13).

The recently available information for HNO_3 includes effects on tree foliage under controlled 12-hour exposures to 50 ppb HNO_3 (approximately 75 $\mu g/m^3$). Foliar damage was also reported in longer, 32- or 33-day exposures in which peak HNO_3 concentrations for the "moderate" treatment (30-60 $\mu g/m^3$) encompassed the range reported in summers during the 1980s in the Los Angeles Basin, as described in section 5.4.2 above (ISA, Appendix 3, section 3.4). During that period, NO_2 concentrations in the Basin ranged up to 0.058 ppm, exceeding the secondary standard (U.S. EPA, 1987). Effects on lichen photosynthesis have been reported from 6.5-hour daily varying exposures with peaks near 50 ppb (approximately 75 $\mu g/m^3$) that extend beyond 18 days (ISA, Appendix 6, section 6.2.3.3; Riddell et al., 2012).

In considering the potential for concentrations of N oxides associated with welfare effects to occur under air quality conditions meeting the current NO_2 standard, we consider the air quality information summarized in section 2.4.1 above. In so doing, we note that air quality at all sites in the contiguous U.S. has met the existing secondary NO_2 standard since around 1991 (Figure 2-22). During the period 1983 to 1991, the 99^{th} percentile of annual mean NO_2 concentrations at sites nationwide was near the level of the standard (Figure 2-22). Further, hourly NO_2 concentrations during this time indicate little likelihood of an occurrence of a 6-hour concentration of magnitude for which plant growth effects were reported from experimental studies (as described in section 5.4.2), as the 98^{th} percentile of 1-hour concentrations rarely exceeded 0.2 ppm, as shown in Figure 2-21.

In considering the potential for HNO_3 concentrations of a magnitude to pose risk of effects to occur in conditions that meet the current NO_2 secondary standard, we recognize, as summarized in section 5.4 above, that the evidence indicates N oxides, and particularly, HNO_3, as "the main agent of decline of lichen in the Los Angeles basin" (ISA, Appendix 3, p. 3-15), where elevated concentrations of N oxides were documented during the 1970s to 1990s (and

likely also occurred earlier). Based on a limited number of studies extending back to the 1980s, HNO_3 has been suspected to have had an important role in these declines, as summarized in section 5.4.2 above. During that time period the Los Angeles metropolitan area experienced NO_2 concentrations in excess of the NO_2 secondary standard (e.g., annual average NO_2 concentrations up to 0.078 ppm in 1979 and above 0.053 ppm into the early 1990s). Surveys in 2008, when NO_2 concentrations were well below the standard, reported that the impacts documented on lichen communities in the 1970s still remained (ISA, Appendix 3, section 3.4). Although the extent to which this relates to lag in recovery or concurrent air pollutant concentrations is unknown, we take note of the risk posed from HNO_3 contact with plant and lichen surfaces. This risk likely relates to the direct exposure of these surfaces to air pollutants, the high deposition velocity of HNO_3 (ISA, Appendix 2, section 2.5.2.1) and its acidity. Given these factors, we recognize that the risk of HNO_3 effects to lichens may be from both direct and deposition-related exposure related to direct contact of the chemical to the lichen surfaces.

In summary, the currently available information is somewhat limited with regard to the extent to which it informs conclusions as to the potential for exposures associated with ecological effects under air quality meeting the existing NO_2 secondary standard. More recent studies extending into more recent periods indicate variation in eutrophic lichen abundance to be associated with variation in metrics representing N deposition (ISA, Appendix 6, section 6.2.3.3). The extent to which these associations are influenced by residual impacts of the historic air quality is unclear.

While new uncertainties have not emerged, uncertainties remain in our interpretation of the evidence, including those related to limitations of the various study types. For example, the various types of studies in the evidence for welfare effects of the different N oxides vary with regard to their limitations, and associated uncertainties. Field studies are limited with regard to identification of threshold exposures for the reported effects and uncertainties associated with controlled experiments include whether the conditions under which the observed effects occur would be expected in the field. A key uncertainty affecting interpretation of studies of historic conditions in the LA Basin relates to the extent to which other air pollutants or local conditions (unrelated to N oxides) may have contributed to the observations of effects, and whether such effects would be expected in response to N oxides in other locations in the U.S. (and the extent to which the conditions unrelated to N oxides differ in other locations). With regard to the risk posed by N oxides, and particularly HNO_3, the evidence, as summarized in sections 5.4.2 and 5.3.3 above indicates the potential for effects of air quality occurring during periods when the current secondary standard was not met. The evidence is limited, however, with regard to support for conclusions related to conditions meeting the current standard.

7.1.3 Particulate Matter

As summarized in section 5.4.3 above, the evidence for ecological effects of PM is consistent with that available in the last review. The ISA causal determinations with regard to ecological effects of PM in the 2012 PM review and in this review focused on deposition-related effects, rather than direct effects of PM in ambient air. In this review, as in the last one, the ecological effects evidence was found to be sufficient to conclude there is likely to exist a causal relationship between deposition of PM and a variety of effects on individual organisms and ecosystems (ISA, Appendix 15; 2009 PM ISA, section 9.4).

With regard to direct effects of PM in ambient air, the associated information on ambient air concentrations associated with effects is well in excess of the existing secondary standards. While some uncertainties remain, new uncertainties have not emerged since the last review. In summary, little information is available on the assessment of direct effects of PM in exposure conditions likely to meet the current standards, and the limited available information does not indicate direct effects to occur under those conditions.

7.2 EVIDENCE AND EXPOSURE/RISK-BASED CONSIDERATIONS FOR S AND N DEPOSITION-RELATED EFFECTS

In this section, we consider the evidence and quantitative exposure/risk information related to ecological effects of N and S deposition associated with S oxides, N oxides and PM in ambient air. We do this in the larger context of evaluating the protection from such effects provided by the existing standards and potential alternative standards. The potential for the three criteria pollutants to all contribute to particular ecosystem effects while also having a potential for independent effects poses challenges to the organization of the discussion. A particular focus of this chapter is on considering quantitative aspects of the relationships between deposition and ecosystem effects that can inform decisions on standards that provide the appropriate control of deposition for the desired level of protection from adverse environmental effects. As recognized in Chapter 5 and the associated appendices, the availability of quantitative information for relating atmospheric deposition to specific welfare effects varies across the categories of effects. We consider here the extent to which such information is available that might support characterization of the potential for effects, and of the protection that might be afforded for such effects, under different air quality conditions.

While recognizing there are multiple organizations that could be applied, we have adopted one that focuses first on consideration of the evidence for welfare effects associated with atmospheric deposition of both S and N compounds, including the nature of effects and associated uncertainties in the evidence (section 7.2.1) and then consideration of the quantitative information and risk estimates particular to first S deposition (section 7.2.2) and then N

deposition (section 7.2.3). Further, within sections 7.2.2 and 7.2.3, we first consider the evidence regarding deposition effects and the quantitative information or analysis results for effects of potential public welfare significance, and then consider relationships between relevant air quality metrics and deposition levels that may be appropriate to target when considering the appropriate degree of public welfare protection that the secondary standards should afford.

7.2.1 Evidence of Ecosystem Effects of S and N deposition

A long-standing evidence base documents the array of effects of acidic deposition in aquatic and terrestrial ecosystems and the effects associated with ecosystem N enrichment. The evidence for acidic deposition effects, extending back many decades, has accrued in part through study of ecosystem acidification that has resulted from many decades of acidifying deposition (ISA, section ES.5.1 and Appendix 4, section 4.6). As discussed in prior chapters, both S and N compounds have contributed to ecosystem acidification, with relative contributions varying with emissions, air concentrations and atmospheric chemistry, among other factors. Ecological effects have been documented comprehensively in waterbodies of the Adirondack and Appalachian Mountains, and forests of the Northeast, at the organism to ecosystem scale. With regard to N enrichment, research on its effects in estuaries and large river systems across the U.S. extends back at least four decades (2008 ISA, section 3.3.2.4; Officer et al., 1984). Further, the evidence base on the effects of N enrichment on terrestrial ecosystems, primarily in grassland and forested ecosystems, extends back to the last review (e.g., 2008 ISA, sections 3.3.3 and 3.3.5). We consider the evidence of these effects, and others more recently understood, as characterized in the ISA and summarized in Chapter 4, in the context of the following questions.

- **To what extent has the newly available information altered our scientific understanding of the ecological effects of atmospheric deposition of N and S compounds?**

The current evidence, including that newly available in this review, supports, sharpens and expands somewhat on the conclusions reached in the 2008 ISA for the review completed in 2012. The long-standing evidence continues to support determinations of causal relationships between acidifying deposition of N and S compounds and N deposition and an array of effects in terrestrial and aquatic ecosystems, as in the last review (ISA, Table ES-1).

A wealth of scientific evidence, spanning many decades, demonstrates effects of acidifying deposition, associated with N and S compounds, in aquatic and terrestrial ecosystems (ISA, sections ES.5.1, IS.5.1, IS.5.3, IS.6.1 and IS.6.3; 2008 ISA, section 3.2; U.S. EPA, 1982, Chapter 7). Accordingly, consistent with the evidence in the last review, the currently available evidence describes an array of acidification-related effects on ecosystems. The current evidence base, which includes an abundance of longstanding evidence, supports conclusions also reached

in the last review of causal relationships between N and S deposition and alteration of soil and aquatic biogeochemistry, alteration of the physiology and growth of terrestrial organisms and of associated productivity, changes in aquatic biota, including physiological impairment, and alteration of species richness, community composition and biodiversity in both aquatic and terrestrial ecosystems (ISA, Table ES-1).

Similarly, a robust evidence base demonstrates ecosystem effects of N enrichment. In both estuarine and freshwater ecosystems, the current evidence, including a wealth of longstanding evidence, also supports conclusions reached in the last review of a causal relationship between N deposition and changes in biota, including altered growth and productivity, and alteration of species richness, community composition and biodiversity due to N enrichment (ISA, sections ES.5.2, IS.6, and IS.7, and Table ES-1). In addition to evidence in freshwater systems, this evidence base also includes longstanding evidence of effects in estuaries along the East and Gulf Coasts of the U.S., as summarized in more detail in Chapters 4 and 5 (ISA, Appendix 7, section 7.2.9). Additional effects of N deposition in wetlands, also recognized in the last review, include alteration of biogeochemical cycling, growth, productivity, species physiology, species richness, community composition and biodiversity.

In terrestrial ecosystems, as in the last review, the evidence supports determination of a causal relationship between N deposition and alteration of species richness, community composition and biodiversity. The ISA additionally determines there to be a causal relationship for alteration of the physiology and growth of terrestrial organisms and associated productivity, a category of effects not included in the 2008 ISA (ISA, Table ES-1). The studies available since the last review provide further evidence that addition of N to sensitive ecosystems "alters plant physiological processes, stimulates the growth of most plants and broadly increases productivity" (ISA, Appendix 6, p. 6-188). Further there is evidence of effects on soil microbes and symbiotic mycorrhizal, with the evidence as a whole indicating the sensitivity of plants, microorganisms and ecosystem productivity to N availability (ISA, Appendix 6, section 6.6.1). With regard to species richness, community composition and biodiversity, the evidence base is expanded from the last review, with regard to observational studies and N addition studies in grass and shrub communities of the Southwest, as summarized in sections 4.3.2.2 and 5.3.3.1 above.

Other evidence of effects causally associated with S deposition in wetland and freshwater ecosystems includes that related to chemical transformation and associated toxicity. This includes alteration of mercury methylation, which was also recognized in the last review. The other category of effects, which was not included in the last review, is that related to sulfide phytotoxicity, and its associated effects in wetland and freshwater ecosystems (ISA, Table ES-1).

- **To what extent have previously identified uncertainties in the evidence been reduced or do important uncertainties remain?**

The evidence base has expanded since the last review, as summarized above, and continues to be strong in documenting roles of SO_X, N oxides and PM (including N and S compounds) in aquatic acidification, nutrient enrichment and other effects. Some uncertainties associated with the evidence in the last review remain, and some additional uncertainties are important. In addition to uncertainties related to the specific air quality circumstances associated with effects (e.g., magnitude, duration and frequency of NO_2 and HNO_3 concentrations associated with effects), there are also uncertainties associated with the effects of N and S deposition expected under changing environmental circumstances, including reduced atmospheric loading with associated changes to soil and waterbody biogeochemistry and meteorological changes associated with changing climate (ISA, section IS.12).

Further, there are important uncertainties associated with the various assessment approaches employed by different study types. For example, uncertainties associated with observational studies include uncertainty regarding the potential influence of historical deposition on species distribution, richness and community composition observed in recent times (ISA, section IS.14.2.1). Further, there are uncertainties contributed by variation in physical, chemical and ecological responses to N and S deposition, and by the potential influence of unaccounted-for stressors on response measures. Uncertainties associated with addition experiments include, among others, those related to the potential for effects to occur over longer periods than those assessed in those studies (section 5.3.4.1). Lastly uncertainties associated with studies reporting atmospheric deposition associated with effects include authors' judgments on magnitude of responses identified as effects, as well as a lack of clarity as to references or baselines from which responses are assessed and with regard to judgments associated with reference or baseline conditions. Additionally, variability in physical, chemical and ecological characteristics of ecosystems contribute uncertainty to such judgments. As noted in the ISA, "[t]he majority of studies that evaluate terrestrial N CLs for N enrichment effects are based on observed response of a biological receptor to N deposition (or N addition as a proxy for deposition), without a known soil chemistry threshold that causes the biological effect" (ISA, p. IS-113).

7.2.2 S Deposition and S Oxides

To inform conclusions in this review related to the SO_X secondary standard, we consider a series of questions below that are intended to facilitate the evaluation of the linkages of SO_X in ambient air with S deposition and associated welfare effects. In considering these questions, we draw on the available welfare effects evidence described in the current ISA, the 2008 ISA for

oxides of N and S, the 2009 ISA for PM, and past AQCDs, and summarized in chapter 4. We do this in combination with the available information from quantitative analyses (and summarized in Chapters 5 and 6 above), both analyses recently developed and those available from the 2009 REA and considering the information now available.

7.2.2.1 Quantitative Information for Ecosystem Risks Associated with S Deposition

The currently available information provides modeling approaches for quantitatively analyzing linkages between S deposition, geochemical processes in soils and waterbodies, and indicators of aquatic and terrestrial ecosystem acidification risk. The use of such modeling approaches for characterizing potential risk of aquatic and terrestrial acidification is well established. Since the last review, aspects of the modeling approaches that quantify processes that are the major determinants of the indicators have been expanded and improved. Further, modeling approaches vary in their complexity, precision, and limitations, and the extent to which they inform different questions.

As recognized in Chapter 5 above, although the approaches and tools for assessing aquatic acidification have often been utilized for S and N deposition in combination, the approach taken in the aquatic acidification REA, summarized in section 5.1 above and described in detail in Appendix 5A, is focused on S deposition. This focus is supported by analyses (as summarized in section 5.1.2.4 above) indicating the relatively greater role of S deposition under the more recent air quality conditions that are the focus of this review. The aquatic acidification REA utilizes available site-specific water quality modeling that relates atmospheric deposition to ANC in a CL-based approach, as described in more detail in Appendix 5A. The site-specific modeling applications and associated estimates of CLs for different ANC thresholds or targets are publicly available in the NCLD.[1] The modeling applications most frequently utilize mass balance modeling tools for watershed processes (e.g., fluxes that affect watershed concentrations of anions and cations), although in some areas, dynamic modeling applications are prevalent (e.g., in the Adirondacks). In summary, the aquatic acidification assessment has utilized well-established site-specific water quality modeling applications with a widely recognized indicator of aquatic acidification, ANC.

Quantitative tools are also available for the assessment of terrestrial acidification related to S deposition, as they were in the last review (section 5.3.2.1; 2009 REA, section 4.3). Recently available studies have addressed a particular area of uncertainty identified for this approach in the last review (related to model inputs for base cation weathering). While updated analyses of terrestrial acidification have not been performed in this review, the findings from the

[1] The surface water acidification CLs used in the REA are from version 3.2.1 of the NCLD (Lynch et al., 2022).

analyses presented in the 2009 REA have been considered in the context of more recently available evidence (section 5.3.2.1; 2009 REA, section 4.3). Quantitative tools and approaches are not well developed for other ecological effects associated with atmospheric deposition of S compounds, such as mercury methylation and sulfide toxicity (summarized in sections 4.4.1 and 4.4.2 above).

In summary, as in the last review, we give primary attention to the quantitative approaches and tools for assessment of aquatic acidification (including particularly that attributable to S deposition). While recognizing the uncertainties associated with results of analyses utilizing these tools in the aquatic acidification REA, as summarized in section 5.1.5 above, we recognize these results to be informative to our purposes in identifying S deposition benchmarks associated with potential for aquatic acidification effects of concern. As described in section 3.3.2 above, this assessment of quantitative linkages between S deposition and potential for aquatic acidification is one component of the approach implemented in this PA for informing judgments on the likelihood of occurrence of such effects under differing air quality conditions.

- **To what extent does the available evidence support the use of waterbody ANC for purposes of judging a potential for ecosystem acidification effects?**

As described in section 4.2.1.2 above, ANC is an indicator of susceptibility or risk of acidification-related effects in waterbodies. Accordingly, the evidence generally indicates that the higher the ANC, the lower the potential for acidification and related waterbody effects, and the lower the ANC, the higher the potential. The support for this relationship is strongest in aquatic systems low in organic material, and the evidence comes predominantly from impacted waterbodies in the eastern U.S. (e.g., in the Adirondack Mountains) and Canada. In waterbodies with relatively higher levels of dissolved organic material (e.g., dissolved organic carbon), however, while the organic acid anions contribute to reduced pH, these anions create complexes with the dissolved aluminum, protecting resident biota against aluminum toxicity (ISA, Appendix 8, section 8.3.6.2). Accordingly, biota in such systems tolerate lower ANC values than biota in waterbodies with low dissolved organic carbon. Thus, while the evidence generally supports the use of ANC as an acidification indicator, the relationship with risk to biota differs depending on the presence of naturally occurring organic acids. Further, such natural acidity affects the responsiveness of ANC to acidifying deposition in these areas. As noted in section 5.1 above, the ecoregions in which ANC is less well supported as an indicator for acidic deposition-related effects due to the prevalence of waterbodies with high dissolved organic material include the Middle Atlantic Coastal Plain (ecoregion 8.5.1), Southern Coastal Plains (ecoregion 8.5.3), and Atlantic Coastal Pine Barrens (ecoregion 8.5.4). The evidence does, however, support the use of waterbody ANC in other areas for purposes of judging a potential for ecosystem acidification effects (section 5.1.2.2).

As summarized in sections 4.2.1.2 and 5.1.1 above, there is longstanding evidence of an array of impacts on aquatic biota and species richness reported in surface waters with ANC values below zero, and in some historically impacted waterbodies with ANC values below 20 µeq/L. The severity in impact is greatest for the ANC values below zero. This evidence derives primarily from lakes and streams of the Adirondack Mountains and areas along the Appalachian Mountains. The evidence base additionally indicates a potential for some increased risk to resident biota, depending on site-specific factors, of ANC levels between 20 and 50 µeq/L. As recognized in the last review, in addition to providing protection during base flow situations, ANC is a water quality characteristic that affords protection against the likelihood of decreased pH from episodic events in impacted watersheds. For example, waterbodies with ANC below 20 µeq/L have been generally associated with increased probability of low pH events, that, depending on other factors as noted above, have potential for reduced survival or loss of fitness of sensitive biota or lifestages (2008 ISA, section 5.1.2.1). In general, the higher the ANC level above zero, the lower the risk presented by episodic acidity. In summary, the available evidence provides strong support for the consideration of ANC for purposes of making judgments regarding risk to aquatic biota in streams impacted by acidifying deposition, and for consideration of the set of targets analyzed in the aquatic acidification REA: 20, 30, and 50 µeq/L (section 5.1 above).

- **What do the quantitative exposure/risk estimates indicate about acidification risks in freshwater streams and lakes for S deposition levels over the past two decades (including the time since the last review)? What are the important uncertainties associated with these quantitative risk estimates?**

In considering this question, we focus on the results of the aquatic acidification REA, as summarized in section 5.1 above (and described in detail in Appendix 5A). In summarizing the acidification risk estimates in Chapter 5, the different scales of analysis make use of water quality modeling-based CLs derived for three different ANC targets (20, 30 and 50 µeq/L). In this way we recognize both the differing risk that might be ascribed to the different ANC targets, as well as the variation in ANC response across waterbodies that may be reasonable to expect with differences in geology, history of acidifying deposition and different patterns of S deposition, and also recognize limitations and uncertainties in the use of ANC as an indicator for model-based risk assessments (section 5.1).

The national-scale analysis involved the 13,824 waterbody sites for which a CL based on ANC target was available (PA, section 5.1.3). As an initial matter, we note the appreciable

reduction in risk over the 20-year period of analysis.[2] For the 2001-03 period, more than 20% of waterbodies analyzed nationally were estimated to be unable to achieve an ANC of 20 µeq/L or greater based on S deposition estimates (Table 5-1). This percentage declines significantly by the 2010-12 period, and by the 2018-20 period, only 1% and 4% of waterbodies analyzed nationally were estimated to be unable to achieve or exceed ANC targets of 20 µeq/L and 50 µeq/L, respectively (Table 5-1).

The ecoregion-scale analyses focus on the 25 level III ecoregions (18 East and 7 West) in which there are at least 50 waterbody sites with CL estimates. This set of 25 ecoregions is dominated by ecoregions categorized as acid sensitive (Table 5A-5) and excludes the three ecoregions identified as having natural acidity related to organic acids (section 5.1.2.1). The ecoregion-scale results across the 20-year period reflect the results at the national scale, but the percentages of waterbodies not able to meet the ANC targets are higher than the national percentages due to the dominance of the acid-sensitive ecoregions among the 25. Specifically, in the most affected ecoregion (ecoregion 8.4.2, Central Appalachians), more than 50% of waterbodies were estimated to be unable to achieve an ANC of 20 µeq/L or greater based on S deposition estimates for the 2001-03 period; the percentage was close to 60% for an ANC target of 50 µeq/L (Figure 5-13). By the 2018-20 period, less than 10% of waterbodies in any of the 25 ecoregions (and less than 5% in all but one) were estimated to be unable to achieve an ANC of 20 µeq/L and less than 15% of waterbodies in the most affected waterbody were estimated to be unable to achieve an ANC of 50 µeq/L (Figure 5-13).

In considering this information we also note the uncertainties associated with such estimates. We recognize uncertainty associated with two overarching aspects of the assessment. The first relates to interpretation of specific thresholds of ANC with regard to aquatic acidification risk and the second relates to our understanding of the biogeochemical linkages between deposition of S and N compounds and waterbody ANC (which is reflected in the modeling employed), and the associated estimation of CLs.

With regard to the first, while ANC is an established indicator of aquatic acidification risk, there is uncertainty in our understanding of relationships between ANC and risk to native biota, particularly in waterbodies in geologic regions prone to waterbody acidity. Such uncertainties relate to a number of factors, including the varying influences of site-specific factors other than ANC. Such factors include prevalence of organic acids in the watershed, as well as historical loading to watershed soils that can influence acidity of episodic high-flow events.

[2] The aquatic acidification risk analyses conducted in the last review focused on the earliest part of this time period (e.g., deposition estimates derived using CMAQ modeling with 2002 wet deposition measurements and 2002 emissions (2009 REA, Appendix 4, p. 4-26).

With regard to the second aspect of the assessment, associated uncertainties are difficult to characterize and assess. Uncertainty in CL estimates is associated with parameters used in the steady-state CL models. While the SSWS and other CL models are well conceived and based on a substantial amount of research and applications available in the peer-reviewed literature, there is uncertainty associated with the availability of the necessary data to support certain model components. Such uncertainties contribute to uncertainty in estimation of the ANC levels that individual waterbodies might be expected to achieve under different rates of S deposition. This estimation is based on site-specific steady-state water quality modeling,[3] with associated limitations and uncertainties. For example, as recognized in sections 4.2.1.3 and 5.1.4 above, the data to support the site-specific model inputs for some areas are more limited than others, with associated greater uncertainties. Further, there are additional uncertainties associated with the estimates of S deposition for use in the analyses of CL exceedances, such as those for the national- and ecoregion-scale analyses (section 6.3.1, Table 6-13).

The strength of the CL estimates and the exceedance calculation rely on the ability of models to estimate the catchment-average base-cation supply (i.e., input of base cations from weathering of bedrock and soils and air), runoff, and surface water chemistry. The uncertainty associated with runoff and surface water parameters relates to availability of measurements, which varies among waterbodies. Further, the ability to accurately estimate the catchment supply of base cations to a water body is difficult, and uncertain (Appendix 5A, section 5A.3). This area of uncertainty is important because the catchment supply of base cations from the weathering of bedrock and soils is the factor with the greatest influence on the CL calculation and has the largest uncertainty (Li and McNulty, 2007).The ISA recognizes the model input for this (base cation weathering [BCw] rate) to be "one of the most influential yet difficult to estimate parameters in the calculation of critical acid loads of N and S deposition for protection against terrestrial acidification" (ISA, section IS.14.2.2.1). Although the approach to estimate base-cation supply for the national case study (e.g., F-factor approach) has been widely published and analyzed in Canada and Europe, and has been applied in the U.S. (e.g., Dupont et al., 2005 and others), the uncertainty in this estimate is unclear and could be large in some cases.

The REA included a quantitative analysis of uncertainty in CL estimates related to state-steady CL modeling inputs that involved many model simulations for the more than 14,000 waterbodies (in 51 ecoregions), drawing on Monte Carlo sampling of model input values, which provides a description of the uncertainty around the CL estimate in terms of the confidence interval for each waterbody mean result. Lower confidence intervals (indicating lower

[3] A small subset of waterbody CLs in Adirondacks region is based on dynamic modeling, simulating response in year 2100 or 3000 based on water quality parameter inputs from the somewhat recent past (Appendix 5A, section 5A.1.5).

uncertainty associated with model inputs) were associated with CLs determined with more extensive and longer-term water quality datasets and low variability in the runoff measurements. Critical load estimates for waterbody sites in the eastern U.S., particularly along the Appalachian Mountains, in the Upper Midwest, and in the Rocky Mountains, had smaller confidence intervals while larger intervals (greater uncertainty) were found for CLs in the Midwest and South and along the CA to WA coast (Appendix 5A, section 5A.3.1).

Consideration of such uncertainties informs the weighing of the findings of the quantitative analyses. For example, in light of the variation in uncertainty associated with CLs among the more to less well studied areas may indicate the appropriateness of a greater emphasis on the former and/or less emphasis on estimates for the upper end of the distribution. This information additionally informs interpretation of the potential risk associated with the different ANC targets.

7.2.2.2 General Approach for Considering Public Welfare Protection

In light of the available evidence, air quality and exposure/risk information, we discuss here key considerations in judging public welfare protection from S deposition in the context of the review of the secondary standard for SO_X.

- **What do the quantitative estimates of aquatic acidification risk indicate about deposition conditions under which waterbodies in sensitive ecoregions might be expected to achieve ANC levels of interest?**

In considering this question, we focus on the results of the aquatic acidification REA at three scales: national-scale, ecoregion-scale and the more localized case study-scale, as described in section 5.1 above. We give particular focus to the ecoregion and case-study analyses, which utilize the waterbody-specific comparisons of estimated deposition and waterbody CLs to provide ecoregion wide and cross-ecoregion summaries of estimated waterbody responses to ecoregion estimates of deposition. In so doing, we have considered the extent to which waterbodies in each ecoregion analyzed were estimated to achieve ANC levels at or above each of the three targets. In this way we recognize the variation in ANC response across waterbodies in an ecoregion that may be reasonable to expect based on both differences in watersheds that can affect sensitivity to S deposition and with different spatial or geographic patterns of S deposition. As summarized in section 6.1.1 above, S deposition levels will vary spatially or geographically due to differences in a number of factors including those related to upwind emissions of S-containing compounds and atmospheric chemistry, as well as patterns of other chemicals that can influence S deposition.

At the national-scale, as summarized in section 7.2.2.1 above, unlike the case for the 2000-2002 period, which was also analyzed in the last review, few waterbodies are estimated to

be receiving deposition in excess of their critical loads for relevant ANC targets under recent deposition levels. More specifically, for S deposition estimates for the most recent time period (2018-2020), only 4% of waterbodies nationally were estimated to exceed CLs for an ANC of 50 µeq/L and 1% for an ANC of 20 µeq/L (Table 5-1). In this time period (2018-2020), median estimates of deposition in all but one of the 69 ecoregions that are represented in these national-scale percentages (ecoregions with at least one site with a CL estimate) are below 4 kg S/ha-yr (Tables 5A-15 and 5A-11).

In the case study analyses, CL estimates for ANC targets of 20, 30 and 50 µeq/L are summarized for waterbodies in five sensitive areas, three areas in the eastern U.S. and two in the western U.S. (Table 5-6). The most well studied of these, the Shenandoah Valley Area case study, includes a Class I area (Shenandoah National Park) and waterbodies in each of three ecoregions (8.4.1, 8.4.4 and 8.3.1). The number of waterbody sites with CLs available in the NCLD for this study area (4977) is nearly an order of magnitude greater than the total for the four other areas combined (524). In the Shenandoah Valley Area, 70% of the waterbodies are estimated to be able to achieve an ANC at or above 20 µeq/L when annual average S deposition is at or below 9.4 kg/ha-yr; the comparable value for 90% of the waterbodies in this case study area is 7.1 kg/ha-yr. The 70^{th} percentile for achieving an ANC at or above 50 µeq/L is 4 kg/ha-yr (Table 5-6). The S deposition estimates for the 70^{th} and 90^{th} percentile of waterbody CLs for the other, less-well-studied case study areas, for which there are appreciably fewer waterbody sites for which modeling has been performed to estimate CLs (and accordingly greater uncertainty), were consistently lower. Yet, the case study area averages of waterbody CLs for achieving ANC at or above each of the three targets (20, 30 or 50 µeq/L) is quite similar across the five case studies, ranging from 9.4 kg/ha-yr for an ANC of 50 µeq/L in Shenandoah Valley Area to 12 kg/ha-yr for an ANC of 20 µeq/L in both Shenandoah and Sierra Nevada Mtns case study areas (Table 5-6).

The ecoregion analyses focused on 25 ecoregions (18 East and 7 West), nearly all of which are considered acid sensitive. Based on waterbody-specific deposition and CL estimates percentages of waterbodies per ecoregion expected to achieve each of the three ANC targets were derived for five deposition time periods from 2001-03 to 2018-20. The ecoregion-specific information has then been summarized in two different ways: (1) in terms of ecoregion median deposition regardless of time period or ecoregion (ecoregion-time period combinations), and (2) in terms of temporal trends in S deposition and waterbody percentages achieving ANC targets.

The first summarization approach relies on the dataset of 125 pairs of ecoregion median S deposition and percentages of waterbodies estimated to achieve ANC at or above one of the three ANC targets based on waterbody-specific deposition estimates. This dataset is compiled from estimates for the five time periods from 2001-03 to 2018-20 and 25 ecoregions (18 East and 7

West), as described in section 5.1.3.2 above. The ecoregion-time period combinations (totaling 90 for the 18 eastern ecoregions) were distributed into bins distinguished by the maximum ecoregion median deposition in the grouping (e.g., ≤15 kg S/ha-yr, ≤10 kg S/ha-yr, ≤5 kg S/ha-yr). In recognition of the increased uncertainty associated with analyses relying on a smaller portion of the full dataset, we focused primarily on the results for the deposition bins representing half or more of the full dataset (which were those in which the highest ecoregion median included is at least 5 kg/ha-yr). Based on this organization, the estimates for the eastern ecoregions indicate that for ecoregion median S deposition at or below 18 kg/ha-yr, at least 90% of waterbodies per ecoregion were estimated to achieve ANC at or above 20 µeq/L in only 73% of the ecoregion-time period combinations (80% of waterbodies per ecoregion in 83% of combinations), and at or above 50 µeq/L in only 60% of the combinations (Tables 5-5 and 7-1). This summary contrasts with that for the 76 combinations in the bin for S deposition at or below 11 kg/ha-yr, for which at least 90% of waterbodies per ecoregion were estimated to achieve ANC at or above 20 µeq/L in 83% of the combinations, and with the bin for at or below 9 kg/ha-yr, for which at least 80% of waterbodies per ecoregion were estimated to achieve ANC at or above 20 µeq/L in all of the combinations in that bin (Tables 5-5 and 7-1).

As shown in Table 7-1,[4] results for ecoregion median deposition at or below 11 kg/ha-yr (and for the bins for lower values) in eastern ecoregions indicate the likelihood of appreciably more waterbodies achieving the acid buffering capacity targets compared to that estimated for the set of ecoregion-time periods reflecting deposition estimates up to 18 kg/ha-yr. More specifically, this reflects an appreciably greater number of waterbodies in more ecoregions achieving ANC at or above 20 µeq/L, at or above 30 µeq/L and also at or above 50 µeq/L (Table 7-1). Additionally, these percentages increase across the bins for the lower deposition estimates, although while also based on smaller proportions of the supporting dataset (i.e., fewer ecoregion-time period combinations in each subsequently lower deposition bin). For example, for the 69 combinations for S deposition at or below 9 kg/ha-yr, at least 90% of waterbodies per ecoregion were estimated to achieve an ANC at or above 20 µeq/L in 87% of the combinations, and at or above 50 µeq/L in 72% of the combinations (Table 7-1). Although fewer ecoregion-time period combinations are associated with still lower S deposition estimates, contributing to increased uncertainty, we also note that for the 63 ecoregion-time periods for which S deposition is estimated at or below 7 kg/ha-yr, at least 90% of waterbodies per ecoregion were estimated to achieve an ANC at or above 20 µeq/L in 92% of the combinations, and at or above 50 µeq/L in 78% of the combinations (Table 7-1). Lastly, for the lowest bin that comprises at least half of the

[4] Table 7-1 summarizes aspects of the more detailed results presented in Table 5-5 for the 90 eastern ecoregion-time period combinations.

full eastern ecoregion dataset (51 ecoregion-time periods with S deposition estimates at or below 5 kg/ha-yr), 90% of waterbodies per ecoregion were estimated to achieve an ANC at or above 20 µeq/L in 96% of the combinations, and at or above 50 µeq/L in 82% of the combinations.

Table 7-1. Summary of the eastern ecoregion and time period combinations achieving different ANC targets with estimated S deposition at or below different values.

S deposition (kg/ha-yr)*	% of combinations included	% of Eastern ecoregion-time period combinations** with at least 90%, 80% or 70% waterbodies per ecoregion achieving ANC target								
		≥90% of waterbodies			≥80% of waterbodies			≥70% of waterbodies		
ANC (µeq/L) at/below:		*20*	*30*	*50*	*20*	*30*	*50*	*20*	*30*	*50*
≤18	100%	73%	67%	60%	88%	87%	81%	92%	90%	89%
≤13	90%	80%	73%	65%	95%	94%	88%	98%	96%	96%
≤11	84%	83%	76%	68%	97%	96%	91%	99%	99%	99%
≤9	77%	87%	81%	72%	100%	99%	93%	100%	100%	100%
≤7	70%	92%	87%	78%	100%	100%	95%	100%	100%	100%
≤6	66%	93%	88%	78%	100%	100%	97%	100%	100%	100%
≤5	57%	96%	92%	82%	100%	100%	96%	100%	100%	100%

* These values are ecoregion median estimates across all waterbody sites in an ecoregion with a CL estimate.
** These percentages are drawn from the more extensive presentation of results in Table 5-5.

We turn now to consideration of the quantitative acidification risk information from a temporal perspective. Given the decreasing temporal trend in S deposition across all ecoregions (section 6.2.1), we also consider the aquatic acidification results at the ecoregion scale across the 20 years represented by the five time periods (2001-03, 2006-08, 2010-12, 2014-16, 2018-20). With regard to percentages of waterbodies per ecoregion estimated to achieve the three ANC targets, an appreciable improvement is observed for the latter three time periods compared to the initial two time periods. By the 2010-2012 time period, more than 70% of waterbodies in all 25 ecoregions are estimated to achieve an ANC at or above 50 µeq/L and at least 85% are able to achieve an ANC at or above 20 µeq/L. By the 2014-2016 period, the percentages are 85% and nearly 90%, respectively. The median deposition for the CL sites in each of the 18 eastern ecoregions during the latter three time periods range from 1.3 kg S/ha-yr to 7.3 kg S/ha-yr (Table 7-2 and Figure 7-2).

As seen in Table 7-2, with each reduction in S deposition in each subsequent time period, more waterbodies in each of the eastern ecoregions are estimated to be able to achieve the ANC targets. Nearly 90% of the 18 eastern ecoregions are estimated to have at least 90% of their waterbodies achieving an ANC of 20 µeq/L in the 2010-12 period and achieving an ANC of 50 µeq/L in the 2014-16 period. When the 7 western ecoregions are included in a summary based on

ANC targets of 20 µeq/L for the West and 50 µeq/L for the East,[5] over 70% of the full set of ecoregions are estimated to have at least 90% of their waterbodies achieving the ANC targets by the 2010-12 period. More than 90% of the ecoregions are estimated to have at least 90% of their waterbodies achieving the ANC targets by the 2014-16 period (Table 7-2).

Table 7-2. Ecoregions estimated to have different percentages of waterbodies achieving different ANC targets for the five deposition periods analyzed.

Time period	% (n) of ecoregions with specified percentage of waterbodies per ecoregion achieving specified ANC											
	ANC:		20 µeq/L			30 µeq/L			50 µeq/L			
	Ecoregion median S deposition (kg/ha-yr)		Percent of waterbodies per ecoregion			Percent of waterbodies per ecoregion			Percent of waterbodies per ecoregion			
	Min	Max	90%	80%	70%	90%	80%	70%	90%	80%	70%	
	East		---------- Of 18 Eastern Ecoregions ----------									
2001-03	4.0	17.3	39% (7)	67% (12)	72% (13)	28% (5)	61% (11)	72% (13)	22% (4)	50% (9)	72% (13)	
2006-08	3.1	14.4	44% (8)	72% (13)	89% (16)	33% (6)	72% (13)	78% (14)	33% (6)	67% (12)	72% (13)	
2010-12	2.3	7.3	89% (16)	100% (18)	100% (18)	83% (15)	100% (18)	100% (18)	61% (11)	89% (16)	100% (18)	
2014-16	1.9	4.6	94% (17)	100% (18)	100% (18)	94% (17)	100% (18)	100% (18)	89% (16)	100% (18)	100% (18)	
2018-20	1.3	3.9	100% (18)	100% (18)	100% (18)	94% (17)	100% (18)	100% (18)	94% (17)	100% (18)	100% (18)	
	All		---------- Of 25 Ecoregions (18 East, 7 West) ----------									
2001-03	1.2	17.3	56% (14)	76% (19)	80% (20)	48% (12)	72% (18)	80% (20)	44% (11)	64% (16)	80% (20)	
2006-08	1.2	14.4	60% (15)	80% (20)	92% (23)	52% (13)	80% (20)	84% (21)	52% (13)	76% (19)	80% (20)	
2010-12	1.0	7.3	92% (23)	100% (25)	100% (25)	88% (22)	100% (25)	100% (25)	72% (18)	92% (23)	100% (25)	
2014-16	1.1	4.6	96% (24)	100% (25)	100% (25)	96% (24)	100% (25)	100% (25)	92% (23)	100% (25)	100% (25)	
2018-20	0.62	3.9	100% (25)	100% (25)	100% (25)	96% (24)	100% (25)	100% (25)	96% (24)	100% (25)	100% (25)	

Note: Estimates for ANC of 50 µeq/L (East) and 20 µeq/L (West) are identical to those for 50 in all 25 ecoregions.

[5] This combination of targets recognizes the naturally and typically low ANC levels observed in western waterbodies while also including a higher target for the East, as described in section 5.1.2.2.

The temporal trends in percentage of waterbodies estimated to achieve the target ANC levels for each of the 25 individual ecoregions (mapped in Figure 5-13 above) document a large difference between the time periods prior to 2010 and subsequent time periods (Figure 7-1). For the S deposition estimated for the 2010-2012 time period, more than 70% of waterbodies are estimated to be able to achieve an ANC of 50 ueq/L in all 25 ecoregions (Figure 7-1, left panel), and 85% to 100% of waterbodies in all ecoregions are estimated to be able to achieve an ANC of 20 ueq/L (Figure 7-1, right panel).

Given the dependency of the ANC estimates on S deposition estimates, this distinction between the period prior to 2010 and the subsequent decade is also seen in the ecoregion deposition estimates (Figure 7-2). The distribution of deposition estimates at waterbody sites assessed in each ecoregion, and particularly the pattern for the higher percentile sites, as presented in Figure 7-2, illustrate the deposition estimates that are driving the REA estimates. For example, among the 25 East and West ecoregions during the two periods prior to 2010, the medians of the ecoregion 90^{th} percentile deposition estimates ranged from approximately 14 to 17 kg/ha-yr, with maximum values above 20 kg/ha-yr. This contrasts with the deposition estimates during the 2010-2020 period when, among all 25 ecoregions, the medians of the ecoregion 90^{th} percentile deposition estimates ranged from approximately 2 to 5 kg/ha-yr, with all ecoregion 90^{th} percentile estimates below 8 kg/ha-yr. The contrast is much less sharp for the ecoregion medians, as the median is a statistic much less influenced by changes in the magnitude of values at the upper end of the distribution (Figure 7-2).

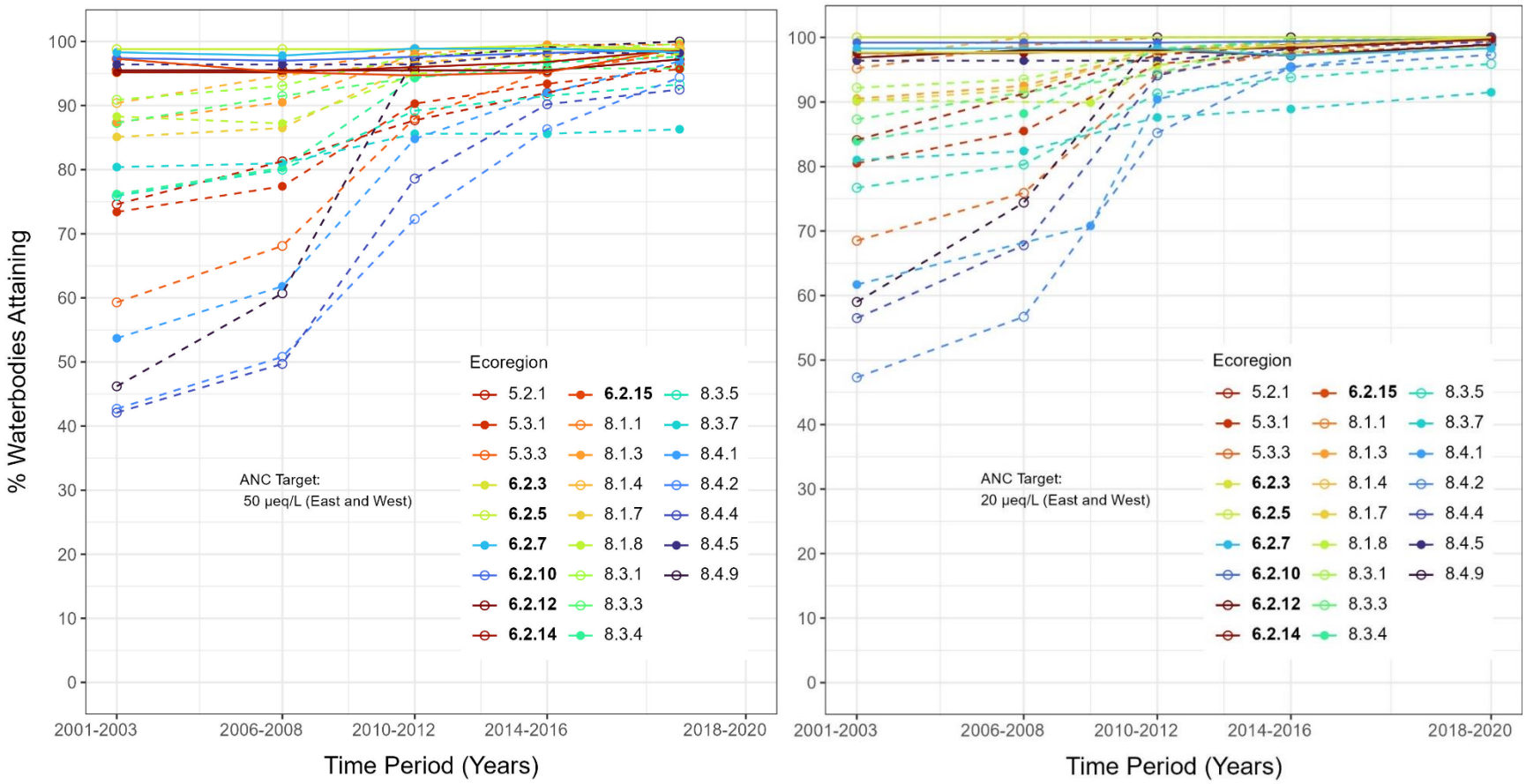

Figure 7-1. Percent of waterbodies per ecoregion estimated to achieve ANC at or above 50 µeq/L (left panel) or 20 µeq/L (right panel). Western ecoregions in bold font and solid lines (*versus* regular font and dashed lines for Eastern ecoregions).

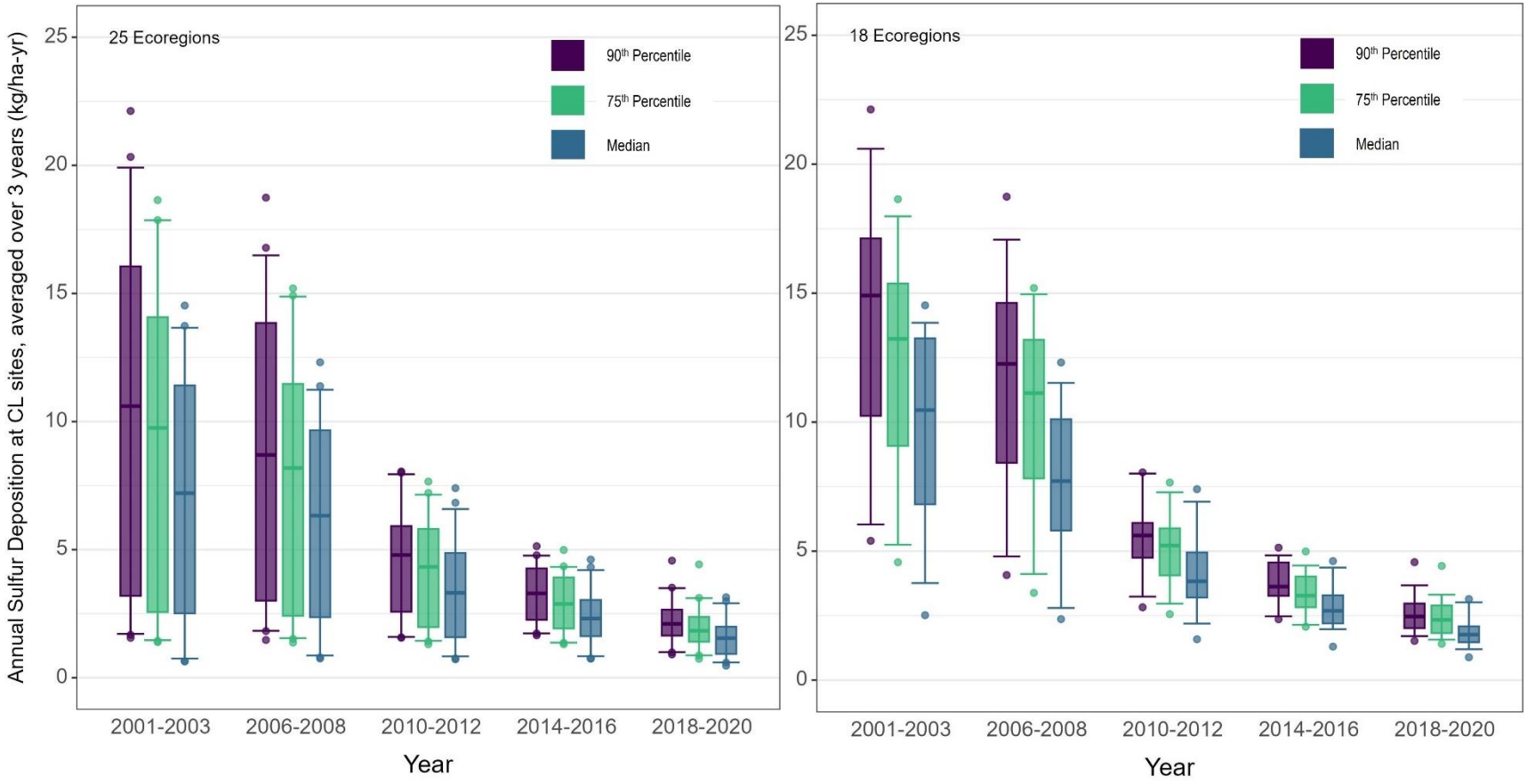

Figure 7-2. Ecoregion 90th, 75th and 50th percentile S deposition estimates at REA waterbody sites summarized for all 25 ecoregions (left) and the 18 eastern ecoregions (right).

In summary, the array of CL-based analyses provides a general sense of the ANC values that waterbodies in sensitive regions across the continental U.S. may be able to achieve, including for areas heavily affected by a long history of acidifying deposition, such as waterbodies in Shenandoah Valley. In the case study for that well studied area (4977 sites in three different ecoregions), 90% of waterbody sites are estimated to be able to achieve an ANC at or above 20 µeq/L (focusing on S deposition only) with S deposition of 7.1 kg/ha-yr and 70% with S deposition of 9.4 kg/ha-yr. For an ANC target at or above 50 µeq/L in the Shenandoah Valley case study, the corresponding deposition estimates are 4.1 and 6.3 kg/ha-yr. These estimates are somewhat similar to the findings of the ecoregion analysis. For example, in that analysis, at least 90% of waterbody sites in 87% of the eastern ecoregion-time period combinations are estimated to be able to achieve an ANC at or above 20 µeq/L with ecoregion median S deposition at or below 9 kg/ha-yr and in 96% of those combinations for S deposition at or below 5 kg/ha-yr. Further, 70% of waterbody sites in all 18 eastern ecoregions are estimated to achieve an ANC at or above 50 µeq/L with ecoregion median S deposition at or below 9 kg/ha-yr. Lastly, consideration of the temporal trend indicates that during the latter half of the 20-year period analyzed (i.e., by the 2010-2012 period), by which time all 25 ecoregions were estimated to have more than 70% of waterbodies able to achieve an ANC at/above 50 µeq/L (and at least 85% able to achieve an ANC at/above 20 µeq/L), median deposition in 95% of the ecoregions was below 8 kg S/ha-yr. By the 2014-2016 and 2018-2020 periods, 24 of the 25 ecoregions were estimated to have more than 90% of waterbodies able to achieve an ANC at/above 50 µeq/L, and median S deposition in all 25 ecoregions was below 5 kg/ha-yr (Figures 7-1 and 7-2).

In considering identification of S deposition levels that may be associated with a desired level of ecosystem protection for a secondary standard, we take note of the implications of the temporal trend in estimated water quality improvements indicated by the increased percentages of waterbodies estimated to achieve more protective ANC levels. The pattern of estimated improving water quality is paralleled by the pattern of declining deposition over the 20-year study period, which is more clear in the upper percentiles of the distribution of values per ecoregion (Figure 7-2). This pattern indicates appreciable difference between the first and second decades of the period in terms of S deposition (at upper percentiles as well as at the median of sites within the 25 ecoregions) and associated aquatic acidification risk. The ecoregion with the highest S deposition in the latter decade had 90th percentile estimates ranging from approximately 8 kg/ha-yr to just below 5 kg/ha-yr (and median estimates with a very similar range) across this decade (Figure 7-2). As noted immediately above, the risk estimates associated with the deposition estimates of this decade indicate generally high percentages of waterbodies per ecoregion as able to achieve or exceed the three ANC targets. Similarly, the ecoregion-time

period binning summary also indicates generally high percentages of waterbodies for ecoregion median S deposition at or below about 8 or 9 kg/ha-yr (Table 7-1). Lastly, the case study CL estimates also indicate appreciable portions of the case study areas that might be expected to attain the 3 ANC targets with deposition below 9 kg/ha-yr. Thus, a range of S deposition, on an areawide basis, that falls below approximately 10-5 kg/ha-yr, or 8-5 kg/ha-yr, appears to be associated with potential to achieve acid buffering capacity levels of interest in appreciable portion of sensitive areas.

- **What does the available information indicate for considering the potential public welfare protection from S deposition-related effects in aquatic ecosystems?**

As an initial matter, we note the integral role of watersheds in aquatic ecosystem health. In so doing, we also recognize the effects of acidic deposition on forested areas that are distinct from effects in water bodies. As recognized in section 4.5 above, given the array of benefits of forested areas to the public, there are public welfare implications of acidifying deposition effects on the natural resources in these areas, with the public welfare significance dependent on the severity and extent of such effects. In light of the more extensive quantitative analyses for aquatic acidification in this review, we focus particularly on the public welfare implications of S deposition-related effects in aquatic ecosystems recognizing their relevance to decision-making in this review.

As recognized in the 2012 review, aquatic ecosystems provide a number of services important to the public welfare, ranging from recreational and commercial fisheries to recreational activities engaged in by the public (77 FR 20232, April 3, 2012). As summarized briefly in section 4.5 above, because aquatic acidification primarily affects the diversity and abundance of aquatic biota, it also affects the ecosystem services that are derived from the fish and other aquatic life found in these surface waters (section 4.5). Fresh surface waters support several cultural services, such as aesthetic and educational services; the type of service that is likely to be most widely and significantly affected by aquatic acidification is recreational fishing, with associated economic and other benefits. Other potentially affected services include provision of food for some recreational and subsistence fishers and for other consumers, as well as non-use services, including existence (protection and preservation with no expectation of direct use) and bequest values (section 4.5).

In light of the considerations above, we recognize that some level of S deposition and associated risk of aquatic acidification, including those associated with past decades of acidifying deposition in the Northeast, can impact the public welfare and thus might reasonably be judged adverse to the public welfare. Depending on magnitude and the associated impacts, there are many locations in which S deposition and associated aquatic acidification can adversely affect the public welfare. For example, there is evidence in some waterbodies, as summarized in

section 5.1.1 above, that aquatic acidification resulting in reduced acid buffering capacity can adversely affect waterbodies and associated fisheries, which in addition to any commercial ramifications, can also have ramifications on recreational enjoyment of affected areas. The evidence is less clear as to what level of risk to an aquatic system, in terms of estimates for achieving various ANC targets across sites within an ecoregion, that might be judged of public welfare significance.

In other secondary NAAQS reviews, the EPA's consideration of the public welfare significance of the associated effects has recognized a particular importance of Class I areas and other similarly protected areas. Accordingly, we note that waterbodies that have been most affected by acidic deposition are in the eastern U.S. and include several Class I areas and a number of other national and state parks and forests (section 5.1.2.1).[6] Such areas were among two of the case studies in the aquatic acidification REA (section 5.1.3.3 above). Thus, while assuring continued improvement of affected waterbodies throughout the U.S. (e.g., through lower S deposition than the levels of the past) may reasonably be considered to be of public welfare importance, such assurance in Class I and similarly protected areas would seem to be of particular importance.

For the purposes of considering the potential public welfare significance of aquatic acidification effects of differing levels of S deposition, we take note of the approach taken in Appendix 5A and section 5.1 to summarize the REA ecoregion-scale results, i.e., in terms of percentages of ecoregions in which differing percentages of waterbodies are estimated to achieve the three acid buffering capacity targets. The presentations above are summarized in such a way in identifying a range of S deposition less than approximately 10 to 5 kg/ha-yr or 8 to 5 kg/ha-yr that may be appropriate to consider for potential alternative standard options in light of REA estimates for achieving the three acid buffering capacity targets. In considering the question below with regard to terrestrial acidification, we also focus on consideration of quantitative information with a similar objective in mind.

- **What does the quantitative information regarding S deposition and terrestrial acidification indicate regarding deposition levels of relatively greater and lesser concern as to the potential for acidification-related effects? What are associated uncertainties?**

As recognized in Chapter 5, the quantitative tools for characterizing waterbody response to acidic deposition are well established and/or have been extensively applied in a greater variety of locations. Further, there is appreciable availability of site-specific water quality measurements in sensitive areas across the U.S. The available quantitative information related to terrestrial

[6] A comparison of Figures 4-4 and 5-6 indicates multiple Class I areas in ecoregions considered acid sensitive.

acidification summarized in Chapter 5 (and presented in more detail in Appendix 5B) includes discussion of soil chemistry modeling analyses (both those described in published studies and an analysis performed in the 2009 REA), studies involving experimental additions of S compounds to defined forestry plots, and observational studies of potential relationships between terrestrial biota assessments and metrics for S deposition (section 5.3). We consider each here in consideration of the questions posed above.

With regard to soil chemistry modeling, we note first the quantitative analyses, performed in the last review, of soil acidification in areas of the northeastern U.S. in which two sensitive tree species, sugar maple and red spruce, are widely distributed. These analyses yielded estimates of acidic deposition CLs associated with three different values for a well-studied indicator of soil acidification, BC:Al ratio[7] (2009 REA, section 4.3). These estimates indicated a range of annual deposition rates (under which ratios were at or above the intermediate target value of 1) that were well above the CL estimates associated with achieving various ANC targets in the aquatic acidification analyses discussed above.[8] Thus, a focus on aquatic acidification might reasonably be expected to also provide protection from soil acidification effects on terrestrial biota. As concluded in the 2009 REA, an important source of uncertainty in the simple mass balance model used in the analysis is the soil weathering parameter (as is also the case in water quality modeling). In this context, we additionally note that studies published since the 2009 REA, including one focused on areas of Pennsylvania, have utilized different estimates for this parameter intended to reduce the associated uncertainty and have reported somewhat higher CL estimates when the updated approach is used (as described more fully in section 5.3.2.1).

With regard to the information available from studies involving S additions to experimental forested areas, the number of tree species that have been included in such experiments is somewhat limited. Although limited in number, the more widely recognized sensitive species (based on field observations) have been included in such studies. We note that the available studies have not reported effects on the trees analyzed plots with additions below 20 kg/ha-yr (in addition to the atmospheric deposition occurring during the experiment).

The recently available quantitative information regarding S deposition and terrestrial acidification also includes two observational studies that report associations of tree growth and/or survival metrics with various air quality or S deposition metrics, providing support to conclusions regarding the role of acidic S deposition on tree health in the U.S., most particularly

[7] Given the toxicity of some forms of aluminum in soil solution, the ratio of base cations to aluminum ions in soil (BC:Al ratio) has commonly been used in assessing risk of acidifying deposition to terrestrial systems (ISA, Appendix 4, section 4.3.5 and Appendix 5).

[8] These deposition rates were also above all of the ecoregion estimates (across the five time periods from 2001 through 2020) considered in the aquatic acidification analyses (Table 5-7).

in regions of the eastern U.S. (summarized in section 5.3.2.3 and described more fully in Appendix 5B, section 5B.3.2). The metrics utilized in the two largest studies include site-specific estimates of average SO_4^{2-} deposition and of average total S deposition over the interval between tree measurements, generally on the order of 10 years (Dietze and Moorcroft, 2011; Horn et al., 2018). In the study that used SO_4^{2-} as the indicator of acidic S deposition, and for which the study area was the eastern half of the contiguous U.S., site-specific average SO_4^{2-} deposition (1994-2005) ranged from a minimum of 4 kg/ha-yr to a maximum of 30 kg/ha-yr (Dietze and Moorcroft, 2011). Review of the study area for this study and a map indicating geographic patterns of deposition during the period of the deposition data indicate the lowest deposition areas to be the farthest western, northeastern and southeastern areas of the eastern U.S. (in which S deposition in the 2000-2002 period is estimated to fall below 8 kg/ha-yr), and the highest deposition areas to be a large area extending from New York through the Ohio River valley (Appendix 5B, Figures 5B-1 and 5B-11). In the second study, deposition at the sites with species for which growth or survival was negatively associated with S deposition ranged from a minimum below 5 kg/ha-yr to a site maximum above 40 kg/ha-yr, with medians for these species generally ranging from around 5 to 12 kg/hr-yr (Appendix 5B, sections 5B.2.2.3 and 5B.2.3; Horn et al., 2018).

As discussed in section 5.3.2 and Appendix 5B, the history of appreciable acidic deposition in the eastern U.S., with its associated impacts on soil chemistry, has the potential to be exerting a legacy influence on tree growth and survival more recently. Further, at a national-scale, the geographic deposition patterns (e.g., locations of relatively greater versus relatively lesser deposition) more recently appear to be somewhat similar to those of several decades ago (e.g., sections 2.5.4 and 6.2.1). This similarity in patterns has the potential to influence findings of observational studies that assess associations between variation in tree growth and survival with variation in levels of a metric for recent deposition at the tree locations. This indicates an uncertainty with regard to interpretation of these studies with regard to a specific magnitude of deposition that might be expected to elicit specific tree responses, such as those for which associations have been found. As recognized in the study by Dietze and Moorcroft (2011), which grouped species into plant functional groups, acidification impacts on tree mortality result from cumulative long-term deposition, and patterns reported by their study should be interpreted with that in mind.

7.2.2.3 Relating SO_X Air Quality Metrics to Deposition of S Compounds

Analyses in Chapter 6 examine the relationships between air concentrations, in terms of various air quality metrics (including design values for the current standards), and S deposition in areas near or removed from ambient air monitoring sites. Analyses include air quality metrics

based on S compounds measured in three different monitoring networks. These include the SLAMS network for SO_2 NAAQS surveillance, as well as the IMPROVE network of $PM_{2.5}$ monitors (which report on particulate S compounds) and the CASTNET network that reports measurements of SO_2 and particulate SO_4^{2-}. The latter two networks do not employ FRM/FEMs established for SO_2 or $PM_{2.5}$ NAAQS surveillance and are generally focused on monitoring in rural or remote areas.

While the data from the CASTNET and IMPROVE networks support analyses of S compounds other than SO_2 (i.e., particulate SO_4^{2-} or summed airborne S compounds), the analyses based on data from NAAQS surveillance monitors are particularly relevant given that the current standards are judged using design value metrics based on measurements at existing FRM/FEM monitor locations, which are mostly located in areas of higher pollutant concentrations near emissions sources. For example, many ambient air SO_2 monitors are sited near large point sources of SO_2 (e.g., electric generating units). Accordingly, information from these monitoring sites can help inform an understanding of how changes in SO_2 emissions, reflected in ambient air concentrations, may relate to changes in deposition and, correspondingly, what secondary standard options might best regulate ambient air concentrations such that deposition in areas of interest is maintained at or below certain levels.

Analyses of relationships between S deposition and ambient air concentrations of S compounds were conducted using ecoregion median S deposition and upwind monitoring site concentrations (in the trajectory-based analyses), as well as S deposition in TDep grid cells with ambient air concentrations at SLAMS monitors in the same grid cells, and TDep total S deposition or NADP wet S deposition at Class I area sites of collocated IMPROVE and CASTNET monitors with ambient air concentrations of SO_2, particulate SO_4^{2-} or both in combination. Information is also analyzed from a 21-year CMAQ simulation. Details of these analyses are described in Chapter 6. In addressing the questions below, we consider the findings of those analyses specific to S deposition associated with SO_X and PM in ambient air.

- **What do the information and air quality analyses available in this review indicate regarding relationships between air quality metrics related to the existing standards, and potential alternatives, and S deposition? What are the uncertainties in relationships using such metrics?**

As characterized in the ISA and summarized in Chapters 2 and 6, S deposition has declined appreciably over the past 20 years (e.g., Figure 6-11). This decline tracks closely with the parallel decline in SO_2 emissions, as discussed in sections 2.5.4 and 6.2.1, above. In the more recent years, the areas of relatively higher S deposition estimates are generally within the Ohio River Valley (southeastern Ohio, West Virginia, and western Pennsylvania), the Gulf Coast (Texas and Louisiana), and a few very small areas in North Dakota and northern California

(Figure 6-11). In addition to source emissions, there are many factors contributing to temporal and spatial variability in S deposition, including frequency of precipitation, and micrometeorological factors relevant to the dry deposition velocity. For example, S deposition in arid areas, such as the Southwest, and in areas close to sources of SO_2 emissions tends to be predominantly dry deposition of SO_2, while S deposition more distant from sources, and in less arid areas, tends to be predominantly wet deposition of SO_4^{2-} (ISA, Appendix 2, section 2.6.5). Thus, in areas of the more arid western U.S., where S tends to be low, S may deposit more from SO_2, while in the wetter eastern U.S., S deposition may be more influenced by wet deposition of SO_4^{2-}.

The analyses in Chapter 6 assess SO_2 concentrations using a metric based on the current form and averaging time of the secondary SO_2 NAAQS, which is the second highest 3-hour daily maximum in a year, as well as an annual average SO_2 air quality metric. With regard to the annual average metric, we focused on the annual average SO_2 concentration, averaged over three years. In light of the many factors contributing variability to S deposition, the analyses focus on a 3-year average of all of the air quality and deposition metrics and include multiple years of data, generally on the order of 20 years and covering a period of declining concentrations and deposition. Of the two metrics analyzed (annual average and second maximum annual 3-hour average), we focus primarily on the annual average of SO_2 concentrations, averaged over 3 years, given the greater stability of the metric and our focus on control of long-term S deposition.

The data and analyses presented in Chapter 6 indicate a significant association of S deposition with SO_2 concentrations with statistically significant correlation coefficients ranging from approximately 0.50 up to above 0.70. These include associations of estimated total and measured wet S deposition with annual average SO_2 concentrations (at same location) in 27 Class I areas based on a 21-year CMAQ simulation (1990-2010) or collocated NADP and CASTNET monitors (2000-2019). These associations are also observed for TDep estimated total S deposition with SO_2 concentrations at SLAMS monitors in the same TDep grid cell, as well as for ecoregion median total S deposition based on TDep with SO_2 concentrations at upwind sites of influence monitors (SLAMS) identified by trajectory-based analyses.

At SLAMS monitor locations, the correlation coefficient for S deposition with annual average SO_2 (averaged over three years) has a value of 0.70 for the full dataset across the five time periods, with similar correlations for dry and wet deposition ($r=0.72$ and $r=0.66$, respectively) and an even higher r value (0.79) for the eastern sites. In the complete dataset and the subset of eastern sites, the statistically significant correlation coefficient ranges from 0.52 to 0.72 in the first three time periods (through 2010-2012) and is much reduced in the latter two time periods. Little correlation is observed in the subset of western sites.

In considering the findings of the trajectory-based analyses, we note the somewhat stronger correlations observed for the weighted metric (which provides for proportional weighting of air concentrations from locations projected to contribute more heavily to a particular ecoregion), compared to the maximum EAQM, particularly for the first two to three time periods of the 20-year period. For example, across all sites, the correlation coefficients for the weighted metric range from 0.71 to 0.81 for the first three periods, while the corresponding coefficients for the maximum metric range from 0.28 to 0.69. This difference is related to the extent to which monitor concentrations can be indicative of atmospheric loading. Conceptually, the weighted maximum EAQM is representing the atmospheric loading for the locations (and associated sources) of the contributing (sites of influence) monitors. We note that this metric, however, is not directly translatable to a standard level which is an upper limit on concentrations in individual areas. Conversely, unweighted concentrations (even from the maximum contributing monitor) are limited in the extent to which they can reflect atmospheric loading due to a number of factors, including monitor and source distribution and magnitude of emissions. The lower correlations observed between deposition and the maximum EAQM in areas of lower concentrations are an indication of this complexity. Across a broad enough range in deposition (e.g., as occurring in the earlier time periods and in the East), a rough correlation is observed, which breaks down across smaller ranges in deposition, as evidenced by the much lower r values for the more recent period with its much lower magnitude of deposition and much smaller range in deposition.

In the context of identifying a range of annual average SO_2 EAQM levels that may be associated with an acceptable level of S deposition, such as ecoregion median S deposition of 5-10 kg S/ha-yr, as discussed above, we take note of several important considerations. First, monitor concentrations of SO_2 can vary substantially across the U.S., reflecting the distribution of sources, and other factors such as meteorology, complicating consideration of how the maximum contributing monitor (as identified in the HYSPLIT analysis described in section 6.2.4 above) relates to S deposition levels in downwind ecosystems. Another consideration is the substantial scatter in the relationship between S deposition estimates and measured SO_2 concentrations with ecoregion median S deposition values below 5 kg/ha-yr. This scatter in the relationship between measured SO_2 concentration and S deposition estimates at these lower deposition levels, contributes increased uncertainty to conclusions regarding potential secondary standard SO_2 metric levels intended to relate to ecoregion median deposition levels at or below 5 kg/ha-yr.

In identifying levels for consideration for a potential annual average SO_2 metric, we consider first the SO_2 concentrations at ecoregion sites of influence identified in the trajectory-based analyses (of the 84 ecoregions in CONUS) across different ranges of downwind ecoregion

S deposition estimates. Figure 7-3 presents the pairs of median deposition estimates and associated upwind sites of influence EAQM-max SO_2 concentrations from the trajectory-based analysis in section 6.2.4 above (specifically, the combined datasets presented in Figures 6-40 and 6-41). In this dataset for all 84 ecoregions, the maximum annual average SO_2 concentrations, averaged over three years, at sites of influence to downwind ecoregions with median S deposition ranging below 9 kg S/ha-yr to 6 kg/ha-yr, were all below 15 ppb, and 75% of the monitor sites of influence concentrations were at or below 10 ppb. For ecoregions with median S deposition below 6 kg/ha-yr, EAQM-max SO_2 concentrations at associated sites of influence were all below approximately 10 ppb (Figure 7-3). In considering this presentation, we note that 9-10 kg/ha-yr is the approximate upper end of the range identified for ecoregion median (or areawide) deposition in section 7.2.2.2 above, and 5 kg/ha-yr is the lower end.

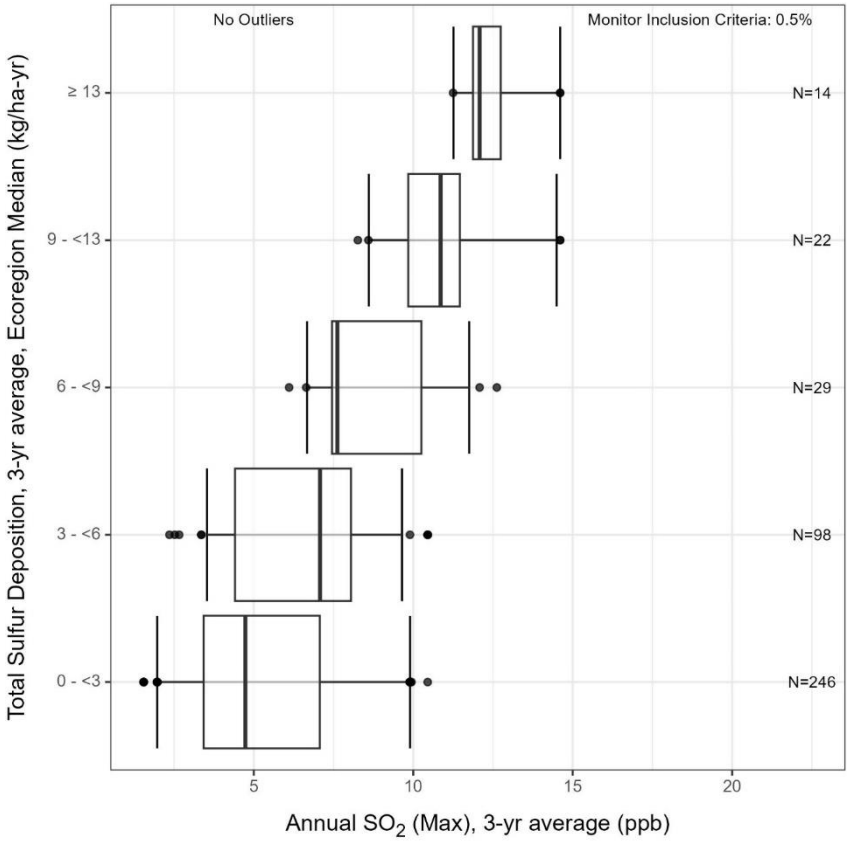

Figure 7-3. Distributions of EAQM -Max annual SO_2 concentrations (3-year average) at ecoregion sites of influence identified in trajectory-based analyses for multiple levels of ecoregion median S deposition (based on zonal statistic) in the five time periods (2001-2003, 2006-2008, 2010-2012, 2014-2016, 2018-20).

In Figure 7-4, we consider a similar presentation focused on the subset of data for the 25 REA ecoregions (with their upwind monitors). This figure presents the distribution of concentrations at maximum upwind monitors with the ecoregion median and 90th percentile

deposition estimates (across the waterbody sites assessed in the REA in each ecoregion). This presentation indicates that for the three highest ecoregion median bins (at or above 9 kg/ha-hr), all of the EAQM-max concentrations are greater than 10 ppb (Figure 7-4, left panel). The next lower bin (for deposition below 9 down to 6 kg/ha-yr), has more than half of the EAQM-max concentrations below 10 ppb. And all of the EAQM-max concentrations associated with ecoregion median deposition in the lowest bins (S deposition below 6 kg/ha-yr) were below 10 ppb. The pattern of declining frequency of EAQM-max concentrations above 10 ppb with lower deposition estimates is also seen for the ecoregion 90th percentile deposition estimates (Figure 7-4, right panel). This pattern suggests that when the highest EAQM-max concentration is somewhat below 15 ppb and down to 10 ppb, the ecoregion median deposition is below 9 kg/ha-yr and the 90th percentile deposition 13 kg/ha-yr. when the highest EAQM-max concentrations is at approximately 11 ppb, or 10 ppb, both the median and 90th percentile deposition are below 9 kg/ha-yr (Figure 7-4).

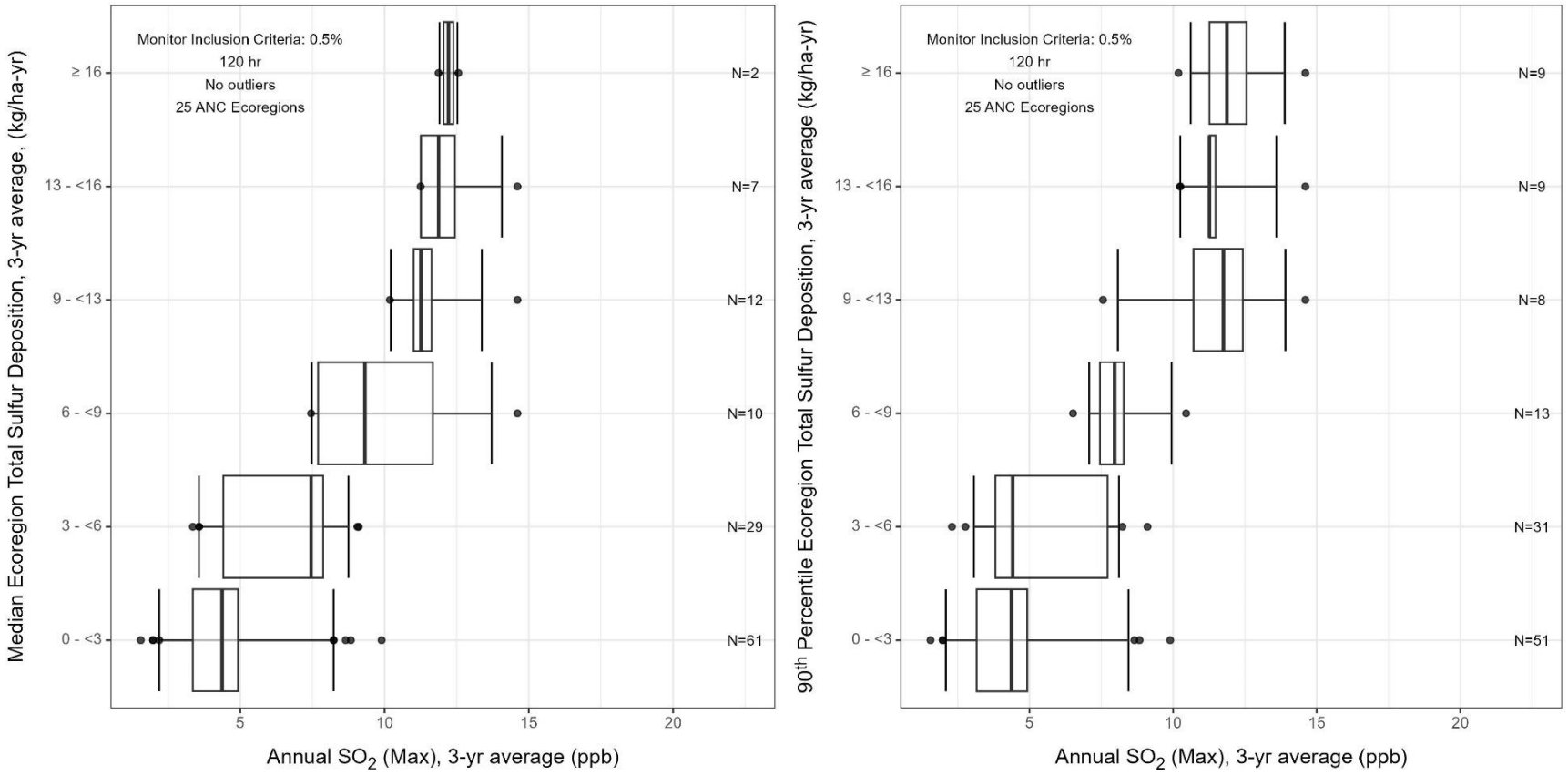

Figure 7-4. Distributions of maximum annual average SO_2 concentrations (3-year average) at ecoregion sites of influence identified in trajectory-based analyses for multiple levels of ecoregion median (left) and 90th percentile (right) S deposition in the 25 REA ecoregions for the five time periods (2001-2003, 2006-2008, 2010-2012, 2014-2016, 2018-20). Ecoregion medians and 90th percentiles derived from TDep estimates at sites with CLs in the ecoregion.

Given the declining trend in S deposition across the five time periods in the aquatic acidification analysis for which there were corresponding estimates of increasing ANC in sensitive ecoregions (as discussed above), we also consider the annual average SO_2 concentration at monitor sites during these same five time periods (Figure 7-5). In so doing, we focus on the most recent time periods analyzed (i.e., since 2010) when, as noted in section 7.2.2.2 above, the REA indicated appreciably improved levels of acid buffering capability in the waterbodies of the 25 analyzed ecoregions in which ANC targets were met or exceeded in a high percentage of water bodies across a high percentage of ecoregions. This presentation indicates that during the most recent time periods (in which ecoregion median S deposition estimates for the 25 REA ecoregions were below 10 kg/ha-yr), the highest 3-year average annual SO_2 concentrations were generally somewhat above 10 ppb (with some exceptions during the 2019-2021 period), and 95% of the concentrations in each of the three most recent periods are just at or below 5 ppb (Figure 7-5, left panel).[9] The distributions of annual average SO_2 concentrations exhibit a similar pattern of concentrations to that for the 3-year averages, suggesting there to be little year-to-year variability in this metric (Figure 7-5).

[9] The outlier annual SO_2 concentration values above 10 ppb during 2019-2021 in Figure 7-5 are at two sites in southern Missouri where the design values for the primary standard are more than three times the level of the primary standard.

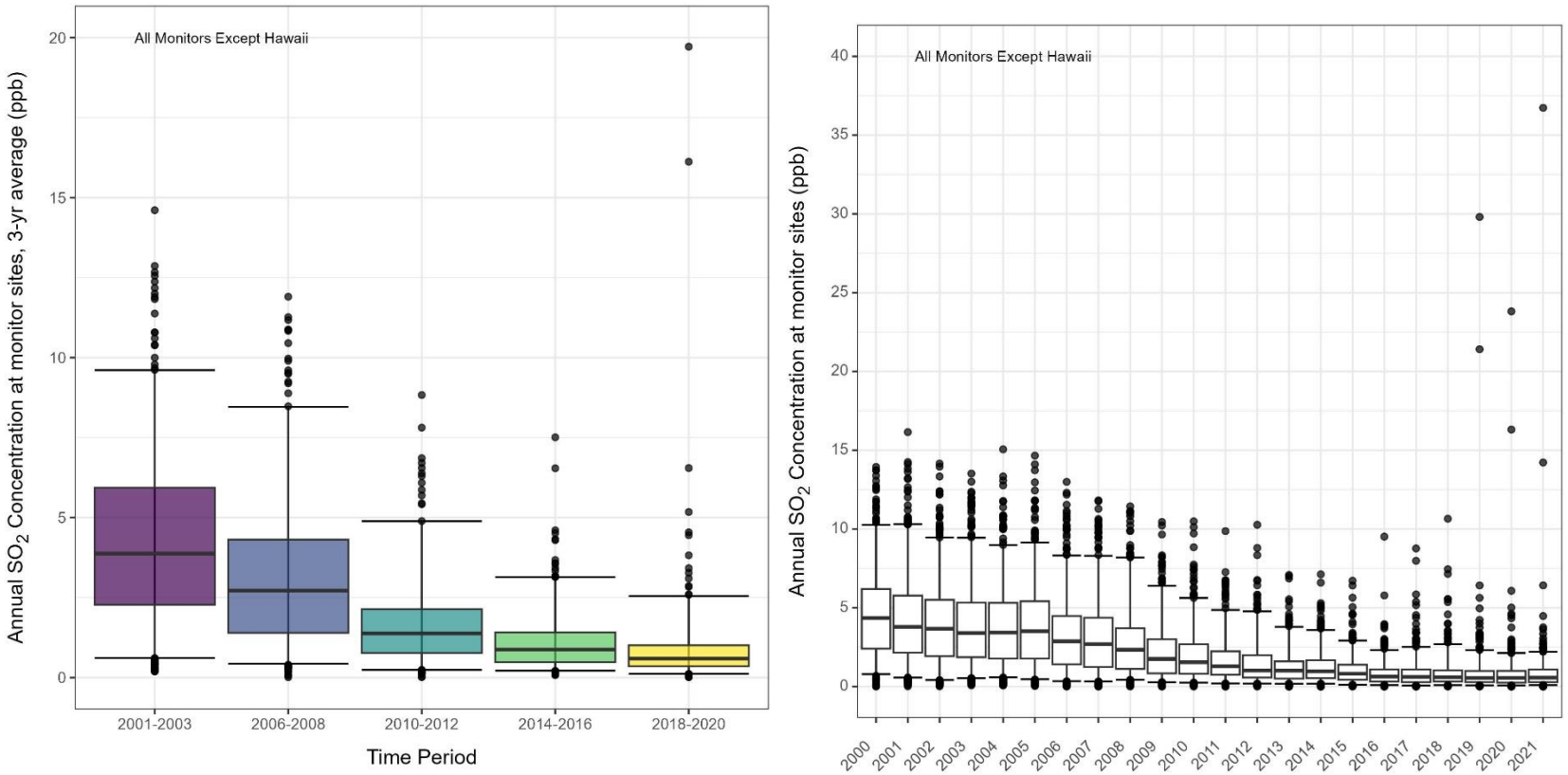

Figure 7-5. Distributions of annual SO_2 concentrations at SLAMS FRM/FEM monitors, averaged across three consecutive years, for the five time periods of the REA (left) and annual averages from 2000 to 2021 (right).

Lastly, although there are significant correlations between SO_2 concentrations and S deposition, there is variability in relationships between SO_2 concentrations at SLAMS monitors and nearby and/or downwind S deposition. This variability relates to the complexity of the atmospheric chemistry, pollutant transport and deposition processes, as summarized in sections 2.1.1 and 2.5 above. There is also uncertainty in these relationships which relates to a number of factors, described more fully in section 6.3 and Table 6-13. These factors include uncertainty in our estimates of S deposition (section 2.5.2)., as well as spatial distribution of monitor sites and their representation of significant SO_2 emissions sources, as well as elements of the trajectory-based analysis, e.g., inclusion criteria for identifying monitoring sites of influence (Table 6-13).These various uncertainties in the data and analyses, and the inherent variability of the physical and chemical processes involved, contribute uncertainty to conclusions concerning ambient air SO_2 concentrations related to S deposition estimates at different scales, although it is unclear, however, how much and in what way each of these uncertainties might impact those conclusions.

In recognition of such uncertainty and variability, REA aquatic acidification analyses and discussion of S deposition levels above have focused on statistics for deposition estimates representing large areas (e.g., ecoregion median and 90^{th} percentile, and case study area average or 70^{th} and 90^{th} percentile CLs). In considering median estimates, however, we have also recognized that it is the higher points on the distribution of deposition estimates within an ecoregion (e.g., 90^{th} percentile) which will contribute most to aquatic acidification risk. In light of this, it is noteworthy, however, that the distribution of S deposition estimates within ecoregions has collapsed in the more recent years of the 20-year analysis period, with 90^{th} percentile estimates falling much close to the medians than in the first decade of the period (Figure 7-2).

- **What do the available information and air quality analyses indicate regarding relationships between air quality metrics based on indicators other than those of the existing standards and S deposition? What are the uncertainties in relationships using such metrics?**

We also assessed relationships between collocated estimates of total S deposition (or wet deposition measurements) and measurements of indicators of atmospheric S-containing pollutants (particulate SO_4^{2-} and the sum of S in SO_2 and particulate SO_4^{2-}) in 27 Class I areas, mostly located in the western U.S (section 6.2.2). The correlations of deposition with the two indicators assessed are moderate and similar for S deposition as NADP wet deposition and as TDep estimates. For example, the correlation coefficients for either total S deposition (TDep) or wet S deposition (NADP) with either SO_4^{2-} in $PM_{2.5}$ (IMPROVE) or total S (CASTNET) in those locations vary from 0.52 to 0.61 (Figures 6-27 and 6-31). These coefficient values, while

somewhat comparable to those for the trajectory-based analysis of downwind deposition and upwind annual average SO_2 concentrations (r=0.49 and 0.56; Table 6-8), they are somewhat lower than the correlation observed between S deposition (TDep) and SO_2 concentrations at SLAMS monitors. For example, for annual average SO_2 concentrations, averaged over three years at SLAMS, the correlation coefficients are 0.70, 0.66 and 0.72 for total, wet and dry S deposition, respectively (Table 6-4).

The analyses for these Class I area sites also indicate poor correlation of total S deposition (TDep) with annual average IMPROVE $PM_{2.5}$ (r=0.33, Figure 6-31). This is not dissimilar to the correlations observed for ecoregion S deposition estimates with annual average $PM_{2.5}$ (3-year average) at upwind sites of influence from the trajectory-based analysis (r=0.22 and 0.48, Table 6-12). While the correlations in this for deposition in eastern ecoregions were much higher (r=0.83 and 0.90), the coefficients were negative for deposition in western ecoregions. The fact that most of the Class I area sites are in the West (20 of the 27 sites) may be an influence on the low correlation observed for that dataset.

In summary, the analyses involving total S deposition and ambient air SO_4^{2-} concentrations are at remote Class I area locations, distant from sources of SO_2 emissions, and the relationship of SO_4^{2-} with S deposition is no stronger than that for SO_2 at SLAMS, which are near sources and which monitor SO_2 (the source for atmospheric SO_4^{2-}). As a result, we find that this analysis does not indicate a clear advantage for an indicator based on SO_4^{2-}measurements (or SO_4^{2-} and SO_2 combined), such as is currently collected at CASTNET sites, over options for a potential annual average standard metric focused on SO_2 concentrations (based on FRM/FEMs), as discussed above. It is also of note that use of SO_4^{2-} measurements, alone or in combination with SO_2 concentrations, as an indicator for a new standard would entail development of sample collection and analysis FRM/FEMs and of a surveillance network.

7.2.3 N Deposition and N Oxides and PM

To inform conclusions in this review related to the N oxides and PM secondary standards, we consider the information supporting quantitative evaluation of the linkages between N oxides and PM in ambient air with N deposition and associated ecological effects. In considering the questions below, we draw on the available welfare effects evidence described in the current ISA, the 2008 ISA for oxides of N and S, the 2009 ISA for PM, and past AQCDs for all three pollutants, and summarized in Chapter 4 above. We do this in combination with the available quantitative information summarized in chapters 5 and 6 above.

7.2.3.1 Quantitative Information for Ecosystem Risks Associated with N Deposition

The currently available evidence, including that previously available, documents aquatic and terrestrial effects of N deposition, as summarized in Chapter 4 and described in detail in the

ISA. As recognized in sections 7.2.1 and 7.2.2.1 above, N deposition has played a role in acidic deposition in both terrestrial and aquatic ecosystems and associated effects in the U.S., although analyses in the aquatic acidification REA indicate a reduced role for N deposition in the time period analyzed (section 5.1.2.4 and Appendix 5A, section 5A.2.1). Additionally, the evidence is extensive and longstanding as to the role of N loading of waterbodies and associated eutrophication. Further, the evidence previously available, with noteworthy additions from the more recently available evidence, describes the role of N deposition in terrestrial N enrichment and associated ecosystem effects.

We consider here the available information that quantitatively relates atmospheric deposition of N to effects on soil and surface water chemistry and relates those effects to specific ecological effects for the different types of ecosystems and categories of effects. Our focus with regard to N deposition is on N enrichment-related effects in light of the relatively greater role played by S in acidic deposition, in recent years (section 5.1.2.4 above). In focusing on N enrichment-related effects, we note the varying directionality of some of these effects, particularly in terrestrial ecosystems, such that the effects of N enrichment can in particular ecosystems and for particular species, seem beneficial (e.g., to growth or survival of those species), although in a multispecies system, effects are more complex with potential for alteration of community composition. Our consideration below of the availability of quantitative information relating atmospheric N deposition to N enrichment-related effects in aquatic and terrestrial ecosystems is in the context of the following question.

- **What does the available evidence base indicate regarding air quality and atmospheric deposition and risk or likelihood of occurrence of ecosystem effects under differing conditions? What are limitations and associated uncertainties in this evidence?**

With regard to acidification-related effects of N deposition, we recognize the approaches and tools referenced in section 7.2.2.1 above could be utilized for S and N deposition in combination, but we focused the analysis of aquatic ecosystem acidification summarized in section 5.1 above on S deposition, based on analyses indicating the relatively greater role of S deposition under the more recent air quality conditions (section 5.1.1.4). Discussion of analyses relating acidifying deposition to terrestrial acidification indicators is presented in section 5.3 above.

In evaluating the available information for the purposes of quantitatively relating N deposition associated with N oxides and PM to waterbody responses (most particularly waterbody eutrophication), we first take note of the appreciable evidence base documenting assessments of N loading to waterbodies across the U.S. (ISA, Appendix 7). In so doing, we note the waterbody-specific nature of such responses and the relative role played by atmospheric

deposition, among other N sources. For example, as recognized in the ISA and Chapters 4 and 5 above, the relative contribution to such loading from atmospheric deposition compared to other sources (e.g., agricultural runoff and wastewater discharges), which varies among waterbody types and locations, can be a complicating factor in quantitative analyses. Additionally, characteristics of resident biota populations and other environmental factors are influential in waterbody responses to N loading (e.g., temperature, organic microbial community structure, aquatic habitat type, among others), as discussed in the ISA (ISA, Appendix 7).

Based on identification of eutrophication as a factor in impacts on important fisheries in some estuaries across the U.S., multiple government and nongovernment organizations have engaged in research and water quality management activities over the past multiple decades in large and small estuaries and coastal waters across the U.S. These activities have generally involved quantitative modeling of relationships between N loading and water quality parameters such as dissolved oxygen (ISA, Appendix 7, section 7.2). As summarized in section 5.2.3 above, this research documents both the impacts of N enrichment in these waterbodies and the relationships between effects on waterbody biota, ecosystem processes and functions, and N loading. The evidence base recognizes N loading to have contributions from multiple types of sources to these large waterbodies, and their associated watersheds, including surface and ground water discharges, as well as atmospheric deposition. Accordingly, loading targets or reduction targets identified for these systems have generally been identified in light of policy and management considerations related to the different source types, as discussed further in section 7.2.3.2 below.

Focused assessments in freshwater lakes, including alpine lakes, where atmospheric deposition may be the dominant or only source of N loading, also provide evidence linking N loading with seemingly subtle changes, as summarized in section 5.2.2. above. Such changes include with regard to whether P or N is the limiting nutrient and shifts in phytoplankton community composition, for which public welfare implications are less clear. This evidence has included observational studies of freshwater lakes of the western U.S. involving statistical modeling, and studies which have utilized NO_3^- concentrations as an indicator of N enrichment (e.g., ISA, Appendix 9). Among the recent evidence in the ISA are long-term monitoring studies of lakes in several mountainous regions, including the Appalachian Mountains, the Adirondacks, and the Rocky Mountains, that have documented reduced surface water NO_3^- concentrations corresponding to decreases in atmospheric N deposition since the 1980s and 1990s (ISA, Appendix 7, section 7.1.5.1).

An additional type of aquatic ecosystem effect recognized in the available evidence for N loading, particularly to freshwaters, relates to an increase in the toxicity of exudates associated with harmful algal blooms (ISA, Appendix 9, section 9.2.6.1). Information available in this

review indicates that growth of some harmful algal species, including those that produce microcystin, are favored by increased availability of N and its availability in dissolved inorganic form (ISA, Appendix 9, p. 9-28). Although this is an active research area, few if any datasets are currently available that quantitatively relate N loading to risk of harmful blooms, including those that may distinguish roles for different deposition components such as deposition of oxidized N or of particulate reduced N distinguished from that of N loading via dry deposition of reduced N.

With regard to terrestrial ecosystems and effects on trees and other plants, we recognize the complexity, referenced above, that poses challenges to approaches for simulating terrestrial ecosystem responses to N deposition across areas diverse in geography, geology, native vegetation, deposition history, and site-specific aspects of other environmental characteristics. In general, limitations particular to the different types of quantitative analyses contribute associated uncertainty to our interpretations. Uncertainties associated with the soil acidification modeling analyses include uncertainties associated with the limited dataset of laboratory-generated data on which the BC:Al targets are based, as well as the steady-state modeling parameters, most prominently those related to base cation weathering and acid-neutralizing capacity (section 5.3.4.1). Uncertainties associated with experimental addition analyses include the extent to which the studies reflect steady-state conditions, with a related limitation of some of these studies associated with a lack of information regarding historic deposition at the study locations that might inform an understanding of the prior issue (section 5.3.4.1). Several aspects of observational or gradient studies of tree growth and survival (or of species richness for herbs, shrubs and lichens) contribute uncertainties to identification of deposition levels of potential concern for tree species effects, including unaccounted-for factors with potential influence on tree growth and survival (e.g., ozone and soil characteristics), as well as the extent to which associations may reflect the influence of historical deposition patterns and associated impact. Thus, while the evidence is robust as to the ability for N loading from deposition to contribute to changes in plant growth and survival, and associated alterations in terrestrial plant communities, there are a variety of factors, including the history of deposition and variability of response across the landscape, that complicate our ability to quantitatively relate specific N deposition rates, associated with various air quality conditions, to N enrichment-related risks of harm to forests and other plant communities in areas across the U.S. (section 5.3.4).

7.2.3.2 General Approach for Considering Public Welfare Protection

As an initial matter, we note that the effects of acidification on plant growth and survival, at the individual level, are generally directionally harmful, including reduced growth and survival. In contrast, the effects of N enrichment can, in particular ecosystems and for particular species, be beneficial or harmful (e.g., to growth or survival of those species). Accordingly, there

is added complexity to risk management policy decisions for this category of effects, including the lack of established risk management targets or objectives, particularly in light of historical deposition and its associated effects that have influenced the current status of terrestrial ecosystems, their biota, structure and function.

Further, we recognize the contribution to N deposition of atmospheric pollutants other than the criteria pollutants N oxides and PM, most significantly the contribution of NH_3 (as described in section 6.2.1 above). This contribution has increased since the last reviews of the NO_2 and PM secondary standards, as seen in Figures 6-17 and 6-18, reflecting increases in NH_3 emissions over that time period. These trends of increased NH_3 emissions and reduced N deposition coincide with decreasing trends in N oxides emissions and associated contributions of oxidized N to total N deposition (Figures 6-3 and 6-19). The TDep estimates of different types of N being deposited at the 92 CASTNET sites indicate that since about 2015, reduced N compounds comprise a greater proportion of total N deposition than do oxidized compounds, with reduced N in recent years generally accounting for more than 50% of total N deposition (Figure 6-19). Further, dry deposition of NH_3 as a percentage of total N deposition at CASTNET sites ranges up to a maximum of 65% at the highest site in 2021 (Figure 6-19). The 75^{th} percentile for these sites is greater than 30%, a noteworthy value given that these sites are generally in the West, with few in the areas of highest NH_3 emissions (Figures 6-20 and 2-9).

As a result of the contrasting temporal trends for emissions of oxidized and reduced N compounds, the influence of ambient air concentrations of N oxides and PM on N deposition appears to have declined over the past 20 years, complicating our consideration of the protection from N deposition-related effects that can be provided by secondary NAAQS for these pollutants. Thus a complicating factor in considering policy options related to NAAQS for addressing ecological effects related to N deposition is NH_3, which is not a criteria pollutant and its contribution to total N deposition, particularly in parts of the U.S. where N deposition is highest (e.g., Figure 6-18 and 6-13).

- **What does the available information indicate for considering the potential public welfare protection from N deposition-related effects in aquatic and terrestrial ecosystems?**

As discussed in section 4.5 above, effects of N deposition in both aquatic and terrestrial ecosystems have potential public welfare implications. For example, in the case of eutrophication in large estuaries and coastal waters of the eastern U.S., the public welfare significance of effects related to decades of N loading is illustrated by the large state, local and national government investments in activities aimed at reducing the loading. This significance relates both to the severity of the effects and the wide-ranging public uses dependent on these waters. These waterbodies are important sources of fish and shellfish production, capable of supporting large

stocks of resident commercial species and serving as breeding grounds and interim habitat for several migratory species, and also provide an important and substantial variety of cultural ecosystem services, including water-based recreational and aesthetic services. Further, these systems have non-use benefits to the public. The relative contribution of atmospheric deposition to total N loading, however, varies widely among estuaries, and has declined in more recent years, contributing a complexity to considerations in this review. While, such complications may not affect smaller, more isolated fresh waterbodies for which N loading is from atmospheric deposition, the evidence with regard to public welfare significance of any small deposition-related effects in these systems is less clear and well established. For example, the public welfare implications of relatively subtle effects of N enrichment in aquatic systems, such as shifts in phytoplankton species communities in remote alpine lakes, are not clear. Additionally, the public welfare implications of HNO_3 effects on lichens (which might be considered direct effects or the result of deposition) are also not clear, and might depend on the extent to which they impact whole communities, other biota or ecosystem structure and function.

With regard to N enrichment in terrestrial ecosystems, the associated effects may vary with regard to public welfare implications. As noted above with regard to impacts of aquatic acidification, we recognize that some level of N deposition and associated effects on terrestrial ecosystems can impact the public welfare and thus might reasonably be judged adverse to the public welfare. Depending on magnitude and the associated impacts, there are situations in which N deposition and associated nutrient enrichment-related impacts might reasonably be concluded to be significant to the public welfare. For example, to the extent forest ecosystem community structures are altered in ways that appreciably affect use and enjoyment of those areas by the public, implication for the public welfare are more obvious.

A complication to consideration of public welfare implications that is specific to N deposition in terrestrial systems is its potential to increase growth and yield of agricultural and forest crops, which may be judged and valued differentially than changes in growth of some species in natural ecosystems. Nitrogen enrichment in natural ecosystems can, by increasing growth of N limited plant species, change competitive advantages of species in a community, with associated impacts on the composition of the ecosystem's plant community. The public welfare implications of such effects may vary depending on their severity, prevalence or magnitude, such as with only those rising to a particular severity (e.g., with associated significant impact on key ecosystem functions or other services), magnitude or prevalence considered of public welfare significance.

- **What does the currently available quantitative information regarding terrestrial ecosystem responses to N deposition indicate about levels of N deposition that may be associated with increased concern for adverse effects?**

Focusing first on the evidence for effects of N deposition on trees, we note that the available quantitative information related to effects on plants, including trees, from N deposition summarized in Chapter 5 (and presented in more detail on Appendix 5B) includes soil chemistry modeling analyses for an indicator of soil acidification, as well as studies involving experimental additions of N compounds to defined field plots, and observational studies of potential relationships between tree growth and survival and metrics for N deposition. We consider the latter two types of studies here, as in Chapter 5 above, with regard to what each provides to inform the question posed above. Estimates from the array of studies indicates N deposition with a range of 7 to 12 kg/ha-yr, on a large area basis, may be a reasonable summary of conditions for which statistical associations have been reported for terrestrial effects, such as tree growth and survival and species richness of herbs and shrubs.

With regard to the information available from experimental addition tree studies, the ranges of N additions that elicited increased tree growth overlapped with those that elicited reduced growth and increased mortality. In considering these studies, we note that while some report observations based on additions over just a few years, others extend over a decade or more. In general, these studies inform our understanding of the effects on tree populations of increased N in forested areas, which can vary, influenced in part by other environmental factors, as well as by species-specific effects on population dynamics. The lowest forest N addition that elicited effects was 15 kg N/ha-yr over a 14-year period occurring from 1988-2002 (Appendix 5B, Table 5B-1; McNulty et al., 2005).

Among the available observational or gradient studies of N deposition and tree growth and survival (or mortality) are three recently available studies that utilized the USFS/FIA dataset of standardized measurements at sites across the U.S. (Dietze and Moorcroft, 2011; Thomas et al., 2010; Horn et al., 2018). These studies cover overlapping areas of the U.S. (see Appendix 5B, Figure 5B-1) and report associations of tree growth and/or survival metrics with various N deposition metrics, which provides support to conclusions regarding a role for N deposition in affecting tree health in the U.S., most particularly in regions of the eastern U.S., where confidence in the study associations is greatest (see summaries in section 5.3.2.3 and Appendix 5B, section 5B.3.2). The metrics utilized include site-specific estimates of average NO_3^- deposition and of average total N deposition over three different time periods (Dietze and Moorcroft, 2011; Thomas et al., 2010; Horn et al., 2018). In considering information from these studies discussed in section 5.3.2 and Appendix 5B, we note the history of N deposition in the eastern U.S. and the similarity between geographic patterns of historical deposition and more

recent deposition patterns in the U.S., which may influence the findings of observational studies, contributing an uncertainty to estimates of a specific magnitude of deposition rate that might be expected to elicit specific tree responses, such as increased or decreased growth or survival.

With regard to tree survival, Dietze and Moorcroft (2011) reported negative associations of mortality in multi-species groups (positive associations for survival) with average NO_3^- deposition at sites across the eastern half of the contiguous U.S. (i.e., higher survival rates in areas of higher NO_3^- deposition estimates). Site-specific average NO_3^- deposition in the analysis (1994-2005) ranged from a minimum of 6 kg/ha-yr to a maximum of 16 kg/ha-yr (Dietze and Moorcroft, 2011). Among 23 species in the northeastern and north-central U.S, the study by Thomas et al. (2010) reported negative and positive associations of N deposition (mean annual average for 2000-04) with survival for eight and three species, respectively. Positive and negative associations were reported with tree growth for 11 and 3 species, respectively. Site-specific average N deposition estimates in the analysis (2000-2004) ranged from a minimum of 3 kg/ha-yr to a maximum of 11 kg/ha-yr (Thomas et al., 2010). The other factors analyzed (e.g., temperature, precipitation, and tree size) did not include pollutants other than N deposition (Thomas et al., 2010).

The much larger study by Horn et al. (2018) of 71 species reported associations of tree survival and growth with N deposition that varied from positive to negative across the range of deposition at the measurement plots for some species, and also varied among species (Appendix 5B, section 5B.3.2.3). The median deposition values across the sample sites for species with significant positive or negative associations generally ranged from 7 to 11 kg N/ha-yr, as described in more detail in section 5.3.2 and Appendix B, section 5B.3.2.3. For species for which the association varied from negative to positive across deposition levels, this range includes those species for which the association was negative at the median deposition value (and for which sample sites were not limited to the western U.S.). Of the six species with negative associations of survival with the N deposition metric across the full range of the N deposition metric, the median deposition values ranged from 8 to 11 kg N ha $ha^{-1}yr^{-1}$ (Appendix 5B, Figure 5B-7). The median deposition values for the 19 other species with hump-shaped (or humped) functions that were negative at the median deposition value (and for which sample sites were not limited to the western U.S.) ranged from 7 to 11 kg N $ha^{-1}yr^{-1}$.

With regard to studies of herb and shrub community response, a number of recently available studies report on addition experiments, as summarized in section 5.3.3.1 and Appendix 5B, section 5B.3.1. The lowest N additions for which community effects have been reported include 10 kg N/ha-yr. With an addition of 10 kg N/ha-yr over a 10-year period, grassland species numbers declined; in a subset of plots for which additions then ceased, relative species numbers increased, converging with controls after 13 years (Appendix 5B, Table 5B-7; Clark

and Tilman, 2008). Recent gradient studies of coastal sage scrub in southern California have indicated N deposition above 10 or 11 kg/ha-year to be associated with increased risk of conversion to non-native grasslands or reduced species richness (Appendix 5B; section 5B.3.2; Cox et al., 2014; Fenn et al., 2010). A larger observational study of herb and shrub species richness in open- and closed-canopy communities using a database of site assessments conducted over a 23-year period and average N deposition estimates for a 26-year period reported significant influence of soil pH on the relationship between species richness and N deposition metric. A negative association was observed for acidic (pH 4.5) forested sites with N deposition estimates above 11.6 kg N/ha-yr and for low pH open canopy sites (woods, shrubs and grasses) with N deposition estimates above 6.5 kg N/ha-yr (section 5.3.3.1).

Lastly, the evidence base includes observational studies that have analyzed variation in lichen community composition in relation to indicators of N deposition (section 5.3.3.2 and Appendix 5B, section 5B.4.2). A recent study focused on relating metrics for community composition to estimated N deposition across sites in the Northwest reported an association of total N deposition in the range of 3 to 9 kg N/ha-yr with areas having 33-43% fewer species that grow well in low N environments and 3 to 4-fold more species that thrive in high N environments (Geiser et al., 2010). In addition to limitations with regard to interpretation, uncertainties associated with these studies include alternate methods for utilizing N deposition estimates as well as the potential influence of unaccounted-for environmental factors (e.g., ozone, SO_2 and historical air quality and associated deposition), as noted in section 5.3.3.2 above.

- **What does the currently available quantitative information regarding aquatic ecosystem responses to N deposition indicate about levels of N deposition that may be associated with increased concern for adverse effects?**

With regard to the evidence for effects of N deposition in aquatic ecosystems, we recognize several different types of information and evidence. This information includes the observational studies utilizing statistical modeling to estimate critical loads, such as those related to subtle phytoplankton species shifts in western lakes. This also includes the four to five decades of research on the impacts and causes of eutrophication in large rivers and estuaries. In considering this diverse evidence base, we take note of the robust evidence base on N loading and eutrophication, with its potentially significant impacts on submerged aquatic vegetation and fish species, particularly in large river systems, estuaries and coastal systems. As noted above, the public attention, including government expenditures, that has been given to N loading and eutrophication in several estuarine and coastal systems are indicative of the recognized public welfare implications of related impacts.

In large aquatic systems across the U.S., the relationship between N loading and algal blooms, and associated water quality impacts (both short- and longer-term), has led to numerous water quality modeling projects to inform water quality management decision-making in multiple estuaries, including Chesapeake Bay, Narraganset Bay, Tampa Bay, Neuse River Estuary and Waquoit (ISA, Appendix 7, section 7.2). These projects often utilize indicators of nutrient enrichment, such as chlorophyll a, dissolved oxygen, and abundance of submerged aquatic vegetation, among others (ISA, section IS.7.3 and Appendix 10, section 10.6). For these estuaries, the available information regarding atmospheric deposition and the establishment of associated target loads varies across the various estuaries (ISA, Appendix 7, Table 7-9). Further, in many cases atmospheric loading has decreased since the initial modeling analyses.

As summarized in section 5.2.3 above, analyses in multiple East Coast estuaries – including Chesapeake Bay, Tampa Bay, Neuse River Estuary and Waquoit Bay – have considered atmospheric deposition as a source of N loading (ISA, Appendix 7, section 7.2.1). Total estuary loading or loading reductions were established in TMDLs developed under the Clean Water Act for these estuaries. Levels identified for allocation of atmospheric N loading in the first three of these estuaries were 6.1, 11.8 and 6.9 kg/ha-yr, and atmospheric loading estimated to be occurring in the fourth was below 5 kg/ha-yr (section 7.3 below).

7.2.3.3 Relating Air Quality Metrics to N Deposition-related Effects of N Oxides and PM

Analyses in Chapter 6 explored how well various air quality metrics relate to S and N deposition. The analyses examine the relationships between air concentrations, in terms of various air quality metrics (including design values for the current standards), and N deposition in areas near or removed from the ambient air monitoring sites. The analyses utilizing data from NAAQS surveillance monitors are particularly relevant given that the current standards are judged using design values derived from FRM/FEM measurements at existing SLAMS. Given their role in surveillance for NAAQS violations, most or many of these monitors are located in areas of relatively higher pollutant concentrations, such as near large sources of NO_2 or PM. Accordingly, information from these monitoring sites can help inform how changes in NO_2 and/or PM emissions, reflected in ambient air concentrations, relate to changes in deposition and, correspondingly, what secondary standard options might best regulate ambient air concentrations such that deposition in sensitive ecosystems of interest is maintained at or below certain levels. In addressing the questions below, we consider the findings of those analyses specific to N deposition associated with N oxides and PM.

- **What do the available information and air quality analyses indicate regarding relationships between air quality metrics related to the existing standards and N deposition? What are the uncertainties in relationships using such metrics?**

In considering the information and analyses regarding relationships between N deposition and N oxides and PM in ambient air, we consider the current forms and averaging times of the secondary PM and NO_2 NAAQS. For N oxides, the current secondary standard is the annual average of NO_2, and that for PM is the average of three consecutive years of annual averages. As in the assessments of S deposition and air quality metrics, the analyses here focus on 3-year average metrics (e.g., annual average NO_2 and N deposition, averaged over three years) and include multiple time periods of data to better assess more typical relationships. For consistency and simplicity, most analyses in Chapter 6 focus on the five 3-year periods also used for S deposition and SO_X: 2001-03, 2006-08, 2010-12, 2014-16 and 2018-20.

As an initial matter, we note that, as discussed in section 6.4.2 above, relationships between N deposition and NO_2 and PM air quality are affected by NH_3 emissions and non-N-containing components of PM. Further, the influence of these factors on the relationships has varied across the 20-year evaluation period and varies across different regions of the U.S. (section 6.2.1). Both of these factors influence relationships between total N deposition and NO_2 and PM air quality metrics.

For total N deposition estimated for grid cells with collocated SLAMS monitors, the correlations with annual average NO_2 concentrations, averaged over three years, are low across all sites and in the East, although somewhat better for the West, with coefficient values of 0.38 and 0.44 for all sites and in the East, respectively, and 0.63 for West (Table 6-6). As noted in section 6.4.2, this likely reflects the relatively greater role of NH_3 in N deposition in the East (which for purposes of the analyses in this PA extends across the Midwest). For N deposition and NO_2 at upwind monitoring sites of influence, the correlation between estimates of total N deposition (wet plus dry) in eastern ecoregions and annual average NO_2 concentrations at monitor sites of influence (identified via trajectory-based modeling) for the five periods from 2001-2020 is low to moderate (0.35 and 0.48 for EAQM-max and EAQM-weighted, respectively), with the earlier part of the 20-year period, when NO_2 concentrations were higher and NH_3 emissions were lower (as indicated by Figures 6-6 and 6-5) having relatively higher correlation than the later part. The correlation is negative or near zero for the western ecoregions, as described in section 6.2.4 above.

As described in section 6.2.1 above, the reductions in NO_2 emissions over the past 20 years have been accompanied by a reduction in deposition of oxidized N. However, increases in NH_3 emissions, particularly in the latter 10 years of the period analyzed (2010-2020), have modified the prior declining trend in total N deposition. That is, coincident with the decreasing

trends in NO_2 emissions and in deposition of oxidized N in the past 10 years there is a trend of increased NH_3 and increased deposition of reduced N, most particularly in areas of the Midwest, Texas, Florida and North Carolina (Figures 6-16 and 6-17). This indicates that while, in the earlier years of the assessment period, controls on NO_2 emissions may have resulted in reductions in deposition of oxidized N, in more recent years they have much less influence on total N deposition (sections 6.2.1 and 6.4). In terms of ecoregion median statistics, Figure 7-6 illustrates a decreasing trend in ecoregion median total N deposition across the period from 2001 through 2012. From 2012 onward, it can be seen that deposition increases, most particularly in ecoregions in which the median % of total deposition that is reduced exceeds 50% (Figure 7-6, left and center panels).

The impact of increasing deposition of reduced N on the 20-year trend in total N deposition is also illustrated by TDep estimates at the nearly 100 CASTNET sites. At these sites, the median percentage of total N deposition comprised by oxidized N species, which is driven predominantly by N oxides, has declined from more than 70% to less than 45% (Figure 6-19). Examination of the components of reduced N deposition indicates the greatest influence on the parallel increase in N deposition percentage comprised of reduced N is the increasing role of NH_3 dry deposition. The percentage of total N deposition at the CASTNET sites has increased, from a median below 10% in 2000 to a median above 25% in 2021 (Figure 6-19).

Recognizing limitations in the extent to which CASTNET sites can provide information representative of the U.S. as a whole, we have also analyzed TDep estimates for the most recent period (2018-2020) with regard to total N deposition percent of total represented by reduced N across the U.S. Figure 7-7 illustrates that in areas with ecoregion median total N deposition above 9 kg/ha-yr (upper panel), the ecoregion median percentage of total N deposition comprised of reduced N is greater than 60% (lower panel). Further, in Figure 7-8, recent (2019-2021) TDep estimates across individual TDep grid cells provide a similar picture showing that areas of the U.S. where total N deposition is highest and is greater than potential targets identified in section 7.2.3.2 above (Figure 7-8, upper) are also the areas with the greatest deposition of NH_3 (Figure 7-8, lower), comprising more than 30% of total N deposition. That is, NH_3 driven deposition is greatest in regions of the U.S. where total deposition is greatest.

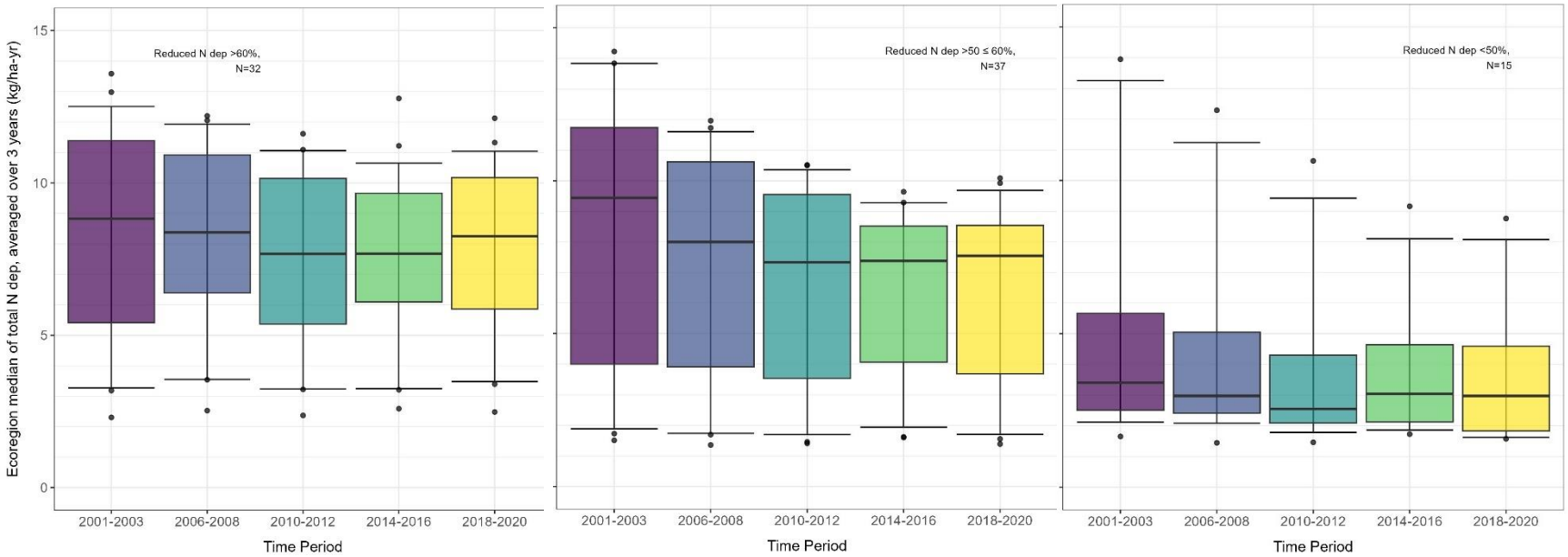

Figure 7-6. Temporal trend in ecoregion median estimates of total N deposition in ecoregions for which 2018-2020 TDep estimated reduced N deposition is >60% (left), 50-60% (middle) and <50%(right).

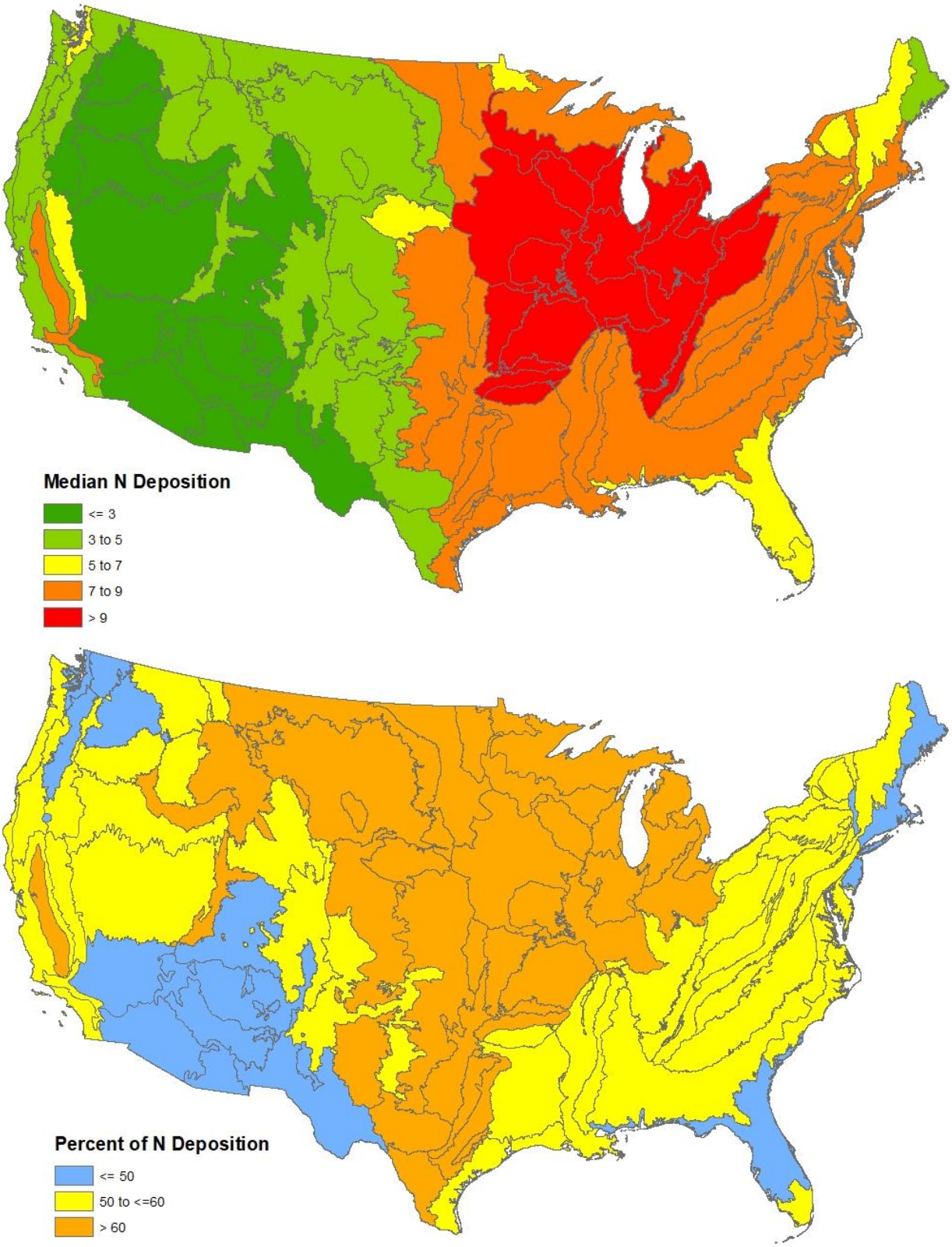

Figure 7-7. Ecoregion median total N deposition (upper panel) and percentage of total comprised of reduced N (lower panel) based on TDep estimates (2018-2020).

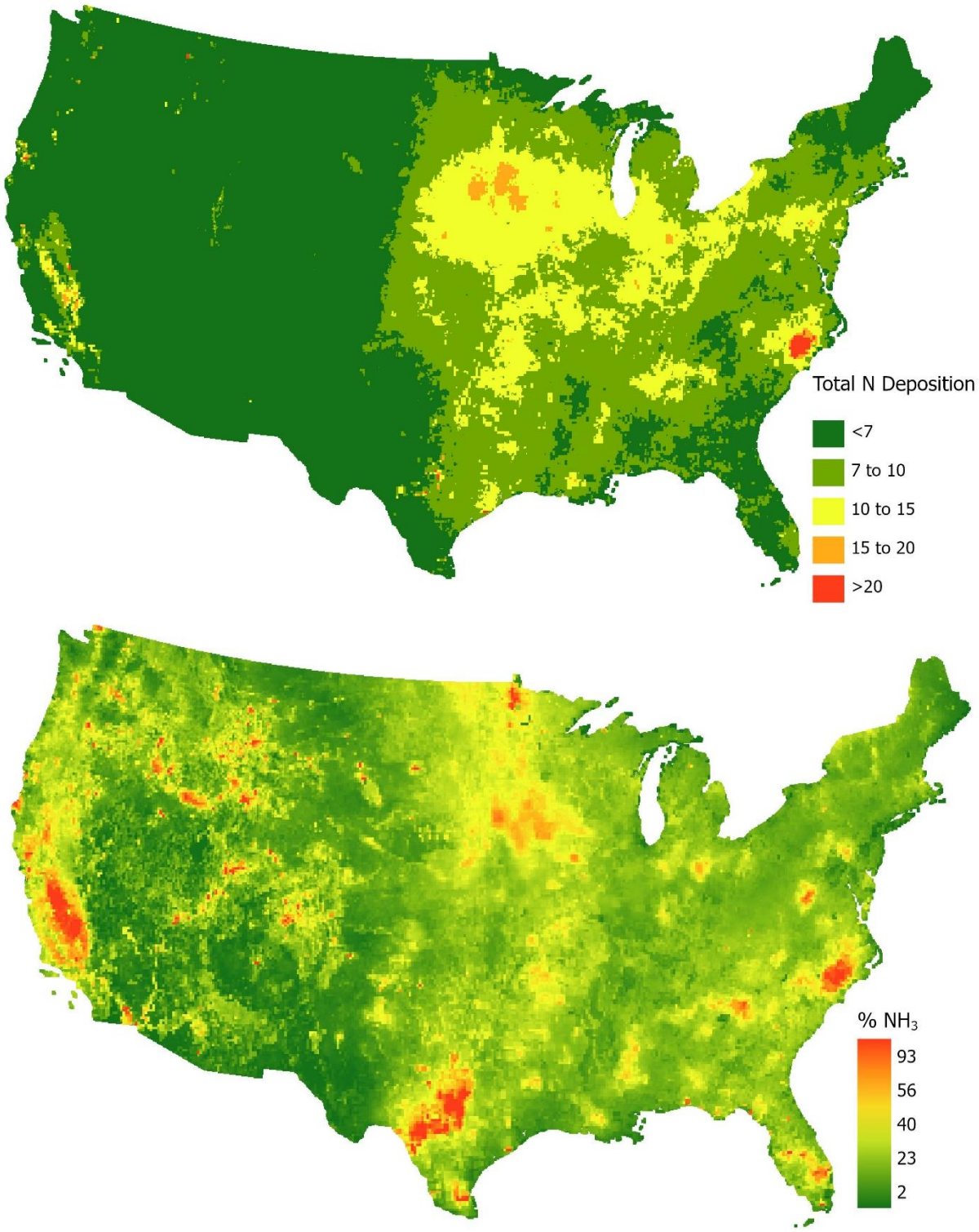

Figure 7-8. Estimated total N deposition (upper panel) and percentage of total comprised by NH_3 deposition (lower panel) based on TDep grid cells (2018-2020).

Regarding ecoregion median N deposition and $PM_{2.5}$ concentrations at upwind sites of influence, as with NO_2 concentrations, the correlation for eastern ecoregions (r=0.53 [max] and

0.62 [weighted]) is better than in western ecoregions, for which there is no correlation at all (section 6.2.4). For N deposition and $PM_{2.5}$ concentrations at SLAMS, as described in section 6.2.3 above, a low to moderate correlation is observed between total N deposition and annual average $PM_{2.5}$ concentrations (r=0.57 across all sites, 0.56 in East, 0.45 in West). In considering the two factors mentioned above, we note, as described in section 6.1 above, some NH_3 transforms to NH_4^+, which is a component of $PM_{2.5}$. As noted above, however, in the areas of greatest N deposition, the portion represented by deposition of gaseous NH_3 generally exceeds 30%. Additionally, while NH_3 emissions have been increasing over the past 20 years, the proportion of $PM_{2.5}$ that is comprised of N compounds has declined. As discussed in section 6.4.2 above, the median % of $PM_{2.5}$ comprised by N compounds at CSN sites declined from about 25% in 2006-2008 to about 17% in 2020-2022 and the highest percentage across sites declined from over 50% to 30% (Figure 6-56). Further this percentages varies regionally, with sites in the nine southeast states having less than 10% of $PM_{2.5}$ mass comprised of N compounds (Figure 6-56).

In summary, in recent years, NH_3, which is not a criteria pollutant, contributes appreciably to total N deposition, particularly in parts of the country where N deposition is highest (as illustrated by comparison of Figures 6-13 and 6-18). This situation, of an increasing, and spatially variable, portion of N deposition not being derived from N oxides or PM, complicates our assessment of policy options for protection against ecological effects related to N deposition associated with N oxides and PM, and on secondary standards for those pollutants that may be considered to be associated with a desired level of welfare protection. That notwithstanding, we have also considered analyses of SLAMS air quality data with regard to trends in annual average NO_2 concentrations (Figure 7-9) and relationships between annual average NO_2 concentrations (in a single year and averaged over three years) and design values for the existing primary standard (Figure 7-10).

From the temporal trend figures for N deposition and NO_2 concentrations, it can be seen that subsequent to 2011-2012, when median N deposition levels in 95% of the eastern ecoregions of the continental U.S.[10] have generally been at/below 11 kg N/ha-yr, annual average NO_2 concentrations, averaged across three years, have been at/below 35 ppb (Figures 7-6 and 7-9). Recognizing that among the NO_2 primary and secondary NAAQS, the 1-hour primary standard (established in 2010) may be the more controlling on ambient air concentrations, we considered the relationship among the two metrics (1-hr and annual). Figure 7-10 (left panel) below illustrates the relationship between 1-hour and annual design values for the existing primary and secondary NO_2 standards. Figure 7-10 (right panel) indicates that single-year annual average

[10]As noted earlier the eastern designation used throughout PA includes areas generally considered the Great Plains.

NO_2 concentrations, averaged over three years, in areas that meet the current 1-hour primary standard have generally been below approximately 35 to 40 ppb.

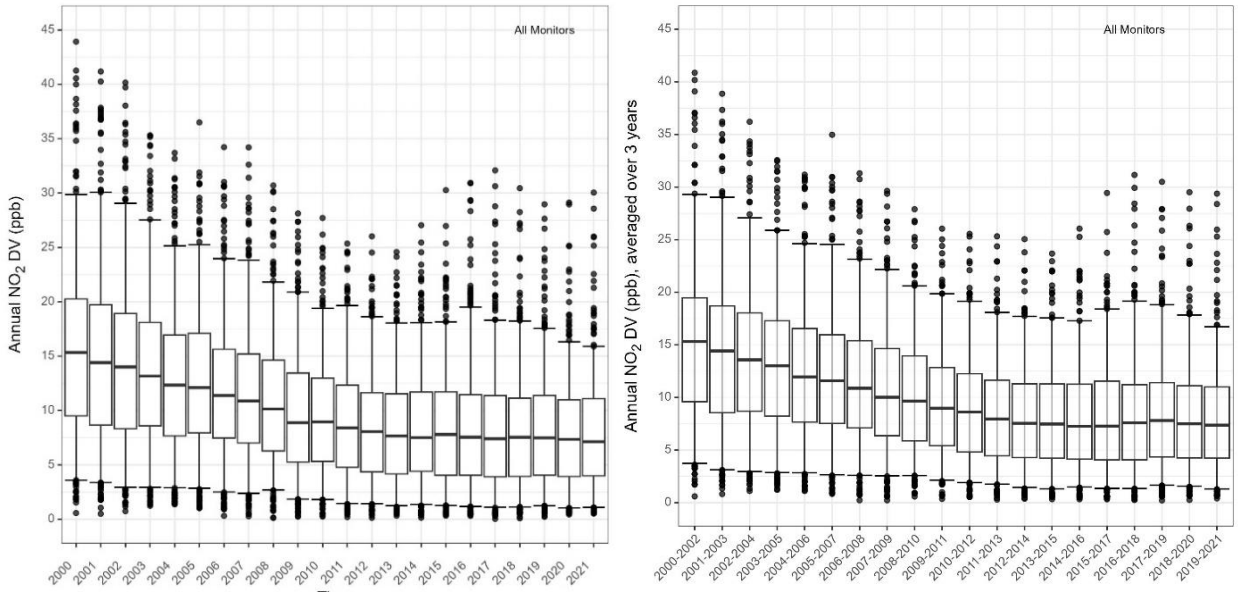

Figure 7-9. Temporal trend in annual NO_2 concentrations at SLAMS across U.S.: design values for existing standard (left) and 3-year averages of design values (right).

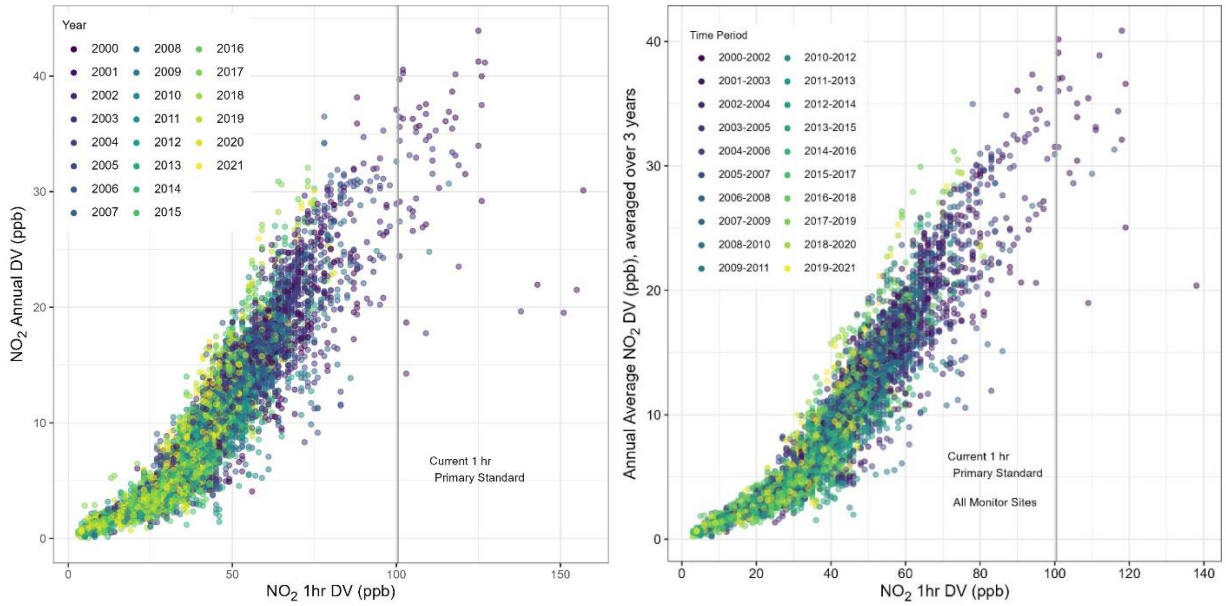

Figure 7-10. Annual NO_2 design values (left) and annual NO_2 concentrations, averaged over three years (right) associated with 1-hour NO_2 design values at SLAMS.

Given this information and these relationships, there may be some potential for a standard set as an NO_2 annual average (annual or averaged over three years), if reduced below the level of the existing standard, to contribute to a degree of control of N deposition (control of N deposition specifically associated with N oxides). However, this information also suggests the potential for future reductions in N oxide-related N deposition to be negated by increasing reduced N deposition. The results also suggest that the $PM_{2.5}$ annual average standard may provide some control of N deposition associated with PM and N oxides. We note, however, that $PM_{2.5}$ monitors, while capturing some compounds that contribute to S and N deposition across the U.S., also capture other non-S and non-N related pollutants as part of the $PM_{2.5}$ mass. Variation in the amounts of each category of compounds varies regionally (and seasonally), and as noted above, N-compounds generally comprise less than 30% of total $PM_{2.5}$ mass. Uncertainties associated with this variation and other uncertainties in the analyses are noted in Chapter 6, along with a characterization of the extent to which each of these uncertainties might impact interpretation of the various analyses (section 6.3).

- **What do the available information and air quality analyses indicate regarding relationships between air quality metrics based on indicators other than those of the existing standards and N deposition? What are the uncertainties in relationships using such metrics?**

As discussed above, Chapter 6 also assessed relationships for collocated measurements and modeled estimates of N compounds other than NO_2 with N deposition in a subset of 27 CASTNET sites located in 27 Class I areas, the majority of which (21 of 27) are located in the western U.S. The analyses indicated some correlations between concentrations of other air quality metrics and N deposition levels in these locations. For example, these results suggest that total N deposition (TDep) in these rural areas has a moderate correlation with annual average air concentrations of nitric acid and particulate nitrate for the 20-year dataset (2000-2020) (Figure 6-32, r=0.57 for TNO_3, r=0.63 for NO_3^-). These values are comparable to the correlation of NO_2 with total N dep (TDep) at western SLAMS, a not unexpected observation given that more than 75% of the 27 CASTNET sites are in the West. A much lower correlation was observed at SLAMS in the East, and with the trajectory-based dataset. As noted in section 6.4.2 above, deposition at the western U.S sites is generally less affected by NH_3. Further, the observed trend of increasing contribution to N deposition of NH_3 emissions over the past decade suggests that such correlations of N deposition with oxidized N may be still further reduced in the future. Thus, the evidence does not provide support for the oxidized N compounds (as analyzed at the 27 Class I sites) as indicators of total atmospheric N deposition, especially in areas where NH_3 is prevalent.

As similarly recognized above for S deposition and SO_X, the analyses involving total N deposition and ambient air NO_3^- (or NO_3^- plus HNO_3) concentrations are at remote locations, distant from sources of N oxides emissions, and the SLAMS, which monitor NO_2 (the primary precursor for atmospheric NO_3^-) are generally in areas near sources. Thus, these analyses do not indicate an advantage or benefit for an indicator based on NO_3^- measurements such as is currently collected at CASTNET sites, over options based on NO_2 as the indicator.

The analyses involving N deposition and N-containing PM components, also performed at the 27 Class I area sites yield similar correlation coefficients as those for 3-year average N deposition (TDep) and $PM_{2.5}$ at SLAMS monitors. For example, the correlation coefficients for annual total N deposition estimates with annual particulate NH_4^+ and NO_3^- combined, or particulate NH_4^+ alone, are all 0.62 (Figure 6-33), which is comparable to the correlation coefficient 0.57 for $PM_{2.5}$ mass design values observed at all U.S. SLAMS (Figure 6-39, upper panel), and also not much different from the value of 0.53 for $PM_{2.5}$ mass (IMPROVE) at the same 27 Class I area sites (Figure 6-32, left panel). Further the graphs of total N deposition estimates versus total N at the 27 Class I area sites indicate the calculated correlations (and slopes) likely to be appreciably influenced by the higher concentrations occurring in the first decade of the 20-year (Figure 6-33). Thus, the available analyses of N-containing $PM_{2.5}$ components at the small dataset of sites remote from sources, also do not indicate an overall benefit or advantage over consideration of $PM_{2.5}$ (discussed in section 7.4 below).

As a whole, the limited dataset with varying analytical methods and monitor locations, generally distant from sources, does not clearly support a conclusion that such alternative indicators might provide better control of N deposition related to N oxides and PM over those options discussed above (and used for the existing standards). It is also of note that use of the NO_3^- or particulate N measurements analyzed with deposition estimates at the 27 Class I area sites, alone or in combination with NO_2, as an indicator for a new standard would entail development of sample collection and analysis FRM/FEMs[11] and of a surveillance network.

7.3 CASAC ADVICE AND PUBLIC COMMENTS

In our consideration of the adequacy of the current secondary standards for SO_X, N oxides and PM, in addition to evidence and air quality/exposure/risk-based information discussed above, we have considered the advice and recommendations of the CASAC, based on their review of the ISA and the earlier draft of this PA, as well as comments from the public on the earlier draft of this PA. A limited number of public comments have been received in the docket

[11] For example, sampling challenges have long been recognized for particulate NH_4^+ (e.g., ISA, Appendix 2, sections 2.4.5; 2008 ISA, section 2.7.3).

for this review[12] to date, including just a few comments on the draft PA, and they were primarily focused on technical analyses and information, which we've considered in developing the final PA (section 1.4 above). The few public commenters that addressed the adequacy of the current secondary standards or potential alternative options to achieve appropriate public welfare protection expressed the view that the available evidence does not indicate the need for revision of the existing standards. The remainder of this section focuses on advice and recommendations from the CASAC regarding the standards review based on review of the draft PA.

The CASAC provided its advice regarding the current secondary standard in the context of its review of the draft PA (Sheppard, 2023). As an initial matter, the CASAC recognized that "translation of deposition-based effects to an ambient concentration in air is fraught with difficulties and complexities" (Sheppard, 2023, pp. 1-2). Further, the CASAC expressed its view that, based on its interpretation of the Clean Air Act, NAAQS could be in terms of atmospheric deposition, which it concluded "would be a cleaner, more scientifically defensible approach to standard setting" and accordingly recommended that direct atmospheric deposition standards be considered in future reviews (Sheppard, 2023, pp. 2 and 5). The CASAC then, as summarized below, provided recommendations regarding standards based on air concentrations, consistent with EPA's interpretations for NAAQS.

With regard to protection from effects other than those associated with ecosystem deposition of S and N compounds, the CASAC concluded that the existing SO_2 and NO_2 secondary standards provide adequate protection for direct effects of those pollutants on plants and lichens, recommending that these standards can be retained without revision for this purpose (Sheppard, 2023, p. 5 of letter and p. 23 of Response to Charge Questions). With regard to deposition-related effects of S and N compounds, the CASAC members did not reach consensus. Advice conveyed from both groups of members concerning deposition-related effects is summarized here.

With regard to deposition-related effects of S and standards for SO_X, the majority of CASAC members recommended a new annual SO_2 standard with a level in the range of 10 to 15 ppb, which these members concluded would generally maintain ecoregion median S deposition below 5 kg/ha-yr[13] based on consideration of the trajectory-based SO_2 analyses (and associated figures) in the draft PA (Sheppard, 2023, Response to Charge Questions, p. 25). They concluded

[12] The docket for this review of the secondary standards for SO_X, N oxides and PM is EPA-HQ-OAR-2014-0128, accessible from www.regulations.gov.

[13] Although the CASAC letter does not specify the statistic for the 5kg/ha-yr value, the analyses referenced in citing that value, both the trajectory analyses and the ecoregion-scale summary of aquatic acidification results, focus on ecoregion medians. So that is how it is interpreted here.

that such a level of S deposition would afford protection for tree and lichen species,[14] and aquatic ecosystems. Regarding aquatic ecosystems, these members cite the ecoregion-scale estimates (from the aquatic acidification REA) associated with median S deposition bins for the 90 ecoregion-time period combinations (summarized in section 5.1.3.2 above) in conveying that for S deposition below 5 kg/ha-yr, 80%, 80% and 70% of waterbodies per ecoregion are estimated to achieve an ANC at or above 20, 30 and 50, respectively, in all ecoregion-time period combinations (Sheppard, 2023, Response to Charge Questions, p 25).[15] In recommending an annual standards with a level in the range of 10-15 ppb, these members stated that such a standard would "preclude the possibility of returning to deleterious deposition values as observed associated with the emergence of high annual average SO_2 concentrations near industrial sources in 2019, 2020, and 2021," citing Figure 2-25 of the draft PA[16] (Sheppard, 2023, Response to Charge Questions, p. 24).

One CASAC member dissented from this recommendation for an annual SO_2 standard[17] and instead recommended adoption of a new 1-hour SO_2 secondary standard identical in form, averaging time, and level to the existing primary standard based on the conclusion that the ecoregion 3-year average S deposition estimates for the most recent periods are generally below 5 kg/ha-yr and that those periods correspond to the timing of the existing primary SO_2 standard (established in 2010), indicating the reduced deposition to be a product of current regulatory requirements (Sheppard, 2023, Appendix A, p. A-2).

With regard to N oxides and protection against deposition-related welfare effects of N, the majority of CASAC members recommended revision of the existing annual NO_2 standard to a level "<10 – 20 ppb" (Sheppard, 2023, Response to Charge Questions, p.24). The justification these members provide is related to their consideration of the relationship presented in the draft

[14] In making this statement, these CASAC members cite two observational data studies with national-scale study areas published after the ISA: one study is on lichen species richness and abundance and the second is on tree growth and mortality (Geiser et al., 2019; Pavlovic et al., 2023). The lichen study by Geiser et al. (2019) relies on lichen community surveys conducted at USFS sites from 1990 to 2012. The tree study by Pavlovic et al. (2023) utilizes machine learning models with the dataset from the observational study by Horn et al. (2018) to estimate confidence intervals for CLs for growth and survival for 108 species based on the dataset first analyzed by Horn et al. (2018).

[15] As seen in Table 7-1, these levels of protection are also achieved in ecoregion-time period combinations for which the ecoregion median S deposition estimate is at or below 7 kg/ha-yr.

[16] This figure is the prior version of Figure 2-28 in section 2.4.2 of this final PA. The figure presents temporal trend in distribution (box and whiskers) of annual average SO_2 concentrations at SLAMS.

[17] Also dissenting from this advice was a member of the CASAC Oxides of Nitrogen, Oxides of Sulfur and Particulate Matter Secondary NAAQS Panel who was not also a member of CASAC (Sheppard, 2023, Response to Charge Questions, p. 23).

PA of median ecosystem N deposition with the weighted[18] annual average NO_2 metric concentrations, averaged over three years, at monitoring sites linked to the ecosystems by trajectory-based analyses and a focus on total N deposition estimates at or below 10 kg/ha-yr (Sheppard, 2023, Response to Charge Questions, p. 24). These members additionally recognize, however, that "when considering all ecoregions, there is no correlation between annual average NO_2 and N deposition" (Sheppard, 2023, Response to Charge Questions, p. 24). A focus on total N deposition estimates at or below 10 kg/ha-yr appears to relate to consideration of total maximum daily load[19] analyses in four East Coast estuaries: Chesapeake Bay, Tampa Bay, Neuse River Estuary and Waquoit Bay (Sheppard, 2023, Response to Charge Questions, pp. 12-14 and 29). Levels identified for allocation of atmospheric N loading in the first three of these estuaries were 6.1, 11.8 and 6.9[20] kg/ha-yr, and atmospheric loading estimated in the fourth was below 5 kg/ha-yr (Sheppard, 2023, Response to Charge Questions, pp. 12-14). Another consideration may be these members' conclusion that 10 kg N/ha-yr is "at the middle to upper end of the N critical load threshold for numerous species effects (e.g., richness) and ecosystem effects e.g., tree growth) in U.S. forests grasslands, deserts, and shrublands (e.g., Pardo et al., 2011; Simkin et al., 2016) and thus 10 kg N/ha-yr provides a good benchmark for assessing the deposition-related effects of NO_2 in ambient air" (Sheppard, 2023, Response to Charge Questions, p. 23).

One CASAC member disagreed with revision of the existing annual NO_2 standard and instead recommended adoption of a new 1-hour NO_2 secondary standard identical in form, averaging time and level to the existing primary standard based on the conclusion that the N deposition estimates for the most recent periods generally reflect reduced deposition that is a product of current regulatory requirements, including the existing primary standards for NO_2 and PM (Sheppard, 2023, Appendix A). This member additionally notes that bringing into attainment the areas still out of attainment with the existing primary standard will provide further reductions in N deposition. This member also notes his analysis of NO_2 annual and 1-hour design values for

[18] As described in section 6.2.4 above, the weighted metric is constructed by applying weighting to concentrations to the monitors identified as sites of influence, with the weighting equal to the relative contribution of air from the monitor location to the downwind ecoregion based on the trajectory analysis (section 6.2.4). Values of this metric are not directly translatable to individual monitor concentrations or to potential standard levels.

[19] Total Maximum Daily Loads or TMDLs are an approach under the Clean Water Act for allocating loading to a waterbody that is projected to allow the waterbody to meet its water quality standards, as described further in section 5.2.3 above.

[20] The CASAC letter states that the Neuse River Estuary TMDL specified a 30% reduction from the 1991-95 loading estimate of 9.8 kg/ha-yr, yielding a remaining atmospheric load target of 6.9 kg/ha-yr (Sheppard, 2023, Response to Charge Questions, p. 13).

the past 10 years (2013-2022) that indicates the current primary NO_2 standard to provide protection for annual average NO_2 concentrations below 31 ppb (Sheppard, 2023, Appendix A).

With regard to PM and effects related to deposition of N and S, the CASAC focused on the $PM_{2.5}$ standards, and made no recommendations regarding the PM_{10} standard. In considering the annual $PM_{2.5}$ standard, the majority of CASAC members recommended revision of the annual secondary $PM_{2.5}$ standard to a level of 6 to 10 $\mu g/m^3$. In describing their justification for this range, these members focus on rates of total N deposition at/below 10 kg/ha-yr and total S deposition at/below 5 kg/ha-yr – as in their advice regarding SO_2 and NO_2 standards (summarized above) – that they state would "afford an adequate level of protection to several species and ecosystems across the U.S." (Sheppard, 2023, Response to Charge Questions, p. 23). In reaching this conclusion for protection from N deposition, the CASAC majority cites studies of U.S. forests, grasslands, deserts and shrublands that are included in the ISA. For S deposition, the CASAC majority notes the Pavlovic et al. (2023) analysis of the dataset used by Horn et al. (2018). Conclusions of the latter study, which is characterized in the ISA and discussed in sections 5.3.2.3 and 7.2.2.2 above (in noting median deposition of 5-12 kg S/ha-yr in ranges of species for which survival and/or growth was observed to be associated with S deposition), is consistent with the more recent analysis in the 2023 publication (ISA, Appendix 6, sections 6.2.3 and 6.3.3).

As justification for their recommended range of annual $PM_{2.5}$ levels (6-10 $\mu g/m^3$), this group of CASAC members makes several statements regarding annual $PM_{2.5}$ concentrations and estimates of S and N deposition for which they cite several figures in the draft PA. Citing figures in the draft PA with TDep deposition estimates and IMPROVE and CASTNET monitoring data, they state "[i]n remote areas, IMPROVE $PM_{2.5}$ concentrations in the range of 2-8 $\mu g/m^3$ for the periods 2014-2016 and 2017-2019 correspond with total S deposition levels <5 kg/ha-yr (Figure 6-12), with levels generally below 3 kg/ha-yr, and with total N deposition levels ≤10 kg/ha-yr (Figure 6-13)" (Sheppard, 2023, Response to Charge Questions, p. 23). With regard to S deposition, these members additionally cite a figure in the draft PA as indicating ecosystem median S deposition estimates at/below 5 kg/ha-yr occurring with $PM_{2.5}$ EAQM-max values in the range of 6 to 12 $\mu g/m^3$ (Sheppard, 2023, Response to Charge Questions, pp. 23-24). These members additionally cite figures in the draft PA as indicating that areas of 2019-2021 total N deposition estimates greater than 15 kg/ha-yr (in California, the Midwest and the East) correspond with areas where the annual $PM_{2.5}$ design values for 2019-2021 range from 6 to 12 $\mu g/m^3$, and other figures (based on trajectory analyses) as indicating ecosystem median N deposition estimates below 10 kg N/ha-yr occurring only with $PM_{2.5}$ weighted EAQM values

below 6 μg/m^3,[21] and $PM_{2.5}$ EAQM-max values below 8 μg/m^3 (Sheppard, 2023, Response to Charge Questions, pp. 23-24). The CASAC also notes that the correlation coefficient for N deposition with the weighted EAQM is 0.52, while the correlation coefficient with the EAQM-max is near zero (0.03). The bases for the N and S deposition levels targeted in this CASAC majority recommendation are described in the paragraphs above.

One CASAC member recommended revision of the annual secondary $PM_{2.5}$ standard to a level of 12 μg/m^3 based on his interpretation of figures in the draft PA that present S and N deposition estimates for five different 3-year time periods from 2001 to 2020. This member observes that these figures indicate ecoregion median S and N deposition estimates in the last 10 years below 5 and 10 kg/ha-yr, respectively. This member concludes this to indicate that the current primary annual $PM_{2.5}$ standard provides adequate protection against long-term annual S and N deposition-related effects (Sheppard, 2023, Appendix A).

Regarding the existing 24-hour $PM_{2.5}$ secondary standard, the majority of CASAC members recommend revision of the level to 25 ug/m^3 or revision of the indicator and level to deciviews and 20 to 25, respectively (Sheppard, 2023, Response to Charge Questions, p 25). These members variously cite "seasonal variabilities" of "[e]cological sensitivities," describing sensitive lichen species to be influenced by fog or cloud water from which they state S and N contributions to be highly episodic, and visibility impairment (Sheppard, 2023, Response to Charge Questions, p 25). These members do not provide further specificity regarding their reference to lichen species and fog or cloud water. With regard to visibility impairment, these members describe the EPA solicitation of comments that occurred with the separate EPA action of reconsidering the 2020 decision on the secondary $PM_{2.5}$ standard in providing requisite protection from visibility effects as the basis for the specific recommendations they make (Sheppard, 2023, Response to Charge Questions, p 25; 88 FR 5562-5663, January 27, 2023).[22]

[21] As noted earlier in this section, weighted EAQM values are not directly translatable to concentrations at individual monitors or to potential standard levels.

[22] The context for solicitation of comment regarding the 24-hour $PM_{2.5}$ secondary standards and the associated target level of visibility protection is provided in the Federal Register notice for that action (88 FR 5558, January 27, 2023), a quotation from which is provided here:

> *With regard to visibility effects, while the Administrator notes that the CASAC did not recommend revising either the target level of protection for the visibility index or the level of the current secondary 24-hour $PM_{2.5}$ standard, the Administrator recognizes that, should an alternative level be considered for the visibility index, that the CASAC recommends also considering revisions to the secondary 24-hour $PM_{2.5}$ standard. In considering the available evidence and quantitative information, with its inherent uncertainties and limitations, the Administrator proposes not to change the secondary PM standards at this time, and solicits comment on this proposed decision. In addition, the Administrator additionally solicits comment on the appropriateness of a target level of protection for visibility below 30 dv and down as low as 25 dv, and of revising the level of the current secondary 24-hour $PM_{2.5}$ standard to a level as low as 25 μg/m^3.*

One CASAC member dissented from this view and supported retention of the existing 24-hr $PM_{2.5}$ standard.

Among the CASAC comments on the draft PA regarding and recommendations for revising the PA[23] was the comment that substantial new evidence has been published since development of the 2020 ISA that supports changes to the draft PA conclusions on N deposition effects. More specifically, the CASAC noted new literature regarding taxonomic groups affected by elevated N deposition, national-scale data documenting adverse ecological effects of elevated N, and lower levels of N deposition and associated quantified ecological effects (Sheppard, 2023, Response to Charge Questions, p. 7). The CASAC raised the issue of more recent studies in the context of its comments on chapters 4 and 5 (Sheppard, 2023, Response to Charge Questions, pp. 7-17). In these comments, the CASAC cites a number of studies published after May 2017 and not included in the ISA, along with many previously available studies that are described in the ISA. The array of topics on which the CASAC recommended updates to the PA includes effects of atmospheric N deposition on various aspects of managed terrestrial ecosystems (including recognition of benefits and disbenefits) and on freshwater and coastal aquatic ecosystems; indicators of acidification and ecosystem N status; comparisons of steady state and dynamic environmental modeling; the influence of climate change; and temporal changes in atmospheric deposition (and associated changes in soil and water quality parameters) on the ecological effects of N and S deposition (Sheppard, 2023, Response to Charge Questions, pp. 8-17). As noted in section 1.4 above, a number of aspects of chapters 4 and 5 in this final PA are revised from the draft PA in consideration of the information that was emphasized by the CASAC in this way while also referring to the ISA and studies considered in it.[24]

7.4 SUMMARY OF STAFF CONCLUSIONS

This section summarizes staff findings and identifies policy options for the Administrator's consideration in this review of the secondary NAAQS for SO_X, N oxides and PM. These conclusions are based on consideration of the assessment and integrative synthesis of the evidence, as summarized in the ISA, and the 2008 ISA, the 2009 PM ISA and AQCDs from prior reviews, and the quantitative information on exposure and air quality summarized above, as well as the advice of the CASAC. Taking into consideration the discussions above in this chapter, this section addresses the following overarching policy question.

[23] Consideration of CASAC comments and areas of the PA in which revisions have been made between the draft and this final document are described in section 1.4 above.

[24] More recent studies cited by the CASAC generally concern effects described in the ISA based on studies available at that time. While the newer studies include additional analyses and datasets, the ISA and studies in it also generally support the main points raised and observations made by the CASAC.

- **Do the current evidence and quantitative analyses call into question the adequacy of protection from ecological effects afforded by the SO_2, NO_2 and PM secondary standards? What alternate standards may be appropriate to consider with regard to protection from ecological effects of SO_X, N oxides and PM?**

In considering this question, we first recognize what the CAA specifies with regard to protection to be provided by the secondary standards. Under section 109(b)(2) of the CAA, the secondary standard is to "specify a level of air quality the attainment and maintenance of which in the judgment of the Administrator … is requisite to protect the public welfare from any known or anticipated adverse effects associated with the presence of such air pollutant in the ambient air." The secondary standard is not meant to protect against all known or anticipated SO_2 related welfare effects, but rather those that are judged to be adverse to the public welfare, and a bright-line determination of adversity is not required in judging what is requisite (78 FR 3212, January 15, 2013; 80 FR 65376, October 26, 2015; see also 73 FR 16496, March 27, 2008). Thus, our consideration of the currently available information regarding welfare effects of the oxides of sulfur and nitrogen and of PM is in this context, while recognizing that the level of protection from known or anticipated adverse effects to public welfare that is requisite for the secondary standard is a public welfare policy judgment to be made by the Administrator.

The general approach in a review of a secondary NAAQS, and accordingly in associated PAs, involves, first, evaluation of the currently available information with regard to key considerations for assessing risk of or protection for the effects of the criteria pollutant of focus. In this evaluation, the PA considers the welfare effects of the pollutant, associated public welfare implications, and also the quantitative information, such as that regarding exposure-response relationships, and associated tools or metrics, as well as associated limitations and uncertainties. The quantitative tools (e.g., metrics for effects and metrics for summarizing exposures) allow for identification and assessment of exposures of concern and, correspondingly, of exposures appropriate for focus in assessing protection afforded by the existing standard(s), and as appropriate, in assessing potential alternatives. The latter part of the general approach in a review and a PA is then consideration of the extent to which the existing standard(s) provides air quality that would be expected to achieve such protection and, as appropriate, potential alternative options (e.g., standard or standards) that could be expected to achieve this desired air quality. This consideration goes beyond a focus on the key exposure metrics and concentrations of potential concern to whether the indicator, form, averaging time, and level of the standard (or suite of standards), together, provide the requisite protection.

As in NAAQS reviews in general, the extent to which the protection provided by the current secondary standards for SO_X, N oxides and PM are judged to be adequate depends on a variety of factors, including science policy judgments and public welfare policy judgments.

These factors include public welfare policy judgments concerning the appropriate benchmarks on which to place weight, as well as judgments on the public welfare significance of the effects that have been observed at the exposures evaluated in the welfare effects evidence. The factors relevant to judging the adequacy of each standard also include the interpretation of, and decisions as to the weight to place on, different aspects of the quantitative analyses of air quality, exposure and risk, and any associated uncertainties. Additionally, to the extent multiple policy options are identified that might be expected to achieve a desired level of protection, decisions on the approach to adopt fall within the scope of the Administrator's judgment. In the end, the Agency's decisions on the adequacy of the current secondary standard and, as appropriate, on any potential alternative standards considered in a review, are largely public welfare policy judgments made by the Administrator. Accordingly, the Administrator's conclusions regarding the adequacy of the current standard will depend in part on public welfare policy judgments, on science policy judgments regarding aspects of the evidence and exposure/risk estimates, and on judgments about the level of public welfare protection that is requisite under the Clean Air Act. Thus, the Administrator's final decisions draw upon the scientific information and analyses about welfare effects, environmental exposures and risks, and associated public welfare significance, as well as judgments about how to consider the range and magnitude of uncertainties that are inherent in the scientific evidence and analyses.

In the discussion below, we address first the SO_2 standard, and its adequacy with regard to protection of the public welfare from effects of SO_X in ambient air other than those associated with ecosystem deposition of S compounds. Next, we address the extent of protection provided by the SO_2 standard from S deposition-related effects of SO_X in ambient air, and consideration of alternate standards for this purpose. In so doing, we focus primarily on the contribution of SO_X in ambient air to ecosystem acidification and particularly aquatic acidification. After addressing SO_X in this way, we next consider the NO_2 standard and its adequacy with regard to protection of the public welfare from effects of N oxides in ambient air other than those associated with ecosystem N deposition, as well as the extent of protection provided by the NO_2 standard from deposition related effects of N oxides in ambient air and consideration of alternate standards for this purpose. Lastly, we address the PM standards and the extent of their protection of the public welfare from ecological effects. In each case, we recognize limitations in the available information and tools and associated uncertainties, which vary in specificity and significance.

The existing SO_2 secondary standard is 0.5 ppm, as a 3-hour average concentration not to be exceeded more than once per year. The evidence of welfare effects at the time this standard was established in 1971 indicated the effects of SO_X on vegetation, most particularly effects on foliar surfaces. The currently available information continues to document the occurrence of visible foliar injury as a result of acute or short exposures (e.g., of a few hours), with greater

exposures (repeated and/or of longer duration) affecting plant growth and yield. As summarized in the ISA, there is "no clear evidence of acute foliar injury below the level of the current standard" (ISA, section IS.4.1, p. IS-37).

We additionally note that across all sites (outside Hawaii, where air quality can be influenced by volcanic emissions) during all years from 2000 through 2021, with the exception of one occurrence in 2010, all design values for the existing 3-hour standard (not to be exceeded more than once in a year) are below the standard level of 0.5 ppm. Further, 95% of values have been below 0.2 ppm in each year of the 22-year period and below 0.1 ppm since 2011 (Figure 2-27). As summarized in section 5.4.1 above, the available evidence does not indicate effects on plants or lichens for short-term air concentrations within this distribution. Thus, as the available evidence does not indicate ecological effects associated with the pattern of concentrations allowed by the existing standard, we find that the currently available information, including that newly available in this review, does not call into question the adequacy of protection provided by the existing SO_2 standard from the direct effects of SO_X in ambient air. Further, we note that the CASAC unanimously made a similar conclusion that the current 3-hour standard provides adequate protection against such direct effects on plants and lichens and should be retained (Sheppard, 2023, p. 23). In light of these considerations summarized immediately above, we conclude that the information available in this review does not call into question the adequacy of the existing standard in providing protection against effects related to the direct action of SO_X on plants and lichens.

With regard to deposition-related effects, we note the range of ecoregion median deposition estimates across U.S. ecoregions analyzed during the 20-year period from 2001 through 2020 extended up through 10 kg S/ha-yr to as high as 20 kg S/ha-yr during years when the existing SO_2 standard was met in all but one occasion (in 2011) in contiguous U.S., and when design values for the standard (second highest 3-hour average in a year) ranged well below 500 ppb (as discussed in section 6.2.1 above). For example, in the earliest 3-yr period (2001-03), when virtually all design values for the existing 3-hour standard were below 400 ppb and the 75^{th} percentile of design values was below 100 ppb (Figure 2-27), total S deposition was estimated to be greater than 14 kg/ha-yr across the Ohio River valley and Mid-Atlantic states, ranging above 20 kg/ha-yr in portions of this area (Figure 6-11). The magnitude of S deposition estimates at the 90^{th} percentile per ecoregion at sites assessed in the aquatic acidification REA was at or above 15 kg/ha-yr in half of the 18 eastern ecoregions and ranged up to nearly 25 kg/ha-yr during this time period (Figure 7-2). The aquatic acidification risk estimates indicate, as illustrated in Figure 5-13 above, that this pattern of S deposition is associated with 20% to more than 50% of waterbody

sites in affected eastern ecoregions[25] being unable to achieve the lowest of the three acid buffering capacity targets (ANC of 20 µeq/L), indicating risks of potential public welfare significance. Considering that these aquatic acidification risk estimates are associated with S deposition during periods when the existing standard has been met (e.g., 2000-2002), it is reasonably concluded that the current evidence and quantitative analyses call into question the adequacy of the existing standard with regard to S deposition-related effects such as aquatic acidification. Thus, we have evaluated options for potential alternative standards that may be more appropriately associated with protection of welfare effects.

For the purposes of evaluating options for potential alternative standards for deposition-related effects of SO_X, we draw on the quantitative analyses and information described in Chapter 5 and summarized in section 7.2.2 above. In this context and for our purposes within this PA, we primarily focus on the aquatic acidification risk estimates, and particularly the ecoregion-scale analyses. In focusing on the aquatic acidification risk estimates for our consideration of acidification risks, we also note the linkages between watershed soils and waterbody acidification, as well as terrestrial effects. Such linkages indicate that protecting waterbodies from reduced acid buffering capacity (with ANC as the indicator) will also, necessarily, provide protection for watershed soils, and may reasonably be expected to also contribute protection for terrestrial effects. That notwithstanding, we recognize there to be limitations of the quantitative analyses and associated uncertainties in their interpretation, as referenced in Chapter 5. Accordingly, in focusing on specific ranges of deposition that may provide protection for waterbody acid buffering capacity for our purposes here, we note there to be relatively greater uncertainty associated with the lower deposition levels. Moreover, we recognize that, in the end, judgments inherent in identification of such a range, include judgments related to the weighing of uncertainties, as well as the consideration of the appropriate targets for public welfare protection, and fall within the purview of the Administrator.

In focusing on the ecoregion-scale findings of the aquatic acidification REA, with particular attention to the 18 well studied, acid-sensitive eastern ecoregions, we consider the ecoregion median S deposition values at and below which the associated risk estimates indicated a high proportion of waterbodies in a high proportion of ecoregions to achieve ANC values at or above the three targets (20, 30 and 50 µeq/L), as summarized in Tables 7-1 and 5-5, above. As an initial matter, we note the approach taken by the CASAC majority in considering these estimates (summarized in section 7.3 above). These members considered the ecoregion-scale analysis summary in Table 5-5 and took note of estimated achievement of ANC at or above the

[25] Aquatic acidification risk estimates for the 2001-2020 deposition estimates in the eight western ecoregions indicated ANC levels achieving all three targets in at least 90% of all sites assessed in each ecoregion (Table 5-4). Ecoregion median deposition estimates were at or below 2 kg/ha-yr in all eight western ecoregions (Table 5-3).

three ANC targets in 80% (for ANC of 20 and 30) or 70% (for ANC of 50) of waterbody CL sites in all ecoregion-time periods for which the ecoregion median S-deposition was below 5 kg/ha-yr (Sheppard, p. 25 of the Response to Charge Questions). We note that the results for ecoregion-time period combinations for median S-deposition in the 18 eastern ecoregions at or below 7 kg/ha-yr also achieve these percentages of waterbodies achieving the three ANC targets (as seen in Tables 7-1 and 5-5 above).[26] The results for median S deposition at or below 7 kg/ha-yr further indicate that 90% of waterbodies per ecoregion achieve ANC at/above targets of 20, 30 and 50 in 96%, 92% and 82%, respectively, of eastern ecoregion-time period combinations. For median S deposition at or below 9 kg/ha-yr, the percentages of ecoregions meeting or exceeding the ANC targets declines to 87%, 81% and 72% (as summarized in section 7.2.2.2., above).

We additionally consider the temporal trend or pattern of ecoregion-scale risk estimates across the five time periods in relation to the declining S deposition estimates for those periods. In so doing, we note the estimates of appreciably improved acid buffering capacity (increased ANC) by the third time period (2010-2012) and so consider the REA risk and deposition estimates for these and subsequent periods. The S deposition estimated to be occurring in the 2010-2012 time period included ecoregion medians (based on CL sites) ranging from 2.3 to 7.3 kg/ha-yr in the 18 eastern ecoregions and extending down below 1 kg/ha-year in the 7 western ecoregions; the highest ecoregion 90th percentile was approximately 8 kg/ha-yr (Table 7-2, Figure 7-2). For this pattern of deposition, more than 70% of waterbodies per ecoregion are estimated to be able to achieve an ANC of 50 ueq/L in all 25 ecoregions (Figure 7-1, left panel), and more than 80% of waterbodies per ecoregion in all ecoregions are estimated to be able to achieve an ANC of 20 ueq/L (Figure 7-1, right panel). Further, by the 2014-2016 period, when both median and 90th percentile S deposition in all 25 ecoregions was estimated to be at or below 5 kg/ha-yr, more than 80% of waterbodies per ecoregion are estimated to be able to achieve an ANC of 50 ueq/L in all 25 ecoregions (more than 90% in 23 of the 25 ecoregions) and more than 90% of waterbodies per ecoregion in all ecoregions are estimated to be able to achieve an ANC of 20 ueq/L (Figure 7-1, right panel).

The estimates of acid buffering capacity achievement for the 2010-12 period deposition — achieving the ANC targets in at least 70% to 80% (depending on the target) of waterbodies per ecoregion — are consistent with the objectives identified by the CASAC (in considering estimates for the 18 eastern ecoregions). The advice from the CASAC emphasized ecoregion ANC achievement estimates of 70%, 80% and 80% for ANC targets of 50, 30 and 20 µeq/L, respectively. The estimates for the later time period are somewhat better, with all ecoregions

[26] Ecoregion median deposition was below 2 kg S/ha-yr in all 35 ecoregion-time period combinations for the eight western ecoregions (Table 5-4).

estimated to achieve the ANC targets in at least 80% to 90% (depending on the target) of waterbodies per ecoregion. Both of these ecoregion-scale ANC achievement results (70% to 80% and 80% to 90%) may be reasonable to consider with regard to acid buffering capacity objectives for the purposes of protecting ecoregions from aquatic acidification risk of a magnitude with potential to be considered of public welfare significance.

In considering the aquatic acidification risk estimates at the ecoregion-scale for the purpose of identifying a range of ecoregion deposition estimates on which to focus in identifying options for potential secondary standards, we consider both sets of potential objectives for acid buffering capacity intended to provide an appropriate degree of protection from S deposition-related effects related to aquatic acidification. With regard to deposition levels, we consider estimates for both the median and for an upper percentile on the distribution of values at sites analyzed in each ecoregion (e.g., the 90^{th} percentile). In so doing, we recognize that the sites estimated to receive the higher levels of deposition are those most influencing the extent to which the potential objectives for aquatic acidification protection are or are not met. With this in mind, we note the appreciable reduction in the 90^{th} percentile deposition estimates, as well as the median, for REA sites in each of the 25 ecoregions analyzed. Although the ecoregion 90^{th} percentile and median estimates ranged up to 22 and 15 kg/ha-yr in the 2001-2003 time period, both types of estimates fall below approximately 5 to 8 kg/ha-yr by the 2010-2012 period, and below 5 kg/ha-yr in later years (Figure 7-2).

Based on all of the above, including the ecoregion-scale acid buffering objectives identified by the CASAC (more than 70% to 80% of waterbody sites in all ecoregions assessed achieving or exceeding the set of ANC targets), the temporal trends in REA aquatic acidification estimates and the temporal trend in ecoregion S deposition, we estimate that such objectives might be expected to be met when ecoregion median and upper (90^{th}) percentile deposition estimates at sensitive ecoregions are generally at and below about 5 to 8 kg/ha-yr. In so doing, we additionally recognize uncertainties associated with the deposition estimates at individual waterbody sites, and with the associated estimates of aquatic acidification risk, as summarized in section 5.1.4 above. As noted in section 7.2.2.2 above, consideration of the case study analyses as well as the ecoregion-scale results for both the ecoregion-time period and temporal perspectives, indicates a range of S deposition below approximately 5 to 8 or 10 kg/ha-yr, on an areawide basis, to be associated with a potential to achieve acid buffering capacity levels of interest in an appreciable portion of acid sensitive areas. Based on this identification of deposition rates at and below about 5 to 8 or 10 kg/ha-yr, we next consider the information regarding patterns of monitoring site SO_2 concentrations associated with these patterns of S deposition.

In considering options for a standard focused on S deposition, we have focused on an averaging time longer than the three hours of the current standard. In so doing, we recognize, in light of the second maximum form of the existing standard and its relatively short averaging time, that this option might reasonably be considered a less than optimal approach for controlling long-term atmospheric deposition of S compounds (and we note the majority of CASAC advice regarding an annual average metric for this purpose). As discussed in section 7.2.2.3 above, the analyses described in Chapter 6 also indicates moderate to strong correlations for S deposition with an annual air quality metric. Accordingly, we conclude it may be more appropriate to consider adoption of a new SO_2 standard with a longer averaging time and more stable form, as well as level, such as a standard with an averaging time of one year, and a form of the average of annual averages across three consecutive years.[27]

In considering options for an annual secondary standard based on consideration of S deposition-related effects, we first note the complexity of identifying a national ambient air quality standard focused on protection from national patterns of atmospheric deposition of concern to the public welfare (rather than on protection from patterns of ambient air concentrations of concern). For example, atmospheric deposition (ecosystem loading) of S, is, in a simple sense, the product of atmospheric concentrations of S compounds, factors affecting S transfer from air to surfaces, and time. Further, atmospheric concentrations in an ecosystem are, themselves, the result of emissions from multiple, distributed sources (near and far), atmospheric chemistry, and transport. Accordingly, consideration of the location of source emissions and expected pollutant transport (in addition to the influence of physical and chemical processes) is important to understanding relationships between SO_2 concentrations at ambient air monitors and S deposition rates in sensitive ecosystems of interest. Further, we recognize that to achieve a desired level of S deposition control in sensitive ecosystems, SO_2 emissions must be controlled at their sources. Accordingly, it is reasonable to consider surveillance for a secondary standard to be at regulatory SO_2 monitors generally sited near large SO_2 sources.

Recognizing the variation across the U.S. in locations and magnitude of sources of SO_X, as well as the processes that govern that transformation of source emissions to eventual deposition of S compounds, we consider the key findings from the suite of analyses summarized in Chapter 6. These include consideration of relationships between S deposition estimates and SO_2 concentrations near SO_2 monitors (both in remote Class I areas and at NAAQS surveillance monitors which are often near large sources) as well as relationships between ecoregion S deposition estimates and SO_2 concentrations at upwind sites of influence, identified by trajectory

[27] Standards established in the last one to two decades have generally utilized 3-year forms in recognition of the importance of stability in air quality management programs (e.g., 88 FR 3198, January 15, 2013).

analyses to account for the relationship between upwind concentrations near sources and deposition in areas more distant (sections 6.2.2 through 6.2.4, above). As evidence of the influence of SO_2 in ambient air on S deposition, all of these analyses demonstrated there to be an association between SO_2 concentrations and nearby or downwind S deposition. The correlation coefficients are strongest in the East and in the earliest two to three time periods when deposition rates and air concentrations were much higher compared to the West and to more recent years, when deposition rates and concentrations are much lower, as described in Chapter 6.

As discussed in section 7.2.2.3 above, we recognize that the trajectory-based analyses and the stronger correlations for the EAQM-weighted compared to the EAQM-max illustrate the fact that atmospheric loading is a primary determinant of atmospheric deposition, as well as the complexity of how to consider concentrations at individual monitors, with variable spatial distribution, in relation to deposition rates. We additionally consider the parallel temporal trends in SO_2 emissions, annual SO_2 concentrations, and annual average estimates of S deposition over the 20-year time period from 2000-2020 (section 6.2.1). These trends additionally document the expected strong correlation of SO_2 emissions with S deposition. With regard to monitor concentrations, we note the appreciably flatter distribution of concentrations prevalent in the latter 10 years of the period in comparison to the initial years, and take note of the fact that the S deposition rates during this time period are appreciably reduced from those in the earlier decade (Figure 7-5). These parallel patterns indicate the role of the central part of the distribution of U.S. monitor concentrations as a potential influence on the higher deposition levels of the past. The much higher atmospheric loading in the first decade (evidenced by the deposition estimates prior to 2010) is associated with a different distribution of ambient air SO_2 concentrations than in the second decade. The distribution in the first decade is characterized by a more broad or normal distribution, while the distribution in the latter decade is more narrow or skewed. Further, we take note of the parallel temporal trends of ecoregion S deposition estimates and the REA aquatic acidification risk estimate across the five time periods analyzed (as discussed above). With all of these linkages in mind, we have considered what the current information indicates regarding options for a standard that may provide protection from aquatic acidification-related risks of S deposition in sensitive ecoregions.

For an annual average standard, based on the air quality analyses and recognizing the various limitations and associated uncertainties, we identify a range of levels extending down from 15 ppb to a level as low as 5 ppb, based on a recognition of the pattern of ambient air SO_2 concentrations across the U.S. in recent times. As discussed above, the current pattern involves a much compressed distribution of concentrations with the bulk of the distribution well below this range of levels. We additionally recognize that the more recent distribution of concentrations is associated with the more recent deposition patterns and the corresponding aquatic acidification

analysis results as discussed in section 7.2.2.2 above (Figures 7-1 and 7-2 above). The information providing support across this broad range, as discussed in sections 7.2.2.2 and 7.2.2.3, varies. Identification of levels in the upper part of the range, generally from 10-15 ppb places greater weight on an objective of ecoregion median and 90th percentile S deposition values below approximately 5 to 8 kg/ha-yr, and on consideration of the trajectory-based analyses of the 20-year dataset of ecoregion S deposition and SO_2 concentrations (SO_2 annual EAQM-max [Figure 7-4]) at upwind sites of influence, and also on uncertainties associated with potential limitations in the data analyzed (including with regard to representation of source locations in the earlier years). Consideration of potential levels in the lower part of the range, generally from 10 down to 5 ppb, would place greater weight on an objective of ecoregion median and 90th percentile S deposition values below 5 kg/ha-yr, and on consideration of the trend analyses that indicate 3-year average annual SO_2 concentrations since 2010 and 2014 were nearly all below 10 ppb. Given the much reduced correlation of S deposition estimates with SO_2 concentrations in the more recent years (e.g., Table 6-4 above), however, we recognize appreciably greater uncertainty associated with interpretation of relationships between S deposition and ambient air SO_2 concentrations below 10 ppb (and with related conclusions regarding deposition levels that might be expected to be associated with such concentrations) and thus with a potential level in the lower part of the range.

In identifying this broad range of levels for consideration with a new annual average SO_2 secondary standard, we take note of a number of limitations in our information that contribute uncertainties that vary in magnitude and type across this range. In general, we recognize uncertainty in identifying a level within this range for a standard that may be expected to achieve a particular degree of S deposition-related protection for ecological effects. This uncertainty is coupled with the uncertainty associated with estimates of aquatic acidification risk in waterbodies across the U.S. associated with specific deposition levels, including with regard to interpretation of risk associated with different levels of acid buffering capacity. Together, we consider there to be greater uncertainty associated with identification of levels in the lower part of the broad range identified here.

We additionally take note of the advice from the CASAC on options for a secondary standard to provide protection from S deposition-related ecological effects. As described in section 7.3 above, the majority of the CASAC recommended adoption of an annual SO_2 standard with a level within the range of 10 to 15 ppb. In so doing, the CASAC majority noted the ecoregion median deposition levels below 5 kg/ha-yr in the periods 2014-2016 and 2018-2020, and conveyed that a standard level in this range (10-15 ppb) would afford protection to tree and lichen species as well as waterbodies, further stating that such a standard would "preclude the possibility of returning to deleterious deposition values" that these members indicate to be

associated with outlier SO_2 concentrations observed in 2019-2021 near a location of industrial sources (Sheppard, Response to Charge Questions, pp. 24-25). The minority of the CASAC recommended adoption of a new 1-hour secondary standard identical in all respects to the existing primary standard form (Sheppard, 2023, pp. 24-25 and Appendix A).

In considering the CASAC advice on levels for an annual average SO_2 standard, we note that the range we have identified above for the option of a new annual SO_2 standard includes the range of levels (10-15 ppb) recommended by the majority of the CASAC (as summarized in section 7.3 above). We additionally note that, as is generally the case here, the information considered by the CASAC majority in drawing its conclusion also focused on an annual average SO_2 metric with a form that involved averaging over three consecutive years.[28] Further, we note an air quality similarity of the identified range for a new annual average standard with the recommendation of the CASAC minority (to establish a 1-hour secondary standard identical to the primary standard) based on observations regarding the relationship between annual average SO_2 concentrations and design values for the 1-hour primary standard indicating that annual average concentrations are generally at or below 10 ppb in areas meeting the current 1-hour primary standard (Figure 2-29).

We additionally consider the extent of control for short-term concentrations (e.g., of three hours duration) that might be expected to be provided by an annual secondary SO_2 standard. In so doing, we note that in areas and periods when the annual SO_2 concentration (annual average, averaged over three years) is below 5-15 ppb, design values for the existing 3-hour standard are well below the standard level of 0.5 ppm (Figure 2-29). Thus, we note that in considering adoption of a new annual standard, it may be appropriate to consider this as an additional secondary SO_2 standard or to consider it in replacement of the existing 3-hour standard given that peak concentrations are currently controlled to lower concentrations, likely in response to the primary standard. We recognize, however, that which of these options — replacing or augmenting the 3-hour standard (with an annual standard) — is concluded to be appropriate and what value within the ranges of levels identified for an annual standard might be appropriate, are in the end decisions made by the Administrator, in light of judgments associated with weighing of the differing aspects of the evidence and air quality information and how to consider their associated uncertainties and limitations.

Turning to consideration of the secondary standard for oxides of N, we note that the existing secondary standard for oxides of N is 53 ppb, as an annual mean in a single year. The

[28] A 3-year form is common to NAAQS adopted over the more recent past. This form provides a desired stability to the air quality management programs which is considered to contribute to improved public health and welfare protection (e.g., 78 FR 3198, January 15, 2013; 80 FR 65352, October 26, 2015; 85 FR 87267, December 31, 2020).

evidence of welfare effects at the time this standard was established in 1971 indicated the direct effects of N oxides on vegetation, most particularly effects on foliar surfaces. The currently available information continues to document such effects, as summarized in sections 4.1 and 5.4.2 above. With regard to NO_2 and NO, the evidence does not indicate effects associated with ambient air concentrations allowed by the existing standard, as summarized in section 7.1.2 above. Accordingly, the evidence related to the N oxides, NO_2 and NO, does not call into question the adequacy of protection provided by the existing standard.

With regard to the N oxide, HNO_3, however, we recognize the evidence of effects associated with air concentrations and associated HNO_3 dry deposition on plant and lichen surfaces, and that there is uncertainty as to the extent to which exposures associated with such effects may be allowed by the existing NO_2 standard, as discussed in section 7.1.2 above (section 5.4.2 and Appendix 5B, sections 5B.4). The limited evidence, however, is not clear as to the potential for such effects to have been elicited by air quality that met the standard. Thus, the available information — while documenting the potential for HNO_3 in ambient air to cause harm — is not clear as to the extent to which it may call into question or support the adequacy of protection provided by the current NO_2 standard. The experimental evidence also does not provide clear indication of ecological effects associated with exposure concentrations that might be allowed by the current standard. We note, however, that depending on judgments as to the weight to place on specific aspects of the evidence and air quality analyses, and associated uncertainties, it may be judged appropriate to consider a more restrictive NO_2 standard that might also be considered to offer the potential for some additional protection from effects related to ecosystem N deposition (as discussed below), and also the potential for increased protection from effects related to airborne nitric acid effects on biota surfaces for which the quantitative evidence is less clear. With regard to the latter, we take note of the relatively high dry deposition velocity of HNO_3, relative to other N-containing compounds and the evidence from field surveys indicating its potential for damage (section 7.1.2). Accordingly, in addition to concluding it is appropriate to consider retaining the existing NO_2 standard, we additionally identify a revision option for the secondary standard for N oxides in consideration of HNO_3-related effects in combination with consideration of ecosystem deposition-related effects discussed below.

In considering options for revision of the secondary standard for N oxides, we have also evaluated the larger information base of effects related to N deposition in ecosystems. In this context, we recognize that ecosystem N deposition is influenced by air pollutants other than N oxides. More specifically, as discussed in sections 6.1 and 6.2.1 above, NH_3 (which is not a CAA criteria pollutant) also contributes to N deposition. The extent of this contribution varies appreciably across the U.S. and has increased during the past 20 years. Thus, we take note of the

fact that a secondary standard for N oxides cannot be expected to effectively control total N deposition.

With regard to N deposition associated with N oxides, the historical trend analyses in section 6.2.1 document the reductions in N deposition that correspond with reductions in emissions of N oxides. These analyses additionally document the increasing role of NH_3 in N deposition since approximately 2010 and the co-occurring tempering of N deposition reductions such that the declining trend that is observed from 2000 through 2010 appears to have leveled off in the more recent years. Further, the areas of highest N deposition appear to correspond to the areas with the greatest deposition of NH_3 (Figure 7-8 above). This associated lessening influence of N oxides on total N deposition is also evidenced by the poor correlations between N deposition and annual average NO_2 concentrations (reported in sections 6.2.3 and 6.2.4 above), most particularly in more recent years and at eastern sites. It may be the result of increasing emissions of NH_3 in more recent years and at eastern sites (section 2.2.3 and Figure 6-5). Together, this finding, particularly since 2010 (and in more localized areas prior to that), complicates our evaluation of the current information with regard to protection from N deposition-related effects that might be afforded by the secondary standard for N oxides. That is, while the information regarding recent rates of ecoregion N deposition may in some individual areas (particularly those for which reduced N, specifically NH_3, has a larger role) indicate rates greater than the range of values identified above for consideration (e.g., 7-12 kg/ha-yr based on the considerations in section 7.2.3 and the benchmark of 10 kg/ha-yr, as conveyed in the advice from the CASAC), the extent to which this occurrence relates to the existing NO_2 secondary standard is unclear.

That notwithstanding, we additionally consider the currently available information related to deposition-related effects of N oxides on ecosystems, as discussed in section 7.2.3 above. In so doing, we recognize the complexities and challenges associated with quantitative characterization of N enrichment-related effects in terrestrial or aquatic ecosystems across the U.S. that might be expected to occur due to specific rates of atmospheric deposition of N over prolonged periods, and the associated uncertainties. Some complexities associated with terrestrial deposition are similar to those for aquatic deposition, such as untangling the impacts of historic deposition from what might be expected from specific annual deposition rates absent that history, while others related to available quantitative information and analyses differ. Further, with regard to many aquatic systems with non-air contributing sources, we recognize the complexity of estimating the portion of N inputs, and associated contribution to effects, derived from atmospheric sources.

Additionally, there are complexities in risk management and policy decisions, including with regard to identifying risk management targets or objectives for an ecosystem stressor like N

enrichment, for which as the CASAC recognized, in terrestrial systems, there are both "benefits and disbenefits" (Sheppard, 2023, p. 8). As noted by the CASAC, "[b]enefits include fertilization of crops and trees and the potential for improved sequestration of carbon in soils and plant biomass" (Sheppard, 2023, p. 8). This also complicates conclusions regarding the extent to which some ecological effects may be judged adverse to the public welfare. Further, with regard to aquatic systems, identification of appropriate risk management targets or objectives for consideration of the relative protection of secondary standards is complicated by the effects of historical deposition that have influenced the current status of soils, surface waters, associated biota, and ecosystem structure and function. For example, changes to ecosystems that have resulted from past, appreciably higher levels of atmospheric deposition have the potential to affect how the ecosystem responds to current, lower levels of deposition or to still further reduced N inputs in the future.

In exploring the potential for a secondary standard to limit N deposition associated with N oxides, we take note of the trends of ecoregion N deposition which differ for ecoregions in which N deposition is driven by reduced N compared to those where reduced N comprises less of the total (e.g., Figures 7-6 and 7-7). The N deposition trends in the latter ecoregions, which include reductions in the upper part of the distribution of ecoregion medians, as well as lower N deposition in the second as compared to the first decade of the 20-year period (corresponding to the decline in NO_2 emissions), appear to document the influence that NO_2 emissions and concentrations have had on N deposition. In light of this relationship and of the recognition that recent levels of N deposition associated with N oxides are much lower than they were in the early part of the 20-year period, we consider the option of a revision of the existing NO_2 standard level to maintain some associated protection from deposition-related effects of N oxides.

With regard to this option, we note the mixed advice from the CASAC regarding an NO_2 annual standard in consideration of N deposition effects (section 7.3 above). The CASAC majority recommended revision of the existing annual NO_2 standard level to a value below 10 to 20 ppb (Sheppard, 2023, p. 24). As described in section 7.3 above, however, the basis for this advice relates to a graph in the draft PA of the dataset of results from the trajectory-based analyses for the weighted annual NO_2 metric (annual NO_2 EAQM-weighted). These CASAC members additionally recognized that these results found no correlation between the ecoregion deposition and the EAQM-weighted values at upwind locations, and as described in section 6.2.4.3 above the correlation coefficients are negative for N deposition with both annual NO_2 EAQMs (-0.17 and -0.06; Table 6-10). While the correlation for the eastern ecoregions and the weighted metric is as high as 0.61 in the 2001-2003 period, it declines for each subsequent time period, and is negative for the most recent period. Further, as noted in section 7.2.2.3 above, the weighted metric values from the trajectory-based analyses are not directly translatable to

individual monitor concentrations or to potential standard levels. Accordingly, the information highlighted by these members for relating N deposition levels to ambient air concentrations cannot reasonably be concluded to provide support for the identified levels. The minority CASAC member recommended revision of the secondary NO_2 standard to be identical to the primary standard based on their conclusion that the recent N deposition levels meet desired targets and that the primary standard is currently the controlling standard (Sheppard, 2023, Appendix A).

The air quality information regarding annual average NO_2 concentrations at SLAMS monitors indicates more recent NO_2 concentrations are well below the existing standard level of 53 ppb. As noted in section 7.2.3.3 above, the temporal trend figures indicate that, subsequent to 2011-2012, when median N deposition levels in 95% of the eastern ecoregions of the continental U.S. have generally been at/below 11 kg N/ha-yr, annual average NO_2 concentrations, averaged across three years, have been at/below 35 ppb. Recognizing that among the NO_2 primary and secondary NAAQS, the 1-hour primary standard (established in 2010) may currently be the controlling standard for ambient air concentrations, we note that annual average NO_2 concentrations, averaged over three years, in areas that meet the current 1-hour primary standard have generally been below approximately 35 to 40 ppb. We note that an annual standard with a level within this range would appear to have conceptual consistency with the advice from the CASAC minority. Thus, for an option to reflect the recent pattern in NO_2 concentrations, and any associated influence on N deposition, as well as to provide additional protection from HNO_3-related effects that may be associated with higher NO_2 concentrations, it may be appropriate to consider an option for revision of the secondary NO_2 standard to an annual standard (averaged across three consecutive years) with a level below the current level of 53 ppb, within a range extending down to 40-35 ppb.

While characterization of such an option as providing <u>some</u> level of protection from N deposition related to N oxides is supported by the quantitative air quality analyses and information regarding air quality and atmospheric chemistry (as discussed in chapter 6), and accordingly, such a standard might be expected to provide some degree of protection from deposition related effects associated with N oxides, we recognize significant uncertainty in understanding the level of protection that would be provided. In addition to the complexity associated with a judgment on the appropriate target level of protection for a national standard for nitrogen, given its contribution to benefits and disbenefits, as well as its multiple sources other than atmospheric deposition (discussed in section 7.2.3 above), this uncertainty relates prominently to the influence of NH_3 on total N deposition separate from that of N oxides, and which in some areas of the U.S. appears to be dominant (as discussed in section 7.2.3.3 above). Further, the extent to which the relative roles of these two pollutants (N oxides and NH_3) may

change in the future is not known. These factors together affect the extent of support for, and contribute significant uncertainty to, a judgment as to a level of N oxides in ambient air that might be expected to provide requisite protection from N deposition-related effects on the public welfare. Thus, the revision option identified here would involve several judgments on the weighing of information and associated uncertainties in several areas. These areas include, but are not limited to the extent to which effects related to HNO_3 may be expected to occur as a result of NO_2 concentrations above the existing standard, and the public welfare significance of such effects; the extent to which a lower annual NO_2 standard could be expected to affect total N deposition across the U.S.; and, the extent of the evidence related to welfare effects associated with deposition related specifically to N oxides. Accordingly, while an option for revision has been identified, in light of considerations raised above, the support for this option is not strong.

Lastly, we turn to consideration of the existing standards for $PM_{2.5}$. As an initial matter, and in light of the discussion in section 7.1.3 above, we do not find the available information to call into question the adequacy of protection afforded by the secondary $PM_{2.5}$ standards from direct effects and deposition of pollutants other than S and N compounds. The evidence indicates such effects to be associated with conditions associated with concentrations much higher than the existing standards.

Regarding S deposition, we note the findings of the air quality analyses in Chapter 6 that indicate appreciable variation in associations between S deposition and $PM_{2.5}$, and generally low correlations and also note the varying composition of PM at sites across the U.S. which may be a factor in the variability in associations. We additionally take note of the atmospheric chemistry which indicates the dependency of S deposition on airborne SO_X, as evidenced by the parallel trends of SO_2 emissions and S deposition. Based on all of these considerations, we find that protection of sensitive ecosystems from S deposition may be more effectively achieved through a revised SO_2 standard than a standard for PM.

With regard to N deposition, as discussed in section 7.2.3.3 above, and in more detail in Chapter 6, air quality analyses of relationships found low to barely moderate correlations between N deposition estimates and annual average $PM_{2.5}$ concentrations at nearby or upwind locations based on the full 20-year dataset, with higher correlations for the early years of the 20-year period and low or no correlation in the later years. We also note the variable composition of $PM_{2.5}$ across the U.S. which contributes to geographic variability in the relationship between N deposition and $PM_{2.5}$ concentrations. For example, as discussed in section 6.4.2, an appreciable percentage of $PM_{2.5}$ mass does not contribute to N deposition, and the highest percentage of $PM_{2.5}$ represented by N compounds at CSN sites in 2020-2022 is 30% (Riverside County, CA). In fact, at an appreciable number of CSN sites, the fraction of $PM_{2.5}$ represented by N compounds is less than 10%. This variability in percentage of $PM_{2.5}$ represented by N (or S)

containing pollutants contributes a high level of uncertainty to our understanding of the potential effect of a $PM_{2.5}$ standard on patterns of N deposition. In light of these considerations and the conclusions above regarding potential for control of S and N deposition from SO_X and N oxides standards, we conclude that the available evidence, as evaluated in this PA, is reasonably judged insufficient to provide a basis for revising the $PM_{2.5}$ annual standard with regard to effects of S and N deposition related to PM.

With regard to options for the annual $PM_{2.5}$ standard, we note that the CASAC did not reach consensus, and provided two sets of recommendations for a revised annual $PM_{2.5}$ standard (section 7.3 above). The CASAC majority recommended revision of the standard level to a value within the range from 6 to 10 $\mu g/m^3$, although we note that the specific rationale for the ends of this range is unclear. The justification provided includes observations regarding annual average $PM_{2.5}$ concentrations in locations for which total N (and S) deposition falls within, and falls above, the preferred deposition ranges identified (Sheppard, 2023, pp. 23-24). For example, the range of annual average $PM_{2.5}$ concentrations these members identify to be associated with deposition within their preferred N deposition range ($PM_{2.5}$ concentrations from 2 to 8 $\mu g/m^3$ and total N deposition at/below 10 kg/ha-yr, based on draft PA graphs of 2014-16 and 2017-19 values) overlaps with the concentration range they identify as being associated with deposition above that range ($PM_{2.5}$ concentrations from 6 to 12 $\mu g/m^3$ and total N deposition above 15 kg/ha-yr in "hotspots" of California, the Midwest and the East, based on draft PA maps depicting 2019-21 deposition estimates and annual $PM_{2.5}$ design values).[29] We note that this overlap indicates a weakness in the associations of N deposition with PM concentrations (and scatter in the dataset) in some areas of the U.S.[30] Further, the expanded air quality analyses in this final PA indicate only low correlation for total N deposition estimates with annual average $PM_{2.5}$ design values in the last 10 years (e.g., r values are less than 0.40 for 2014-16 and 2018-20 at SLAMS [Table 6-7]). Among other factors, this reduction in correlation may relate to the reduced presence of N compounds in $PM_{2.5}$ mass in the more recent period, as discussed in section 6.4.2 above. In total, we take note of the appreciable uncertainty regarding relationships of N (and S) deposition with $PM_{2.5}$ concentrations across the U.S. The minority CASAC member recommended revision of the secondary annual $PM_{2.5}$ standard level to equal the primary

[29] For example, the justification provided for the range of levels recommended by the CASAC majority for a revised $PM_{2.5}$ annual standard (6 to 10 $\mu g/m^3$) refers both to annual average $PM_{2.5}$ concentrations (3-yr averages) ranging from 2 to 8 $\mu g/m^3$ in 27 Class I areas (as corresponding to N deposition estimates at or below 10 kg/ha-yr) and to annual average $PM_{2.5}$ concentrations (3-year averages) ranging from 6 to 12 $\mu g/m^3$ (at design value sites in areas of N deposition estimates greater than 15 kg/ha-yr), as summarized in section 7.3 above.

[30] As discussed in section 6.2.1 above, these areas of highest N deposition estimates coincide with areas of the U.S. in which NH_3 deposition is also the highest (Figure 6-13, bottom and Figure 6-18, bottom), and also where NH_3 deposition is estimated to comprise the majority of total N deposition (Figure 7-8; 7.2.3.3).

standard level of 12 μg/m^3 based on their conclusion that the recent N (and S) deposition levels meet desired targets and that the primary annual $PM_{2.5}$ standard is currently the controlling standard for annual $PM_{2.5}$ concentrations (Sheppard, 2023, Appendix A).

Although we recognize there to be appreciable uncertainty associated with a basis for a revised annual $PM_{2.5}$ standard related to effects of S and N deposition related to PM, as discussed above, we also recognize that decisions on the NAAQS also draw on judgements with regard to the weight to place on various uncertainties, and so, in light of all the considerations described above, and based on the air quality information that suggests some low to moderate correlation of N-deposition with the annual $PM_{2.5}$ metric, that is stronger in the West (as summarized in section 7.2.3.3 above), we suggest that it may be appropriate to consider some revision of the level of the $PM_{2.5}$ annual secondary standard. For this option, it may be appropriate to consider levels below the current level of 15 μg/m^3, such as a level of 12 μg/m^3 (the level of the currently controlling primary standard), recognizing uncertainty with regard to the extent of N deposition-related control and associated protection that might be achieved. In so doing, we note that this option is that recommended by the CASAC minority.

With regard to other PM standards, we take note of the lack of information that would call into question the adequacy of protection afforded by the existing PM_{10} secondary standard for ecological effects, and thus conclude it is appropriate to consider retaining this standard without revision. As to the 24-hour $PM_{2.5}$ standard, we note the advice of the majority of CASAC, summarized in section 7.3 above, with regard to revision of this standard to a lower level or to an indicator of deciviews. In conveying these recommendations, these CASAC members generally expressed the view that the existing standard was not adequate to protect against short-term events. In justifying this view, the members make the general statements that there are "seasonal variabilities" in "ecological sensitivities," and that sensitive lichen species are dependent on fog or cloud water-related deposition, in which the members state S and N contributions can be highly episodic. These members do not, however, provide further specificity regarding the basis for these references to lichen species and fog or cloud water. While the available evidence as characterized in the ISA recognizes there to be N deposition associated with cloud water or fog (ISA, Appendix 2), it does not provide estimates of this deposition or describe associated temporal variability, or specifically describe related effects on biota. Thus, we do not find that the evidence available in this review, as documented in the ISA, or cited by the CASAC, calls into question the adequacy of protection provided by the 24-hour $PM_{2.5}$ standard from ecological effects. Further, with regard to their specific revision recommendations for a revised level or indicator of the 24-hour $PM_{2.5}$ secondary standard, the CASAC members cite discussion in the January 2023 proposal to revise the $PM_{2.5}$ secondary standard to protect against visibility effects. We note that considerations as to the adequacy of protection provided

by the $PM_{2.5}$ standard from visibility effects are being addressed in the review of the PM NAAQS Reconsideration (88 FR 5558, January 27, 2023), and are not included in the review that is the subject of this PA.

In summary, based on the current evidence and quantitative air quality, exposure and risk information, with associated limitations and uncertainties, in light of all of the considerations above, we identify several options that may be appropriate for the Administrator to consider. The potential policy options that could inform the Administrator's decisions on the NAAQS providing the "requisite" public welfare protection and that are supported by the science include both options to address protection for direct effects of the pollutants in ambient air and options to address protection for effects related to S deposition and to N deposition. A summary of these options is shown in Table 7-3 and described below.

To address protection of the public welfare from effects of SO_X in ambient air, we recognize options appropriate to consider for protection from both direct and deposition-related effects. With regard to protection against the direct effects of SO_X in ambient air, we conclude it is appropriate to consider retaining the current secondary standard. To address protection of the public welfare from effects related to S deposition, we conclude it is appropriate to consider adoption of a new annual SO_2 standard. This option involves establishing a SO_2 annual mean standard, averaged across three years, with a level within the range of levels extending below 15 to 5 ppb. In light of the extent to which peak concentrations (e.g., 3-hour averages) may be otherwise controlled as discussed above, it may also be appropriate to consider adoption of such an annual standard as a replacement for the current 3-hour standard.

With regard to protection from effects of N oxides and/or PM and N deposition, three options are identified in consideration of: limitations in the available evidence, and associated uncertainties related to interpretation of the evidence and air quality information; relationships between the two pollutants and associated effects; and connections of effects elicited by N oxides in ambient air and deposited onto biota surfaces. One option is to retain the existing NO_2 and PM standards, based on the judgment that the current evidence does not call into question the adequacy of protection of the public welfare from both direct effects of N oxides and PM in ambient air and effects related to N deposition associated with these pollutants. To the extent different judgments are made, two options for revision are also identified that might be appropriate to consider with regard to both protection from direct effects of N oxides in ambient air and some increased protection from N deposition associated with N oxides and PM.

The option to retain the existing NO_2 and PM standards is based on judgments that the evidence for direct effects of N oxides and PM does not call into question the adequacy of protection provided by these standards and also judgments that weigh heavily the limitations and associated uncertainties associated with the available information. These limitations and

associated uncertainties relate to the evidence base for ecosystem effects related to N deposition associated with N oxides and PM, and with the air quality information related to the limited potential for control of N deposition in areas across the U.S., in light of variation in the composition of both oxides of N and of PM. The first set of limitations and uncertainties relates to quantitative relationships between N deposition and ecosystem effects, based on which differing judgments may be made in decisions regarding protection of the public welfare. In the case of protection of the public welfare from adverse effects associated with nutrient enrichment, we additionally recognize the complexity associated with identification of appropriate protection objectives in the context of changing conditions in aquatic and terrestrial systems as recent deposition has declined from the historical rates of loading. The second set of limitations and uncertainties relates to relatively lower correlations in more recent time periods of air quality metrics for N oxides with N deposition in ecosystems and the variation in PM composition across the U.S., particularly that between the eastern and western U.S. This latter set of limitations is considered to relate to the emergence of NH_3 as a greater influence on N deposition than N oxides and PM over the more recent years. Further, this influence appears to be exerted in areas with some of the highest N deposition estimates for those years.

For N oxides, the options of retention or revision of the existing standard are based on consideration of the air quality information that suggests control of N-deposition associated with N oxides with an annual NO_2 standard, and taking into account limitations in the available evidence and associated uncertainties related to interpretation of the evidence of terrestrial biota effects of nitric acid, which may be the direct effects most sensitive to oxides of N in ambient air. We note that such effects may be considered to be both direct effects and also deposition-related effects as they relate to direct contact with biota surfaces by dry deposition (e.g., ISA, Appendix 3, section 3.4, Appendix 5, section 5.2.3 and Appendix 6, section 6.3.7). The options, as described earlier in this section, include retaining or revising the current secondary NO_2 standard. For the revision option, it may be appropriate to consider levels below 53 ppb and extending down to approximately 40-35 ppb.

With regard to the annual $PM_{2.5}$ secondary standard, based on the air quality information that suggests some correlation of N-deposition with the annual $PM_{2.5}$ metric, which is stronger in the West, it may be appropriate to consider revision of the level of the $PM_{2.5}$ annual secondary standard. For this option, it may be appropriate to consider levels below the current level of 15 $\mu g/m^3$, such as a level of 12 $\mu g/m^3$ (the level of the currently controlling primary standard), recognizing uncertainty with regard to the extent of N deposition-related control and associated protection that might be achieved. We note that this option is that recommended by the CASAC minority.

In addition to the options identified above, we recognize the potential value in consideration of a standard or suite of standards with alternate indicator(s) that may target specific chemicals that deposit N and S (e.g., NO_3^-, SO_4^{2-}, NH_4^+). In so doing, however, we note a number of information gaps that would need to be filled to inform identification of specific options of this type. One example relates to the depth of our understanding of the distribution of these chemicals in ambient air, including relationships between concentrations near sources and in areas of deposition, such as protected areas. In this context we recognize that, depending on the indicator selected, the relationship exhibited between concentrations of the indicator and N or S deposition at the same location may not be expected to hold for concentrations of the indicator in more distant locations, including locations near emissions sources. Additionally, we recognize the practical considerations associated with establishing new standards with new indicators related to establishment of regulatory measurement methods and surveillance networks, that would yield effective implementation of the standards. Thus, while we note the potential value in such approaches, as also recognized by the CASAC, we also recognize the additional data collection and analysis needed to develop a foundation that might support their adoption.

We additionally note that the Administrator's decisions regarding secondary standards, in general, are largely public welfare judgments, as described above. We note that different public welfare policy judgments could lead to different conclusions regarding the extent to which the current and various alternative standards might be expected to provide the requisite protection of the public welfare. Such public welfare judgments include those related to identification of effects of public welfare significance, as well as with regard to the appropriate weight to be given to differing aspects of the evidence and air quality information, and how to consider their associated uncertainties and limitations. For example, different judgments might give greater weight to more uncertain aspects of the evidence. There are, additionally, judgments with regard to the appropriate objectives for the requisite protection of the public welfare. Such judgments are left to the discretion of the Administrator. Thus, in identifying a broad array of options for consideration above (summarized in Table 7-3 below), we also note that decisions on the approach to take in achieving the desired air quality and public welfare protection fall within the scope of the Administrator's judgment.

Table 7-3. Summary of current standards and range of potential policy options for consideration.

Current Secondary Standards					
Pollutant	**Indicator**	**Averaging Time**	**Level**	**Form**	**Basis**
SO_X	SO_2	3 hours	0.5 ppm	Not to be exceeded more than once per year	Direct effects on vegetation
N Oxides	NO_2	1 year	53 ppb	Annual	Direct effects on vegetation
PM	$PM_{2.5}$	1 year	15 μg/m^3	Annual, averaged over three years	Ecological effects related to deposition, as well as effects on visibility and climate, and materials damage (with only the former considered in this review)
		24 hours	35 μg/m^3	98th percentile, averaged over three years	
	PM_{10}	24 hours	150 μg/m^3	Not to be exceeded more than once per year on average over three years	
Policy Options for Consideration: For protection from both direct effects of the pollutants on biota and from ecological effects of ecosystem deposition of N and S associated with the pollutants.					
SO_X	Adoption of an annual average SO_2 standard, averaged over three years, with a level within the range of levels below 15 ppb down to 5 ppb, and retention of the existing 3-hour SO_2 standard Or Replacement of existing 3-hour standard with an annual average SO_2 standard based on air quality data indicating annual standard to also provide the pertinent control for short-term concentrations.				
N Oxides	Retention of the existing annual NO_2 standard Or Revision of the level of the existing standard to within a range below 53 ppb to as low as 40-35 ppb, in combination with consideration of a form averaged over three years				
PM	Retention of the existing suite of standards Or Revision of the current annual $PM_{2.5}$ standard level to within a range below 15 μg/m^3 to that of the current primary standard (12 μg/m^3)				
Potential Options for Consideration in Future Reviews					
SO_X, N Oxides and PM	Alternate indicator(s)	The potential for establishment of a revised standard or suite of standards with alternate indicator(s) that may target specific chemicals that deposit N and S (e.g., particulate NO_3^-, SO_4^{2-}, NH_4^+) is associated with a number of uncertainties and complications that include uncertainties in relationships between concentrations near sources and in areas of deposition, as well as complications related to establishment of measurement methods and design of regulatory monitoring networks.			

7.5 AREAS FOR FUTURE RESEARCH RELATED TO KEY UNCERTAINTIES

In this section, we highlight several key uncertainties associated with reviewing and establishing the secondary standards for SO_X, oxides of N and PM, and additionally recognize that research in these areas, and perhaps others not highlighted here, may additionally be informative to the development of more efficient and effective control strategies. Accordingly, areas highlighted for future welfare effects and atmospheric chemistry research include model development, and data collection activities to address key uncertainties and limitations in the current scientific evidence. These areas are similar to those highlighted in past reviews, such as those that follow:

- Data and tools to relate concentrations of specific pollutants in ambient air with deposition. This could include expansion of existing monitoring networks (either in number or in the number of pollutants measured) to enable more geographically representative comparisons of local deposition and local air quality concentrations.
- Research to further develop and improve modeling tools that relate atmospheric deposition of specific compounds to changes in soil conditions, which influence watershed aquatic impacts as well as effects on resident vegetation, in areas characterized by different soil types and geology.
- Improved understanding of the relationship between wildfires and deposition of SO_X, N oxides and PM.
- Continued refinement of the TDep methodology to estimate national total deposition. This could include efforts to continually evaluate and improve the air quality model simulation inputs to TDep.
- Additional work to improve accuracy of estimates of BCw, a critical parameter in modeling to characterize risks associated with aquatic and terrestrial acidification.
- To address uncertainty associated with characterizing risks associated with terrestrial acidification, additional research might contribute to an improved understanding of effects on sensitive vegetation of various levels of BC:Al in different soil types.
- Improved understanding of relationships between soil N and carbon to N ratios, as indicator metrics, and effects on key ecological receptors.
- Although addition or exposure studies are somewhat limited, studies assessing important tree species included in Horn et al. 2018 would help improve confidence.
- Research to improve understanding of the linkages between deposition, geochemical metrics and ecological effects of freshwater ecosystem eutrophication. Currently available studies of waterbodies in the western U.S. have included investigations of nutrient limitation and diatom assemblages. Studies in eastern lakes and streams have primarily focused on NO_3^- leaching. Information is limited for relationships between additional ecological endpoints (e.g., effects on fish and invertebrate communities) and NO_3^- concentrations (or other chemical indicators).

- Research relating specific indicators of acidification or nutrient enrichment to ecological effects and to ecosystem services (e.g., fish harvest, recreation, etc).
- Research to address key limitations and uncertainties in modeling watershed N loading, including atmospheric deposition to indicators of eutrophication (e.g., disolved oxygen and chlorophyll A). For example, data to better estimate estuary-specific parameters (e.g., as used in Evans and Scavia Model); improved modeling tools that combine watershed loading and influence on estuarine indicators.
- Information is limited relating N deposition to specific endpoints in wetlands. Additional research would contribute to an improved understanding of relationships between N deposition and chemical and ecological responses across a range of wetland types and across geographic regions.
- Regarding aquatic eutrophication, research in several areas would advance assessment approaches. These include research on appropriate endpoints or indicators; important mediating factors (e.g., drought, temperatures, seasonality, dissolved organic carbon, recovery from acidification) and characterization of their role in key processes, as well as on the extent of differences among N compounds with regard to their role in key processes.

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APPENDICES

APPENDIX 5A

RISK AND EXPOSURE ASSESSMENT FOR AQUATIC ACIDIFICATION

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5A.1 AQUATIC ACIDIFICATION AND OVERVIEW OF ANALYSES

Air emissions of sulfur oxides (SO_X), oxides of nitrogen, and reduced forms of nitrogen (NH_X) react in the atmosphere through a complex mix of reactions and thermodynamic processes in gaseous, liquid, and solid phases to form various acidifying compounds. These compounds are removed from the atmosphere through wet (e.g., rain, snow), cloud and fog, or dry (e.g., gases, particles) deposition. Deposition of SO_X, oxides of nitrogen, and NH_X leads to ecosystem exposure to acidification. The 2020 ISA concludes that the body of evidence is sufficient to infer a causal relationship between acidifying deposition and adverse changes in freshwater biota (see ISA, Appendix 8). Freshwater systems of the U.S. include lakes, rivers, streams, and wetlands. Changes in biogeochemical processes and water chemistry caused by deposition of nitrogen (N) and sulfur (S) to surface waters and their watersheds have been well characterized for decades and have ramifications for biological functioning of freshwater ecosystems.

When S or N deposition leaches from soils to surface waters in the form of sulfate (SO_4^{2-}) or nitrate (NO_3^-), an equivalent number of positive cations, or countercharge, is also transported. This maintains electroneutrality. If the countercharge is provided by base cations such as calcium (Ca^{2+}), magnesium (Mg^{2+}), sodium (Na^+), or potassium (K^+), rather than hydrogen (H^+) and aluminum (Al_3^+), the acidity of the soil water is neutralized, but the base saturation of the soil is reduced. Continued SO_4^{2-} and/or NO_3^- leaching can deplete available base cation pools in the soil. As the base cations are removed, continued deposition and leaching of SO_4^{2-} and/or NO_3^- - (with H^+ and Al_3^+) leads to acidification of soil water, and by connection, surface water. Loss of soil base saturation is a cumulative effect that increases the sensitivity of the watershed to further acidifying deposition.

These chemical changes in water quality can occur over both long- and short-term timescales. Short-term (i.e., hours or days), often termed episodic, periods of increased acidity can also have significant biological effects. Episodic chemistry refers to conditions during precipitation or snowmelt events when proportionately more drainage water is routed through upper soil horizons that tends to provide less acid neutralizing than deeper soil horizons. Surface water chemistry has lower pH and acid neutralizing capacity (ANC) during these events than during baseflow conditions. Acid neutralizing capacity is a water quality measurement of a waterbody's ability to neutralize acid inputs or its "buffering capacity against acidification" (ISA, p. ES-14). Models often simulate calculated ANC, e.g., as the difference between the total amount of strong base ions (sum of base cations, SBC) and the total amount of strong acid anions

(sum of acid anions, SAA).[1] In this assessment, the calculation is performed as in equation. 5A-1):

$$ANC = SBC - SAA = (Ca^{2+} + Mg^{2+} + K^{+} + Na^{+} + NH_4^{+}) - (SO_4^{2-} + NO_3^{-} + Cl^{-}) \quad (5A\text{-}1)$$

Acid neutralizing capacity and pH are related to one another as they both are measures of acidity in surface waters and low pH values correspond to low ANC values. However, pH in natural waters is dependent on the amount of carbon dioxide, organic acids, and aluminum solubility, which impacts the relationships between the two parameters. The amount of carbon dioxide (CO_2) dissolved in surface waters is affected by biological activity and temperature, which decreases pH but does not impact ANC. Dissolved organic carbon (DOC), which includes organic acids (e.g., fulvic and humic acids, carboxylic acids, and amino acids), also lowers pH values in surface waters and changes the relationship between pH and ANC (ISA, Appendix 4, section 4.3.9).

The principal factor governing the sensitivity of aquatic ecosystems to acidification from acidifying deposition is geology (particularly surficial geology; [Greaver et al., 2012]). Levels of acidifying deposition are generally low in the western contiguous U.S. (western CONUS) but can be higher in the eastern CONUS (ISA Appendix 8, section 8.5.1). In the eastern CONUS, acid-sensitive ecosystems are generally located in upland, mountainous terrain underlain by weathering resistant bedrock. Surface waters most sensitive to acidification are largely found in the Northeast, southern Appalachian Mountains, Florida, the Upper Midwest, and the mountainous West. (ISA, Appendix 8, section 8.5.1).

Acidification of freshwater ecosystems occurs in response to either N or S deposition alone or in combination. This is because both N and S deposition can act as acidifying agents. The effects of acidifying deposition on biogeochemical processes in soils have ramifications for the water chemistry and biological functioning of associated surface waters. Surface water chemistry integrates direct air-to-water deposition with deposition impacts on soil chemistry of hydrologically connected terrestrial ecosystems within the watershed (ISA, Appendices 4, 7 and 8). Acid-sensitive freshwater systems can either be chronically acidified or subject to occasional episodes of decreased pH, decreased ANC, and increased inorganic Al concentration (ISA, Appendix 7, section 7.1).

In this assessment, the impact of N and/or S deposition on aquatic acidification was evaluated using a critical load (CL) approach. This CL approach provides a means of gauging whether a group of lakes, streams, and rivers (i.e., waterbodies) in each area receives a level of N

[1] The two measures (measured or titrated ANC and calculated ANC) can differ greatly, depending mainly on the amount of organic acidity and dissolved Al in the water (ISA, Appendix 7, p. 7-23).

and/or S deposition that corresponds to that associated with a specified value for the water quality metric used as indicator of acidification. For this analysis, ANC was used as the indicator, with target levels identified to correspond to different levels of acidification-related risk to biota. Depending on the ANC target, low CL values may mean that the watershed has a limited ability to neutralize the addition of acidic anions and, hence, is susceptible to acidification. The greater the CL value, the greater the ability of the watershed to neutralize additional acidic anions.

5A.1.1 Analysis Scales

A multi-scale analysis was completed that assessed aquatic acidification at three levels of spatial extent: national, ecoregion, and case study (Figure 5A-1). The national-scale assessment focused within the contiguous U.S. (CONUS) due to insufficient availability data for Hawaii, Alaska, and the territories. The Omernik ecoregion classifications (level III) were used for the ecoregion-scale analyses. Case study locations were areas likely to be most impacted and for which sufficient data were available. Further discussion of these spatial scales can be found below. Since acidification of waterbodies is controlled by local factors such as geology, hydrology, etc. the aquatic CLs for acidification are unique to the waterbody itself and information about the waterbody, like water quality, is needed to determine its critical load. For these reasons, CLs were determined at the waterbody level and then summarized at the national, ecoregion, and case study level. The national assessment is a combined summary of aquatic CLs across the CONUS.

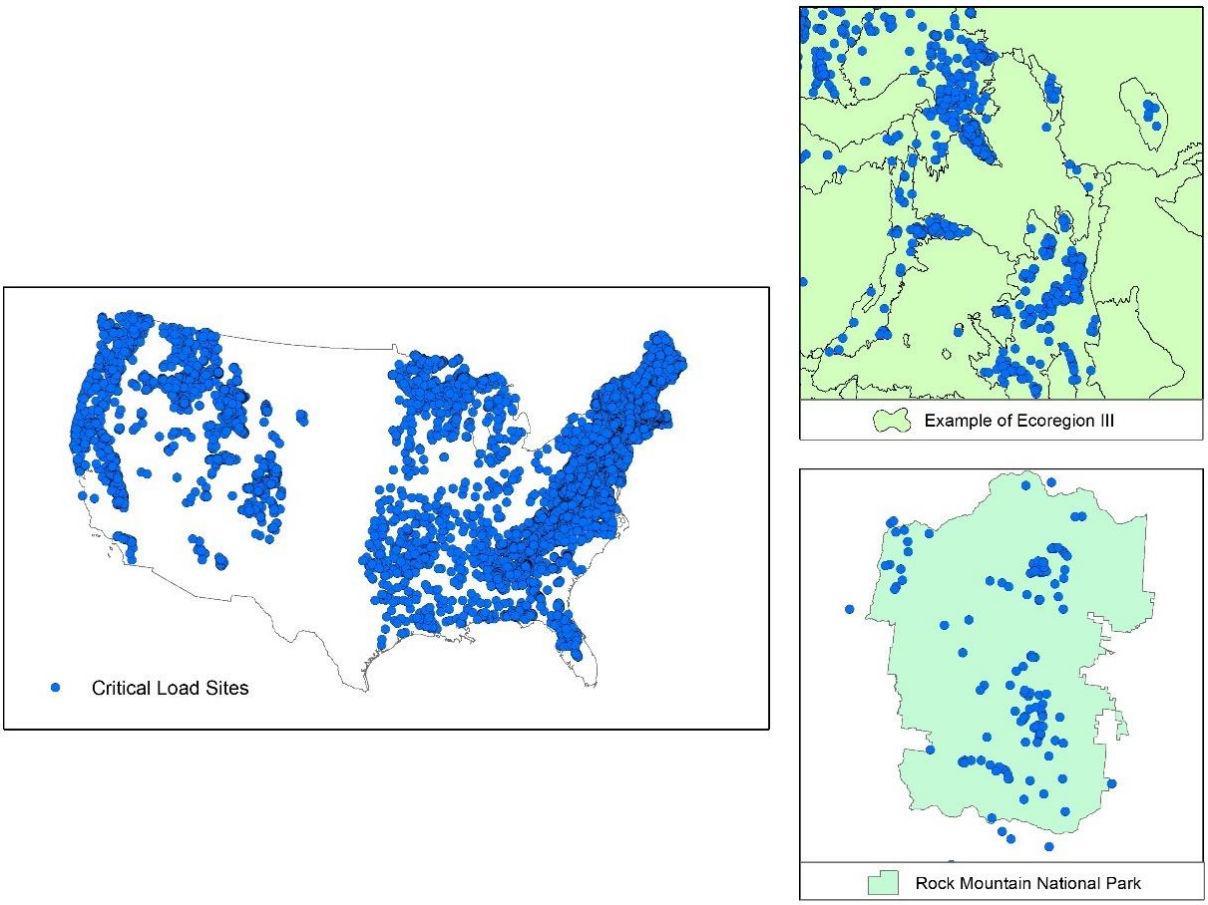

Figure 5A-1. Three scales of the analysis: national, level III ecoregion, and case study.

It is important to note that aquatic ecosystems across the CONUS exhibit a wide range of sensitivity to acidification because of a host of landscape factors, such as geology, hydrology, soils, catchment scale, and vegetation characteristics that control whether a waterbody will be impacted by acidifying deposition. Consequently, variations in ecosystem sensitivity must be considered in order to characterize sensitive populations of waterbodies and relevant regions across the CONUS. The EPA's Omernik ecoregions classifications was used to define ecologically relevant, spatial aggregated, acid sensitive regions across the CONUS in order to better characterize the regional differences in the impact of deposition driven acidification.

Ecoregions are areas of similarity regarding patterns in vegetation, aquatic, and terrestrial ecosystem components. Available ecoregion categorization schemes include the EPA's Omernik classifications (Omernik, 1987). Omernik's ecoregions are categorized using a holistic, "weight-of-evidence" approach in which the relative importance of factors may vary from region to region. The method used to map ecoregions is described in Omernik (1987) and classifies regions through the analysis of the patterns and the composition of biotic and abiotic characteristics that affect or reflect differences in ecosystem quality and integrity. Factors

include geology, physiography, vegetation, climate, soils, land use, wildlife, and hydrology. Three hierarchical levels were developed to distinguish coarser (more general) and finer (more detailed) categorization. Level I is the coarsest level, dividing the CONUS into 12 ecoregions. At level II, the continent is subdivided into 25 ecoregions and the contiguous U.S. into 20 (Figure 5A-2). Level III is a further subdivision of level II and divides North America into 105 ecoregions with 84 in the CONUS. Level IV is a subdivision of level III into 967 ecoregions for the CONUS.

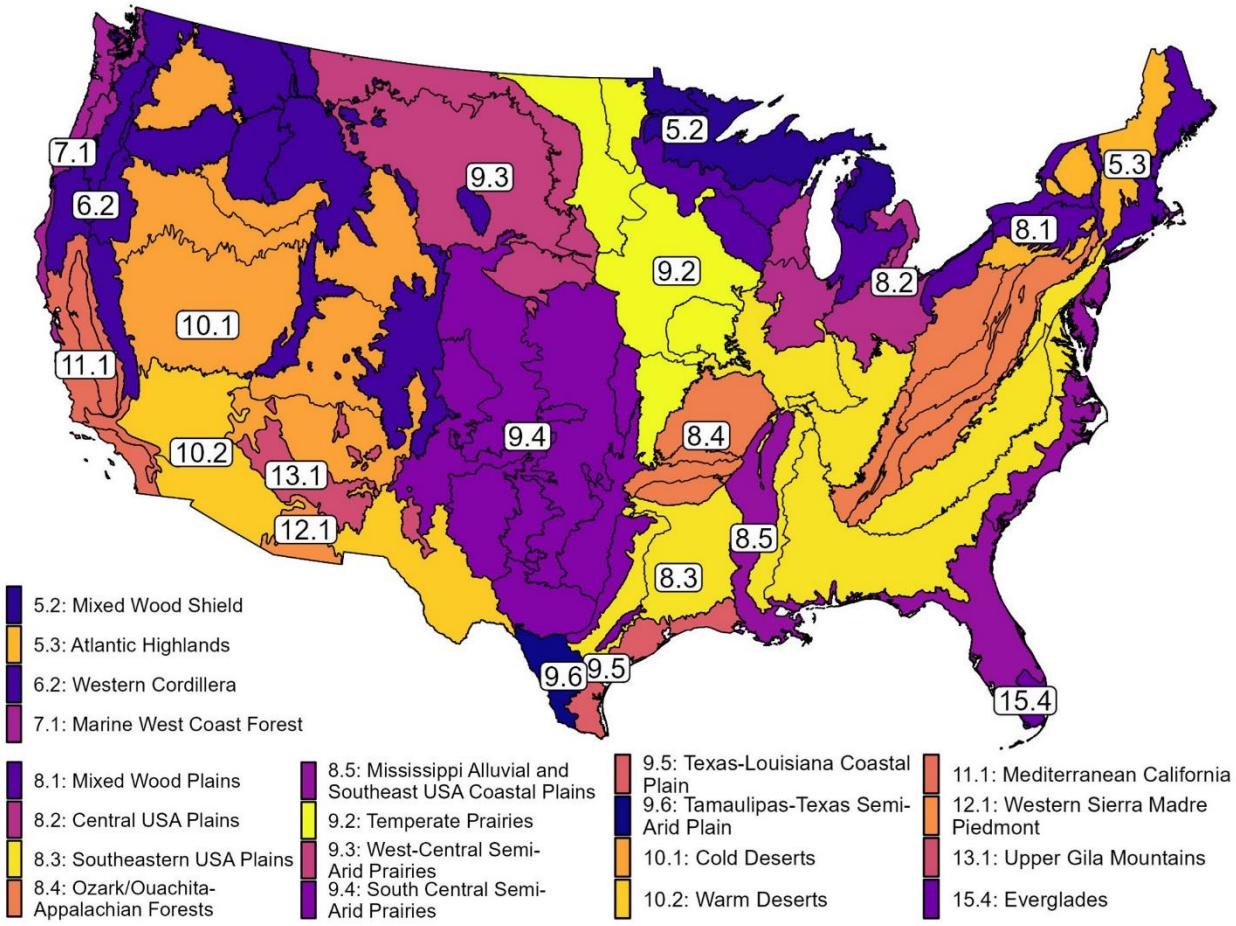

Figure 5A-2. Level II ecoregions with level III subdivisions.

The case study scale represents the smallest scale at which we performed our analyses and is intended to give some insight into the local impact of aquatic acidification. Five case study areas across the U.S. were examined: Shenandoah Valley Area, White Mountain National Forest, Northern Minnesota, Sierra Nevada Mountains, and Rocky Mountain National Park (section 5A.2.3). These areas include a number of parks and national forests that vary in their sensitivity to acidification, but represent high value or protected ecosystems, such as Class 1 areas, wilderness, and national forests.

5A.1.2 Method - Aquatic Critical Load Approach

The impacts of N and/or S deposition on aquatic ecosystems were evaluated using a CL approach. The CL approach was used to characterize the risk of N and/or S deposition on aquatic acidification across the CONUS with a focus on acid sensitive areas. In this assessment, the CL approach provides a means of gauging whether an individual or group of waterbodies (i.e., lake or stream) in a given area receives an amount of deposition that results in the waterbody not being able to achieve the target ANC level (as described in 5A.1.3). Critical load exceedances were summarized at the national, ecoregion III, and case study levels to understand the spatial extent of deposition-driven acidication across the CONUS. Special consideration was given to naturally occuring aquatic acidification in order to focus the analysis on deposition-driven impacts to aquatic biota. Uncertainty associated with the CL estimate was also estimated and factored into the CL exceedance determination.

5A.1.3 Ecological Risk and Response

The biological impact of acidifying deposition is mediated through changes in water quality that in turn impact biota (ISA, Appendices 7 and 8). Deposition of N and/or S can effect biogeochemical changes in aquatic systems that may induce biologically harmful effects. Surface water chemistry is then a good indicator of the risk of acidification on the biotic integrity of freshwater ecosystems, because it integrates soil and water processes that occur within a watershed. Changes in surface water chemistry reflect the influence of acidic inputs from precipitation, gases, and particles, as well as local geology and soil conditions. Surface water chemical factors such as pH, Ca^{2+}, ANC, base cations, ionic metals concentrations, and DOC are affected by acid deposition and, accordingly, are commonly used indicators of acidification. Although ANC does not directly cause effects on biota, it relates to pH and aluminum levels, and biological effects are primarily attributable to low pH and high inorganic aluminum concentration (ISA, section ES.5.1).

The most widely used measure of surface water acidification, and subsequent recovery under reduced acid deposition, is ANC (ISA, Appendix 7, section 7.1.2.6). This is because ANC is associated with the surface water constituents that directly cause or reduce acidity-related stress, in particular pH, Ca^{2+}, and inorganic Al concentrations and ANC is generally a more stable measurement than pH, and it reflects sensitivity and effects of acidification in a linear fashion across the full range of ANC values (ISA, Appendix 7, section 7.1.2.6). These water quality parameters are indicators of aquatic acidification for which there is evidence of effects on aquatic systems including physiological impairment, reduced fitness or survival, alteration of species richness, community composition and structure, and biodiversity in freshwater ecosystems.

The evidence of effects on biota from aquatic acidification indicates a range of severity with varying levels of ANC, pH and inorganic Al, with effects on biota ranging from phytoplankton and invertebrates to fish communities (ISA, Appendix 8, section 8.5). For example, a review by Lacoul et al. (2011) of aquatic acidification effects on aquatic organisms in Atlantic Canada observed that the greatest differences in phytoplankton species richness occurred across a pH range of 4.7 to 5.5 (ANC range of 0 to 20 μeq/L), just below the range (pH 5.5 to 6.5) where bicarbonate becomes rapidly depleted in the water (ISA, Appendix 8, section 8.3.1.1). Under acidifying conditions, these phytoplankton communities shifted from dominance by chrysophytes, other flagellates, and diatoms to dominance by larger dinoflagellates. In benthic invertebrates residing in sediments of acidic streams, Al concentration is a key influence on the presence of sensitive species. Studies of macroinvertebrate species have reported reduced species richness at lower pH, with the most sensitive group, mayflies, absent at the lowest levels. Values of pH below 5 (which may correspond to approximant ANC concentrations below 0 μeq/L)[2] were associated with the virtual elimination of all acid sensitive mayfly and stonefly species over the period from 1937-42 to 1984-85 in two streams in Ontario (Baker and Christensen, 1991). In a more recent study, Baldigo et al. (2009) showed macroinvertebrate assemblages in the southwestern Adirondack Mountains were severely impacted at pH <5.1, moderately impacted at pH from 5.1 to 5.7, slightly impacted at pH from 5.7 to 6.4 and usually unaffected above pH 6.4 (Figure 5A-3). In Atlantic Canada, Lacoul et al. (2011) found the median pH for sensitive invertebrate species occurrence was between 5.2 and 6.1 (ANC of 10 and 80 μeq/L), below which such species tended to be absent. For example, some benthic macroinvertebrates, including several species of mayfly and some gastropods are intolerant of acid conditions and only occur at pH ≥5.5 (ANC 20 μeq/L) and ≥6, (ANC 50 μeq/L) respectively. (ISA, section 8.3.3).

[2] pH and ANC were related to one another using a generalized relationship base on equilibrium with atmospheric CO_2 concentration (Cole and Prairie, 2010).

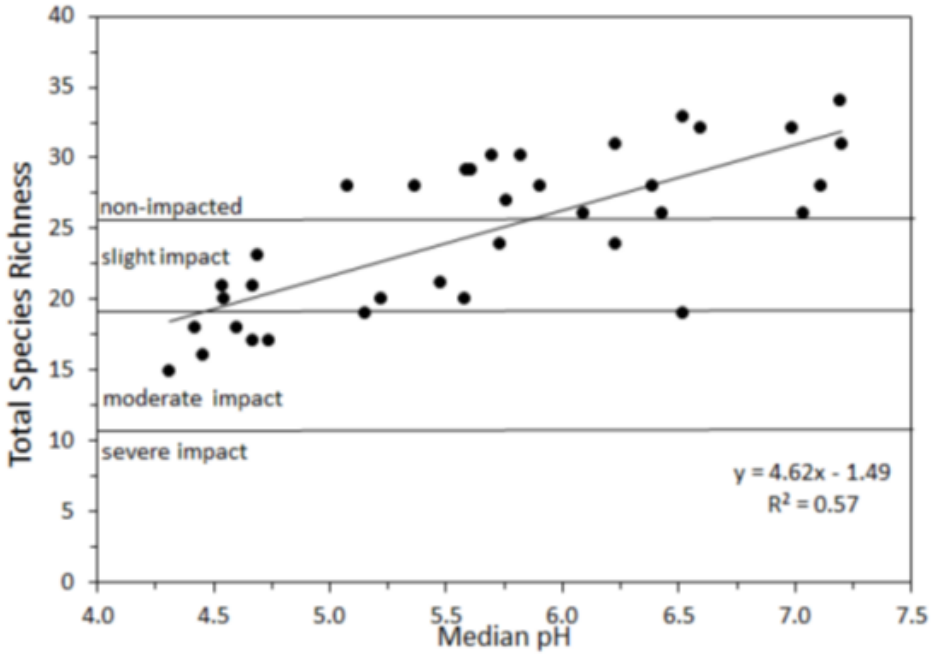

Figure 5A-3. Total macroinvertebrate species richness as a function of pH in 36 streams in western Adirondack Mountains of New York, 2003-2005. From Baldigo et al. (2009); see ISA, Appendix 8, section 8.3.3, and p. 8-12.

Responses among fish species and life stages to changes in ANC, pH and Al in surface waters are variable. Early life stages such as larvae and smolts are more sensitive to acidic conditions than the young-of-the-year, yearlings, and adults (Baker et al., 1990; Johnson et al., 1987; Baker and Schofield 1985). Studies showed a loss of fish whole-body sodium in trout when stream pH drops below 5.1 (ANC 0 μeq/L) indicating loss of the ability to ionoregulate. Some species and life stages experienced significant mortality in bioassays at relatively high pH ((e.g., pH 6.0−6.5; ANC 50-100 μeq/L for eggs and fry of striped bass and fathead minnow) (McCormick et al., 1989; Buckler et al., 1987)), whereas other species were able to survive at quite low pH without adverse effects. Many minnows and dace (Cyprinidae) are highly sensitive to acidity, but some common game species such as brook trout, largemouth bass, and smallmouth bass are less sensitive. A study by Neff et al. (2008), investigated the effects of two acid runoff episodes in the Great Smoke Mountains National Park on native brook trout using an in-situ bioassay. The resulting whole-body sodium concentrations before and after the episodes showed negative impacts on physiology. More specifically, the reduction in whole-body sodium when stream pH dropped below 5.1 (ANC 0 μeq/L) indicated that the trout had lost the ability to ionoregulate (ISA, Appendix 8, section 8.3.6.1). Field and laboratory bioassay studies indicate variation in pH ranges among fish species (Figure 5A-4).

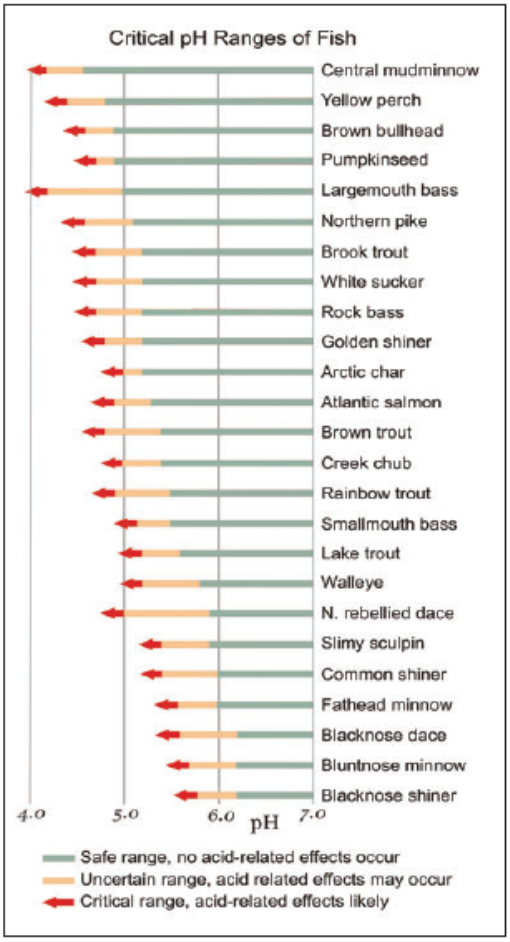

Figure 5A-4. Critical aquatic pH range for fish species. Notes: Baker and Christensen (1991) generally defined bioassay thresholds as statistically significant increases in mortality or by survival rates less than 50% of survival rates in control waters. For field surveys, values reported represent pH levels consistently associated with population absence or loss. Source: Fenn et al. (2011) based on Baker and Christensen (1991) (ISA, Appendix 8, Figure 8-3).

As noted in the ISA, "[a]cross the eastern U.S., brook trout are often selected as a biological indicator of aquatic acidification because they are native to many eastern surface waters and because residents place substantial recreational and aesthetic value on this species" (ISA, Appendix 8, p. 8-26). Compared to other fish species in Appalachian streams, this species is relatively pH sensitive. For example, "[in many Appalachian mountain streams that have been acidified by acidic deposition, brook trout is the last fish species to disappear; it is generally lost at pH near 5.0 (MacAvoy and Bulger, 1995), which usually corresponds in these streams with ANC near 0 μeq/L (Sullivan et al., 2003)" (ISA, Appendix 8, p. 8-21).

As described in section 4.2.1 episodic acidification during storm events can pose risks in low ANC streams. For example, streams with ANC around 20 μeq/L or less at base flow may be

considered vulnerable to episodic acidification events that could reduce pH and ANC to levels potentially harmful to brook trout and other species. Streams with suitable habitat and annual average ANC greater than about 50 μeq/L are often considered suitable for brook trout in southeastern U.S. streams and reproducing brook trout populations are expected (Bulger et al., 2000). Streams of this type provide "sufficient buffering capacity to prevent acidification from eliminating this species and there is reduced likelihood of lethal storm-induced acidic episodes" (ISA, Appendix 8, p. 8-26). Results of a study by Andren and Rydin (2012) suggested a threshold of Al less than 20 ug/L and pH higher than 5.0 for healthy brown trout populations by exposing yearling trout to a pH and inorganic Al gradient in humic streams in Scandinavia (ISA, Appendix 8, section 8.3.6.2). Another recently available study that investigated the effects of episodic pH shifts fluctuations in waterbodies of eastern Maine reported that episodes resulting in pH dropping below 5.9 (ANC of ~50 μeq/L) have the potential for harmful physiological effects to Atlantic salmon smolts if coinciding with the smolt migration in eastern Maine rivers (Liebich et al., 2011; ISA, Appendix 8, section 8.3.6.2).

Investigations of waterbody recovery from historic deposition have reported on episodic acidification associated with the high SO_4^{2-} remaining in watershed soils. For example, monitoring data in the Great Smoky Mountains National Park indicated that while the majority of SO_4^{2-} entering the study watershed was retained, SO_4^{2-} in wet deposition moved more directly and rapidly to streams during large precipitation events, contributing to episodic acidification of receiving streams and posing increased risk to biota (ISA, Appendix 7, section 7.1.5.1.4). High flow episodes in historically impacted watersheds of the Appalachians have been reported to appreciably reduce stream ANC (Lawrence et al., 2015).

There is often a positive relationship between pH or ANC and number of fish species, at least for pH values between about 5.0 and 6.5, or ANC values between about 0 and 50 to 100 μeq/L (Cosby et al., 2006; Sullivan et al., 2006; Bulger et al., 1999). This is because energy cost in maintaining physiological homeostasis, growth, and reproduction is high at low ANC levels (Sullivan et al., 2003; Wedemeyer et al., 1990). As noted in section 4.2.1.1.2, surveys in the heavily impacted Adirondack mountains found that lakes and streams having an annual average ANC < 0 μeq/L and pH near or below 5.0 generally support few or no fish species to no fish at all, as illustrated in Figure 5-3 below (Sullivan et al., 2006; ISA, Appendix 8, section 8.3.6.3.

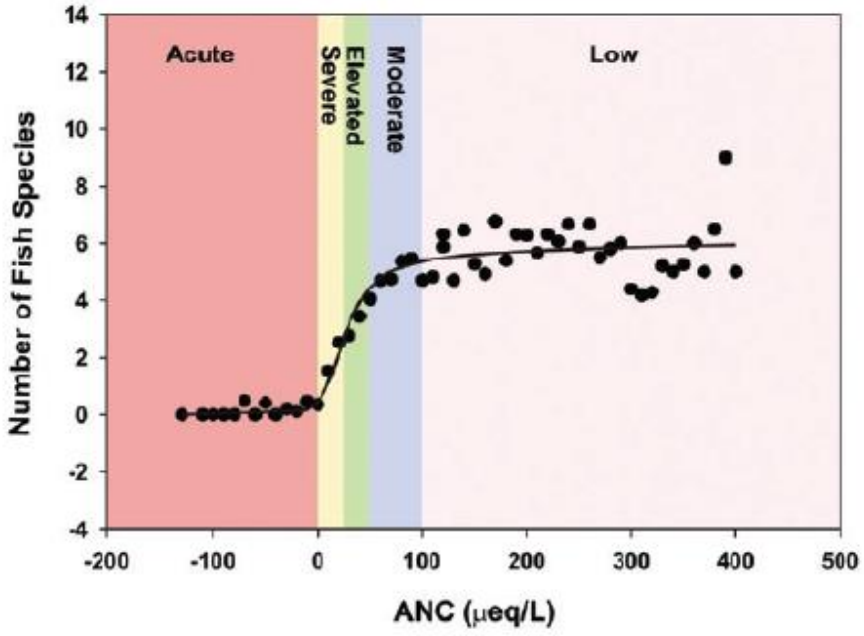

Figure 5A-5. Number of fish species per lake *versus* acidity status, expressed as ANC, for Adirondack lakes. Notes: The data are presented as the mean (filled circles) of species richness within 10 μeq/L ANC categories, based on data collected by the Adirondacks Lakes Survey Corporation. Source: Modified from Sullivan et al. (2006). (ISA, Appendix 8, Figure 8-4)

The data presented in Figure 5A-5 above suggest that there could be a loss of fish species in these lakes with decreases in ANC below approximately 50 to 100 μeq/L (Sullivan et al., 2006). For streams in Shenandoah National Park, a statistically robust relationship between ANC and fish species richness was also documented by Bulger et al. (2000). However, interpretation of species richness relationship with ANC can be difficult and misleading, because more species tend to occur in larger lakes and streams as compared with smaller ones, irrespective of acidity (Sullivan et al., 2006) because of increased aquatic habitat complexity in larger lakes and streams (Sullivan et al., 2003; ISA, Appendix 8, section 8.3.6.3).

Observations of effects in watersheds impacted by historic acidification can also reflect the influence of episodic high flow events that lower pH and ANC appreciably below the baseflow ANC (as described above). Studies described above are summarized below in the context of ANC ranges: <0, 0-20, 20-50, 50-80, and >80 μeq/L:

- At ANC levels <0 μeq/L, aquatic ecosystems have exhibited low to a near loss of aquatic diversity and small population sizes. For example, planktonic and macroinvertebrates communities shift to the most acid tolerant species (Lacoul et al., 2011) and mayflies can be eliminated (Baker and Christensen, 1991). A near to complete loss of fish populations can occur, including non-acid sensitive native species such as brook trout (*Salvelinus fontinalis*), northern pike (*Esox lucius*), and others (Sullivan et al., 2003, 2006; Bulger et al., 2000), which is in most cases attributed to elevated inorganic monomeric Al

concentration (Baldigo and Murdoch, 1997). At this level, aquatic diversity is at its lowest (Bulger et al., 2000; Baldigo et al., 2009; Sullivan et al., 2006) with only acidophilic species being present.

- In waterbodies with ANC levels between 0 and 20 μeq/L, acidophilic species dominate other species (Matuszek and Beggs, 1988; Driscoll et al., 2001) and diversity is low (Bulger et al., 2000; Baldigo et al., 2009; Sullivan et al., 2006). Plankton and macroinvertebrate populations have been observed to decline, and acid-tolerant species have outnumbered non-acid sensitive species (Liebich et al., 2011). Sensitive species are often absent (e.g., brown trout, common shiner, etc.) while non-sensitive fish species populations may be reduced (Bulger et al., 2000). Episodic acidification events (e.g., inflow with ANC <0 μeq/L and pH< 5), may have lethal impacts on sensitive lifestages of some biota, including brook trout and other fish species (Matuszek and Beggs, 1988; Driscoll et al., 2001).
- Levels of ANC between 20 and 50 μeq/L have been associated with the loss and/or reduction in fitness of aquatic biota that are sensitive to acidification in some waterbodies of the Adirondacks and Appalachians. Such effects included reduced aquatic diversity (Kretser et al., 1989; Lawrence et al., 2015; Dennis and Bulger, 1995) with some sensitive species missing (Bulger et al., 2000; Sullivan et al., 2006). In historically impacted watersheds, waterbodies with ANC below 50 μeq/L are more vulnerable to increased potential for harm associated with episodic acidification (ISA, Appendix 8, section 8.2). Comparatively, acid tolerant species, such as brook trout may have moderate to healthy populations, (Kretser et al., 1989; Lawrence et al., 2015; Dennis and Bulger, 1995).
- At an ANC between 50 and 80 μeq L-1, the fitness and population size of some sensitive species have been affected in some historically impacted watersheds. Levels of ANC above 50 μeq/L are considered suitable for brook trout and most fish species because buffering capacity is sufficient to prevent the likelihood of lethal episodic acidification events (Driscoll et al.; 2001; Baker and Christensen; 1991). However, depending on other factors, the most sensitive species have been reported to experience a reduction in fitness and/or population size in some waterbodies (e.g., blacknose shiner [Baldigo et al., 2009; Kretser et al., 1989; Lawrence et al., 2015; Dennis and Bulger, 1995]). Fish species richness has also been reported to be affected in some Adirondack streams at ANC 50 (Sullivan et al., 2006).
- Values of ANC >80 μeq/L have generally not been associated with harmful effects on biota (Bulger et al., 1999; Driscoll et al., 2001; Kretser et al., 1989; Sullivan et al., 2006).

5A.1.4 Chemical Criterion and Critical Threshold

Most aquatic CL studies conducted in the U.S. use surface water ANC as the principal metric of water quality change in response to changes in a N and/or S deposition, which is known as the chemical criterion. The ANC is generally a more stable measurement than pH because ANC is insensitive to changes in CO_2 and it reflects sensitivity and effects of acidification in a linear fashion across the full range of ANC values. The critical threshold is then the value of the chemical criterion (ANC) beyond which it is negatively impacted. For the

analyses in this assessment, CLs were evaluated with respect to three different ANC thresholds to account for variation in waterbodies with regard to risk of episodic acidification events, associated uncertainties, and potential for differing science policy judgments on these uncertainties: 20 μeq/L, 30 μeq/L and 50 μeq/L based on section 5A.1.3. Most aquatic CL studies conducted in the U.S. since 2010 use an ANC of 20 and/or 50 μeq/L, because 20 μeq/L is considered by the authors to provide protection for "natural" or "historical" range of ANC and 50 μeq/L provides overall ecosystem protection (DuPont et al., 2005; McDonnell et al., 2012, 2014; Sullivan et al., 2012a, 2012b; Lynch et al., 2022; Fakhraei et al., 2014; Lawrence et al., 2015). In the Mountain west, vulnerable lakes and streams to deposition driven aquatic acidification are often found in the mountains where surface water ANC levels are low and typically vary between 0 and 30 μeq/L (Williams and Labou, 2017; Shaw et al., 2014). For these reasons, various studies, including some represented in the National Critical Loads Database (NCLD), have used an ANC threshold of 50 μeq/L for the eastern and 20 μeq/L for the western CONUS (denoted as "50/20" μeq/L). In the analyses in this assessment, we have calculated CL exceedances for ANC thresholds of 20, 30 and 50 μeq/L across the CONUS, and also for the 50/20 (E/W)[3] application of ANC thresholds. An ANC of 80 μeq/L was considered; however, it was determined that many waterbodies, particularly, in acid sensitive regions of CONUS never had an ANC that high and would never reach an ANC that high naturally.

5A.1.4.1 Natural Acidic Waterbodies

Some waterbodies are naturally acidic because of multiple factors, but most commonly due to acidic rock within the waterbodies watershed, low base cation weathering rates linked to the type of bedrock, and high DOC with the surface waters. Natural or historical level of ANC concentration are typically above 20 μeq/L (Sullivan et al., 2012a; Shaw et al., 2014). Sullivan et al. (2012a) using Model of Acidification of Groundwater in Catchment (MAGIC) simulations for pre-industrial (1850), suggested that in pre-industrial times, there were no acidic lakes (ANC ≤ 0 μeq/L) and only ~6% of modeled lakes had ANC < 20 μeq/L in the Adirondack mountains, NY. For these reasons, most recent CL studies (since 2010) use 20 μeq/L as a minimum ANC threshold. For waterbodies where their natural or historical level of ANC is lower than the selected ANC threshold, the calculated CL is invalid or not achievable at any level of deposition. In those cases, the CL was evaluated, but was not included in the results and summary assessments.

[3] Consistent with regional definitions based on groups of states that were employed in the last review, in this REA for this current review, the West includes the states of ND, SD, CO, WY, MT, AZ, NM, UT, NV, ID, CA, OR, and WA (2009 REA, Appendix 1, p. 1-21). Accordingly, an ecoregion is designated western if it intersects or overlaps with these ten states, and eastern ecoregions are those not designated as western.

5A.1.5 Critical Load Data

Aquatic CLs used in this assessment came from the National Critical Load Database version 3.2 (Lynch et al., 2022) and include recent studies identified in the ISA (e.g., Lawrence et al., 2015; Fakhraei et al., 2014; Sullivan et al., 2012a; Fakhraei et al., 2016). The NCLD is comprised of CLs calculated from several common models: (1) steady-state mass-balance models such as the Steady-State Water Chemistry (SSWC), (2) dynamic models such as MAGIC (Cosby et al., 1985) or Photosynthesis EvapoTranspiration Biogeochemical model (PnET-BGC) (Zhou et al., 2015) run out to year 2100 or 3000 to model steady-state conditions and (3) regional regression models that use results from dynamic models to extrapolate to other waterbodies (McDonnell et al., 2012; Sullivan et al., 2012a). These approaches differ in the way watershed base cation weathering was determined (e.g., F-Factor or dynamic model).

Figure 5A-6 shows the unique locations for 13,000+ CLs used in this assessment. Critical load waterbodies are concentrated in areas that are acid sensitive in the eastern U.S. and the Rocky Mountain and Pacific Northwest regions of the west. Not all waterbodies are sensitive to acidification. Small to medium size lakes size (>200 Ha) and streams (1- 3 orders) tend to be the waterbodies that are impacted by deposition driven acidification. Rivers are not typically impacted by deposition driven acidification. Data in the NCLD are generally focused on waterbodies impacted by deposition-driven acidification. A waterbody is represented as a single CL value. In many cases, a waterbody has more than one CL value calculated for it because different studies determined a value for the same waterbody. When more than one CL exists, the CL from the most recent study was selected or averaged when the publications are from the same timeframe.

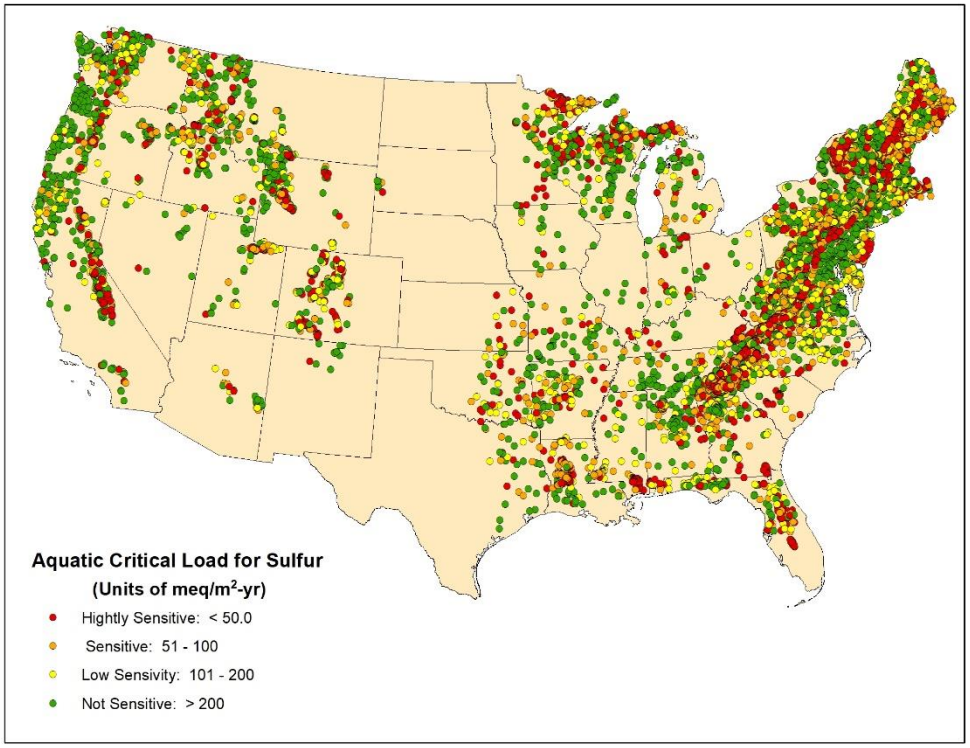

Figure 5A-6. Unique waterbody locations with CL estimates used in this assessment. Lower values are red and orange; the lowest bin includes CLs of zero (section 5A.1.6).

5A.1.5.1 Steady-State Water Chemistry Model and F-Factor

Critical loads derived with the Steady State Water Chemistry (SSWC) model used available water chemistry data, and are based on the principle that excess base cation production within a catchment area should be equal to or greater than the acid anion input, thereby maintaining the ANC above a pre-selected level (Scheffe et al., 2014; Miller, 2011; Dupont et al., 2005; and Vermont Department of Environmental Conservation (VDEC), 2003, 2004, 2012). The SSWC model assumes a mass balance and that all SO_4^{2-} in runoff originates from sea salt spray and anthropogenic deposition. The acidity CL can be defined for S only (CLS) and S and N (CLSN). A Cl for S only is calculated based on the principle that the acid load should not exceed the non-marine, base cation inputs minus a nutrient base cation uptake and ANC buffer to protect selected biota from being damaged (Eq. 5A-2):[4]

$$CLS = BC^*_{dep} + BC_w - Bc_u - nANC_{crit} \quad \textbf{(5A-2)}$$

Where:

[4] The F-factor approach to the SSWC model uses an integrated watershed estimates of the base cation inputs of BC*dep, BCw, and Bcu defined as base cation flux (BC*o), e.g., $BC^*o = BC^*_{dep} + BC_w - Bc_u$.

BC^*_{dep} (BC; Ca+Mg+K+Na) = the sea-salt corrected non-anthropogenic deposition of base cations (the asterisk denotes the correction for base cations of marine origin [Henriksen et al., 2002]);

BC_w (BC; Ca+Mg+K+Na) = the average watershed weathering flux;

Bc_u (Bc: Ca+Mg+K) = the net long-term average uptake of base cations in the biomass (i.e., the annual average removal of base cations due to harvesting);

$nANC_{crit}$ = the lowest ANC-flux that protects the biological communities.

BC_u = zero for these CLs.

For these CLs based on both S and N, the SSWC model was modified to incorporate a simplified N framework whereby N components that account for nitrogen removal from long-term nitrogen immobilization and denitrification were included in the model (Eq. 5A-3): [5]

$$CLSN = BC^*_{dep} + BC_w + N_u + N_i + N_{de} - Bc_u - nANC_{crit} \quad \textbf{(5A-3)}$$

Where:

N_u = N removal through removal of trees with harvesting;

N_i = N removal from long-term N immobilization;

N_{de} = N removal from the soil through microbial denitrification.

The sum of N_u, N_i, and N_{de} defines the minimum CL for N (CLN_{min}) as the amount of N deposition that does not lead to acidification in the watershed. The variable, N_i, was set equal to 4.30 meq/m^2-yr (McNulty et al., 2007) and N_{de} was set equal to 7.14 meq/m^2-yr (Ashby et al., 1998) for sites in the east. For western states, a combined value of N_i+N_{de} = 11.0 eq/ha-yr was used based on Nanus et al. (2012). For Sullivan et al. (2012b), N_u also includes removal of N via uptake by tree boles that were harvested, which was based on literature values summarized by McNulty et al. 2007. Nitrogen removal can also be incorporated into the acidity CL and CL exceedances (Ex) using the using the NO_3^- leaching flux, $N_{le,}$ (Henriksen and Posch, 2001) (Eq. 5A-4):

$$Ex = S_{dep} + N_{le} - CLS \quad \textbf{(5A-4)}$$

Where:

N_{le} = the sum of the measured concentrations of nitrate (NO_3^- eq/L) and ammonia (NH_4^+ eq/L) in the runoff (Q_s m/yr) as $([NO_3^-]+[NH_4^+])*Q_s$.

[5] The F-factor approach to the SSWC model uses an integrated watershed estimates of the base cation inputs of BC*dep, BCw, and Bcu defined as base cation flux (BC*o), e.g., $BC^*o = BC^*_{dep} + BC_w - Bc_u$.

See sections 5A.1.6 “Critical Load Exceedance” and 5A.1.6.2 “Acidifying Contribution of Nitrogen Deposition” below regarding how exceedances of CLs of S and of S and N combined are calculated and how N_{le} was determined.

5A.1.5.2 MAGIC Model and Regional Linear Regression Models for Estimating BC_w Input to SSWC

Sullivan et al. (2012b) CLs used a modified form of the SSWC model (see Eq. 5A-3) where base cation weathering was derived using a new method based on MAGIC model estimates of BC_w and regional linear regression models (see Sullivan et al., 2012b and McDonnell et al., 2012), rather than the F-factor method for estimating BC_w.

The MAGIC model was used to calculate watershed-specific BC_w for input to regional linear regression models that estimated BC_w in all watersheds, including those without MAGIC values. The BCw estimates were then used as input to the SSWC model. MAGIC is a lumped-parameter model of intermediate complexity, developed to predict the long-term effects of acidic deposition on surface water chemistry (Cosby et al., 1985). The model simulates soil solution chemistry, weathering rates, and surface water chemistry to predict the monthly and annual average concentrations of the major ions in these waters (see Cosby et al., 1985 for more details about the model itself). The base cation weathering terms in MAGIC represent the catchment-average weathering rates for the soil compartments. In a one soil-layer application of MAGIC, the weathering rates in MAGIC thus reflect the catchment-average net supply of base cations to the surface waters draining the catchment. The sum of the MAGIC weathering rates for the individual base cations is therefore identical in concept to the base cation weathering term, BC_w, in the SSWC CL model (Eq. 5A-2). Base cation weathering rates in MAGIC are calibrated parameters. The calibration procedure uses observed deposition of base cations, observed (or estimated) base cation uptake in soils, observed stream water base cation concentrations, and runoff (Q_S). These observed input and output data provide upper and lower limits for internal sources of base cations in the catchment soils. The two most important internal sources of base cations in catchment soils are modeled explicitly by MAGIC: primarily mineral weathering and soil cation exchange. During the calibration process, observed soil base saturation for each base cation and observed soil chemical characteristics are combined with the observed input and output data to partition the inferred net internal sources of base cations between weathering and base cation exchange.

The watershed-specific BC_w values calculated by the MAGIC and input to a regional regression model provided for watershed specific BC_w values for 500+ monitoring locations in the Appalachian Mountains of Virginia and West Virginia. Water chemistry and landscape variables were used as the predictor variables in regression analyses to extrapolate BC_w. Each of

the calibrated MAGIC study watersheds was placed in an ecoregion category based on which ecoregion contained most of the watershed area and three separate regression models were developed for each ecoregion (Table 5A-1). Watershed averages were used to represent the spatial variability within each watershed for the landscape characteristics, except for watershed area. Regression models were established using stepwise linear regression using 'best subsets' to evaluate candidate models and constrain the number of independent predictor variables during model selection. Water quality predictor data were collected during several regional surveys, as compiled by Sullivan and Cosby, 2004). These surveys included the National Stream Survey (NSS), Environmental Monitoring and Assessment Program (EMAP), Virginia Trout Stream Sensitivity Study (VTSSS), and stream surveys conducted in Monongahela National Forest. One water quality sample, generally collected during the spring between 1985 and 2001, was used to characterize each watershed (Sullivan and Cosby, 2004).

Table 5A-1. Multiple regression equations to estimate BC_w from either water chemistry and landscape variables or from landscape variables alone, stratified by ecoregion.

Ecoregion	n	Equation	r^2
Central Appalachian	24	BC_w = -37.5 + 0.6 (SBC) + 0.9 (NO_3) + 0.006 (WS Area)	0.93
Ridge and Valley	42	BC_w = 107.0 + 0.5 (SBC) - 0.06 (Elevation) - 3.2 (Slope)	0.86
Blue Ridge	26	BC_w = 27.1 + 0.6 (CALK) + 0.6 (NO_3)	0.90
Note: These equations are presented in Table S2 of the Supplemental Materials for Sullivan et al. (2012b). SBC is the sum of base cations; CALK is calculated ANC. The r^2 values are for correlation of BC_w predicted by the regression equations with the BCw calculated by MAGIC based on the site-specific water chemistry data for these sites, as presented in Table 2 and Figure 3 of McDonnell et al. (2012).			

5A.1.5.3 MAGIC model and Hurdle Modeling for Estimating BC_w Input to SSWC

For McDonnell et al. (2014) and Povak et al. (2014) CLs used a modified form of the SSWC model that excluded the N terms. Building on the framework of Sullivan et al. (2012b) and McDonnell et al. (2012), McDonnell et al. (2014) and Povak et al. (2014) expanded the study area and developed new statistical models to better predict BC_w and evaluate CLs of S. Their studies expanded the area to include the full Southern Appalachian Mountain region and surrounding terrain from northern Georgia to southern Pennsylvania, and from eastern Kentucky and Tennessee to central Virginia and western North Carolina.

As with Sullivan et al. (2012b) and McDonnell et al. (2012), the MAGIC model was used to calculate watershed-specific BC_w for 140 stream locations containing both measured soil chemistry and water chemistry data (see section above for a description of MAGIC). In addition, McDonnell et al. (2014) aggregated all known water quality data that totaled 933 sample

locations in order to develop a statistical model to predict ANC and BC_w for all streams in the Southern Appalachian Mountain region. Water chemistry data were collected between 1986 and 2009, with stream ANC calculated as the equivalent sum of the base cation concentrations (Ca^{2+}, Mg^{2+}, K^+, Na^+, ammonium [NH_4^+]) minus the sum of the mineral acid anion concentrations (chloride [Cl^-], NO_3^-, SO_4^{2-}). Base cation weathering flux, BC_w, was estimated as the net internal source of base cations between weathering and base cation exchange for the watershed based on the MAGIC model calibrations, which used observed stream and soil chemistry data, and atmospheric deposition estimates to simulate surface water and soil solution chemistry (McDonnell et al., 2014).

A random forest regression modeling technique was used to generate estimates of BC_w and ANC for the region. This was based on a suite of initial candidate predictor variables chosen to represent potential broad- to fine-scale climatic, lithologic, topoedaphic, vegetative, and S deposition variables that have the potential to influence ANC and BC_w. To represent the landscape conditions that influence specific locations along a stream, all candidate landscape predictor variables were expressed on a 30 m grid basis across the Southern Appalachian Mountain domain This resolution allowed for the creation of "flowpaths" for the development of a topographically determined stream network. All data values for each target grid cell and upslope grid cells were averaged based on the technique described in McDonnell et al. (2012).

A total of 140,504 watersheds were represented (i.e., delineated) with the use of a hydrologically conditioned digital elevation model derivatives drawn from the National Hydrography Dataset (NHD+) (https://www.epa.gov/waterdata/nhdplus-national-hydrography-dataset-plus). The CLs from McDonnell et al. (2014) and Povak et al. (2014) were then calculated with the SSWC model (Henriksen and Posch, 2001) using the estimates of BC_{dep}, BC_w, Bc_u, Q_s and the ANC criterion set to a value of 50 µeq/L for each stream node. See McDonnell et al. (2014) and Povak et al. (2014) for additional methods detail.

5A.1.6 Critical Load Exceedance

A critical load exceedance (Ex) is when deposition is greater than the critical load. Critical Load exceedances define when the benchmark for which the CL is derived is likely to be exceeded. Uncertainty associated with the CL estimates were taken into account in the calculation of CL exceedances. Specifically, based on preliminary analyses, a 6.25 meq S/m^2-yr or 1 kg S/ha-yr range of uncertainty was used in the exceedance calculation.[6,7] For that reason,

[6] Based on the CL uncertainty analysis (see section 5A.3**)**, on average the magnitude of the uncertainty for aquatic CLs is 4.29 meq S/m^2-yr or 0.69 kg S/ha-yr and a confidence interval of ±2.15 meq/m^2-yr or ±0.35 kg S/ha-yr.

[7] Critical load estimates have been converted from meq/m^2-yr to kg S/ha-hr by dividing by 6.25. This takes into account conversions from milliequivalents to equivalents, equivalents to kg S, and m^2 to ha.

we conclude that CLs are exceeded where deposition is above 3.125 meq S/m^2-yr or 0.5 kg S/ha-yr and are not exceeded where deposition is below 3.125 meq S/m^2-yr or 0.5 kg S/ha-yr. The exceedances that fall within this range are described as being "at" the CL.[8] This factor is generally confirmed by the CL uncertainty analysis (see section 5A.3**)**. For comparisons of deposition to CL falling within this range, it is judged unclear whether the CL is exceeded.[9]

Aquatic CL exceedances can be considered with respect to S and combined N and S deposition. When considering only S deposition (i.e., N deposition is zero), the exceedance is expressed as the difference between the CL of S, total S deposition, and an uncertainty of ±3.125 meq S/m^2-yr or ±0.5 kg S/ha-yr (Eq. 5A-6).

$$\text{Exceedance (Ex)} = (\text{Total S deposition} - \text{CLS}) > 3.125 \text{ meq S/m}^2\text{-yr} \quad \textbf{(5A-6)}$$

In most cases, deposition of both S and N contributes to the exceedance. Calculating a combined S and N Ex is more complex because both S and N contribution to acidification needs to be factored in the exceedance. Given that not all N deposition to a watershed will contribute to acidification, the N deposition removed by long-term N processes in the soil and waterbody (e.g., N uptake and immobilization) defines a "minimum" CL for N, noted as CLN_{min}. Nitrogen deposition inputs below what is removed do not acidify, but the amount above this minimum will likely contribute to acidification.

Exceedance of both N and S is a two-step calculation where if N removal is greater than N deposition, only S deposition contributes to the Ex (Eq. 5A-8**)**. However, if deposition of N is greater than what is removed, the amount is not removed (Eq. 5A-9):

When minimum CLNmin ≥ Total N deposition, then

$$\text{Ex(N+S)} = \text{Total S deposition} - \text{CLS} \quad \textbf{(5A-8)}$$

When minimum CLNmin < Total N deposition, then

$$\text{Ex(N+S)} = \text{Total S + N deposition} - \text{CLS} + \text{CLNmin} \quad \textbf{(5A-9)}$$

[8] The approach used here is generally consistent with the approach described in Chapter VII: Exceedance Calculation (2015) of CLRTAP (2014-2021) Manual on Methodologies and Criteria for Modeling and Mapping Critical Loads and Levels and Air Pollution Effects Risks and Trends (available at: https://www.umweltbundesamt.de/en/cce-manual).

[9] The approach used here is generally consistent with the approach described in Chapter VII: Exceedance Calculation (2015) of CLRTAP (2014-2021) Manual on Methodologies and Criteria for Modeling and Mapping Critical Loads and Levels and Air Pollution Effects Risks and Trends (available at: https://www.umweltbundesamt.de/en/cce-manual).

There are different methods for determining the contribution of N deposition to aquatic acidification. Section 5A.1.6.2 below described the two most common methods and how they are handled in the CL exceedance calculations.

5A.1.6.1 Deposition

The amount of deposition used in the critical load exceedance calculation was determined from the Total Deposition (TDep) model (https://nadp.slh.wisc.edu/committees/tdep/) (Schwede and Lear (2014). See section 2.5.1 for more details. Both total N and S deposition were determined to be the deposition level at the grid cell of the stream reach or lake location. For each waterbody total N and S deposition was determined for each year from 2000 to 2020. Three-year averages were calculated for these periods: 2001-03, 2006-08, 2010-12, 2014-16 and 2018-20 to be used in the different analyses. Critical load exceedances were then calculated for each of these five periods and summed nationally and by level III ecoregion.

5A.1.6.2 Acidifying Contribution of Nitrogen Deposition

Unlike sulfur, not all N deposition leads to acidification. In fact, in some systems, none of the entering N deposition acidifies because it is retained in biomass (terrestrial and aquatic) and soils or is lost to the atmosphere by denitrification (ISA, Appendix 7, section 7.1.2.1). The contribution of N deposition that acidifies is difficult to estimate and uncertain because the underlying processes that store and release N in a watershed are complex, making them hard to measure or model. Different methods have been developed to determine the amount of N deposition that acidifies related to aquatic CL exceedances. There are two common approaches in the studies that derived CLs used in this assessment: the first approach is based on the amount of "N leaching" to the waterbody determined by the amount of dissolved N in the water measured as the concentration of nitrite and runoff as presented in Henriksen and Posch (2001). The second approach is the use of a "set value" based on long-term estimate of N immobilization and denitrification as described by McNulty et al. (2007).

While the majority of atmospherically deposited N is either denitrified or accumulates in watershed soils, vegetation, or groundwater (Galloway et al., 2008), the relative partitioning of N loss via denitrification versus watershed storage is poorly known (Galloway et al., 2003). The amount of N leaching to a waterbody that is not retained within the waterbody's biota is the actual amount that contributes to acidification in the surface water. This depends on the amount of N immobilized in the watershed, the amount exported to the drainage waters from the soils, and how much uptake there is within the waterbody itself (Bergström, 2010; ISA Appendix 9, section 9.1.1.2). As the different forms of N deposition enter a watershed, they undergo many biogeochemical changes that result in N being stored in the soil and vegetation and being released to the drainage water. As N deposition enters the watershed it can be quickly taken up

by the microorganisms in the soils and vegetation (particularly NH_3) and incorporated into biomass. This is the amount of N immobilized in the watershed. Nitrogen immobilization or accumulation is the conversion of inorganic N to organic N. The amount that is immobilized can be variable, but in most upland forest areas in the U.S. most of the atmospheric deposition is retained in the soil (Nadelhoffer et al., 1999). Lovett et al. (2000) found immobilization of N to be 49% to 90% of the atmospheric input based on N measured in stream water because of factors such as vegetation type, age of vegetation, soil type, soil condition, the amount of nitrification, management activities, etc. that control the amount of N accumulating. Several different data compilations indicate also that 80 to 100% of N deposition is retained or denitrified within terrestrial ecosystems that receive less than about 10 kg N/ha-yr (2008 ISA,, section 3.3.2.1; 2020 ISA, Appendix 4, section 4.6.2.2). Using compiled data collected during the mid- to late 1990s and focusing on lakes and streams in 83 forested watersheds of the Northeast, Aber et al. (2003) suggested that in northeastern watersheds that receive less than about 8 to 10 kg N/ha-yr, nearly all N deposition is retained or denitrified (ISA, Appendix 4, section). In the West, a study of mixed conifer forests of the Sierra Nevada and San Bernardino Mountains estimated 17 kg N/ha-yr as the N deposition load associated with the onset of NO_3 leaching (Fenn et al., 2011). Two studies in the Rocky Mountains indicated that the onset of NO_3 leaching in alpine catchments occurs at approximately 10 kg N/ha-yr (Baron et al., 1994; Williams and Tonnesen, 2000).

Nitrogen is removed or exported from the watershed by being volatilized in fires, denitrified or leached to drainage waters (ISA, Appendix 4, sections 4.3 and 4.7). Denitrification is the process by which nitrate is converted into gaseous N, most commonly in water saturated soil, and returned to the atmosphere. Like with immobilization, many factors control the rate of denitrification, making it difficult to estimate on a site-by-site basis without directly measuring it. Accordingly, rates vary widely across sites. For example, Groffman (1994) observed rates of denitrification of 4 to 135 kg N/ha-yr in very poorly drained soils on nutrient-rich parent material and rates of 1.2 to 5.3 kg N/ha-yr in soils that were better-drained or less nutrient-rich (2008 ISA, section 3.3.2.1; 2020 ISA, Appendix 4, Table 4-7). The N remaining, that isn't volatilized, denitrified, or immobilized, can be leached in drainage water as nitrate or dissolved organic nitrogen (DON) and has the potential to acidify surface waters. Nitrate concentrations or concentrations of DON in streams impacted by acidification (typically 1-3 order streams) are often very low, near zero, during the growing season when the N entering the watershed is incorporated into soil or vegetation (Campbell et al., 2000; MacDonald et al., 2002; Dise et al., 2009).

Recent studies from some regions of the U.S. (e.g., Eshleman et al., 2013; Driscoll et al., 2016; Strock et al., 2014; Eshleman and Sabo, 2016; ISA, Appendix 7, section 7.1.5.1) showed

declines in concentrations of NO_3^- in surface waters that are consistent with declines in N deposition. Using the Lake Multi-Scaled Geospatial and Temporal Database of the Northeast Lakes of the U.S. (LAGOS-NE) containing water quality data from 2,913 lakes, Oliver et al. (2017) identified atmospheric deposition as the main driver of declines in total N (TN) deposition and lake TN:total P (TP) ratios from 1990 to 2011. In additional, monitored lakes and streams as part of the EPA's Long-term Monitoring (LTM) program have average annual nitrate concentrations of 9.5 and 7.64 µeq/L, respectively, from 1990 to 2018 (Table 5A-2).[10] Average annual nitrate concentrations have decreased during the past decade to 7.19 and 4.40 µeq/L. These areas receive 5 to 8 kg N/ha-yr deposition annually.

Table 5A-2. Average annual nitrate concentrations for the EPA's Long-term Monitoring program for lakes and streams.

Areas	Years	Average (95% CI) (µeq/L)
New England Lakes	1990 – 2018	2.36 (2.155 – 2.565)
	1990 – 1999	2.33 (1.947 – 2.713
	2000 – 2009	2.45 (2.165 – 2.745)
	2010 – 2018	0.56 (0.46 – 0.66)
Adirondacks Lakes	1990 – 2018	16.64 (15.966 – 17.318)
	1990 – 1999	18.48 (17.183 – 19.779)
	2000 – 2009	16.70 (15.602 – 17.796)
	2010 – 2018	13.82 (12.736 – 14.907)
Appalachian Streams	1990 – 2018	7.64 (7.092 – 8.187)
	1990 – 1999	11.50 (10.334 – 12.675)
	2000 – 2009	6.59 (5.774 – 7.40)
	2010 – 2018	4.40 (3.744 – 5.049)

We recognize that estimating the contribution of N deposition to acidification of surface waters is difficult and uncertain because N cycling in an ecosystem is inherently variable and data are limited across the U.S. to model it, however, it is important to the review that an estimate be determined for aquatic acidification. Given the availability of data and what was used in the 2008 review, we chose the N leaching method to estimate the contribution of total N deposition to acidification that uses water quality and runoff data to estimate the amount of total N deposition leaching to the drainage water that acidify (Henriksen and Posch, 2001).

This method is based on Henriksen and Posch (2001) where the exceedance for these CLs is determined using the Nle (see Eq. 5A-10):

[10] The EPA's Long-Term Monitoring program tracks changes in surface water chemistry in the four regions shown below, known to be sensitive to acid rain: New England, the Adirondack Mountains, the Northern Appalachian Plateau, and the central Appalachians (https://www.epa.gov/power-sector/monitoring-surface-water-chemistry#tab-6). Data from this program are available at: https://doi.org/10.23719/1518546.

$$Ex(N+S) = \text{Total S deposition} + Nle - CLS \quad \textbf{(5A-10)}$$

Where:

Nle = the sum of the measured concentrations of nitrate (NO_3^- μeq/L) and ammonia (NH_4^+ μeq/L) in the runoff (Qs m/yr) as ($[NO_3^-]+[NH_4^+]$)*Qs.

Factoring in the CL uncertainty, Eq. 5A-11 is:

$$Ex(N+S) = ((\text{Total S deposition} + Nle) - CLS) > 3.125 \text{ meq } /m^2\text{-yr} \quad \textbf{(5A-11)}$$

The advantage of using the leaching estimate, Nle, (units of meq/m^2-yr) is that for some waterbodies it is based on measured water quality parameters that integrate all the N processes occurring in the watershed. However, it is an indicator of conditions at the time of the measurement, which may or may not be representative of long-term leaching. Steady-state CLs are intended to represent the long-term leaching amount, which may or may not be well represented under current conditions. For example, if a forest is a watershed is young, it would be growing fast, and be able to immobilize most of the N deposition. However, that would not be the case for old growth forests, which leach N at a much higher rate than younger forests (Goodale et al., 2000). Old growth forests are thought of as the steady-state condition. If future forests are older, then the leaching estimate based on current water quality would under-estimate the acidification affect. But if future forests are like today's forests, then the leaching value would better represent acidification impacts. Further studies in other old-growth forests are needed to better understand the mechanisms causing long-term change in N cycling with forest development (ISA Appendix 4, sections 4.3.2 and 4.3.6).

The Nle estimate, which is used for calculating the contribution to acidification from N deposition is based on the calculated flux of N to the waterbody. This is estimated by multiplying the concentration of nitrate as N within the waterbody by the annual surface water runoff to the waterbody. Nitrogen leaching measurements are not typically collected across the U.S. For that reason, annual leaching is estimated as a function of annual runoff (eq 5A-10), which we recognize is a source of uncertainty for this estimated value. We chose to use an annual runoff (based on 30-year "Normals"[11]) that is included as a catchment parameter in the national hydrology dataset developed by the U.S. EPA and U.S. Geological Survey (NHDplus, version

11 A "normal" is the 30-year average of a particular variable's measurements, calculated for a uniform time period. Climate normals are derived from weather and climate observations captured by weather stations. The official normals are calculated by the National Centers for Environmental Information at the U.S. NOAA for a uniform 30 year period, and consist of annual/seasonal, monthly, daily, and hourly averages and statistics of temperature, precipitation, and other climatological variables from almost 15,000 U.S. weather stations. (https://www.ncei.noaa.gov/products/land-based-station/us-climate-normals#:~:text=A%20%22normal%22%20is%20the%2030,observations%20captured%20by%20weather%20stations.).

2).[12] Site-specific catchment annual runoff values were used for each waterbody with a CL. We decided to use these annual runoff values because they are expected to better reflect long-term and temporal patterns in runoff relevant to the mass-balance steady-state CL approach.

Some nitrate measurements used to estimate the Nle value date back to the 1980-1990s and for that reason may not reflect more recent N leaching rates. Also, many of the waterbodies with CLs have no nitrate measurements, hence, no way to calculate the leaching directly. Another limitation is that a single water quality measurement is from a single sample which cannot reflect the variability of nitrate during the year and for that reason may over or under-estimate Nle. For waterbodies with no or few nitrate measurements, a "regional approach," described immediately below, was used to estimate Nle values in equation 5A-11. We recognize this regional approach provides additional uncertainty to the leaching estimate, as recognized in section 5A.3.2 below; however, it provides an integrated regional average estimate that is based on numerous available water quality data and long-term runoff data at the catchment level where the waterbody is located. We recognize that multiple water quality measurements over many years for each waterbody and waterbody specific runoff or flow would be more desirable to estimate the contribution of N deposition that is acidifying deposition, however, those data are not readily available.

The regional aggregation was done for level III and level II and level I ecoregions. Water quality data associated with the CLs was drawn from the NCLD, version 3.2, and was supplemented with data from EPA's LTM program.[13] We decide to focus on the water quality data within the NCLD, version 3.2, because they represent the type of waterbodies (i.e., small lakes/ponds, 1-3 order streams, etc.) that the CLs are based on. Measurements from within each ecoregion III, II, and I were averaged to create three different values from which a single aggregated value was chosen to replace the measured value for the CL. The ecoregion average for level III was used unless there were fewer than 30 water quality measurements, in which case the level II ecoregion average was used, and if there were fewer than 30 measurements in level II, the level I ecoregion average was used. See Table 5A-3 for the number of measured used in the aggregation and Nle value.

[12] This dataset is available at: https://www.epa.gov/waterdata/nhdplus-national-hydrography-dataset-plus.
[13] These data, equaling 16,900+ measurements across the CONUS were downloaded from https://www.epa.gov/power-sector/monitoring-surface-water-chemistry#tab-6 in February 2020.

Table 5A-3. Regional aggregation of N leaching for ecoregion II and III, based on water quality data for sites in NCLD, version 3.2.

Name	Code	No.Sites	Average N Leaching (meq/m^2-yr)
Ecoregion III			
Northern Appalachian and Atlantic Maritime Highlands	5.3.1	3729	0.7
Blue Ridge	8.4.4	2703	1.7
Southern Rockies	6.2.14	444	1.2
Ridge and Valley	8.4.1	1719	3.0
Middle Rockies	6.2.10	552	1.3
Sierra Nevada	6.2.12	566	1.0
Northern Lakes and Forests	5.2.1	894	0.6
Acadian Plains and Hills	8.1.8	630	0.5
Piedmont	8.3.4	573	4.8
Northeastern Coastal Zone	8.1.7	526	1.8
Central Appalachians	8.4.2	495	3.0
Idaho Batholith	6.2.15	212	8.8
Cascades	6.2.7	229	1.4
Southeastern Plains	8.3.5	413	5.6
Northern Piedmont	8.3.1	265	16.1
Wasatch and Uinta Mountains	6.2.13	114	1.7
Atlantic Coastal Pine Barrens	8.5.4	263	5.2
North Central Appalachians	5.3.3	230	2.7
Northern Allegheny Plateau	8.1.3	224	3.3
North Cascades	6.2.5	169	1.0
South Central Plains	8.3.7	157	0.6
Southwestern Appalachians	8.4.9	127	2.4
Columbia Mountains/Northern Rockies	6.2.3	96	0.9
Southern Coastal Plain	8.5.3	149	1.6
Middle Atlantic Coastal Plain	8.5.1	118	13.5
Coast Range	7.1.8	119	4.0
Eastern Great Lakes Lowlands	8.1.1	92	1.5
Klamath Mountains	6.2.11	85	1.2
North Central Hardwood Forests	8.1.4	101	1.7
Interior Plateau	8.3.3	89	7.2
Blue Mountains	6.2.9	65	0.3
Ozark Highlands	8.4.5	61	4.0
Eastern Cascades Slopes and Foothills	6.2.8	32	0.6
Ouachita Mountains	8.4.8	51	3.2
Strait of Georgia/Puget Lowland	7.1.7	39	4.5
Mississippi Valley Loess Plains	8.3.6	41	0.4
Arkansas Valley	8.4.7	39	1.7

Name	Code	No.Sites	Average N Leaching (meq/m^2-yr)
Arizona/New Mexico Mountains	13.1.1	27	NA
California Coastal Sage, Chaparral, and Oak Woodlands	11.1.1	25	NA
Central Basin and Range	10.1.5	17	NA
Western Allegheny Plateau	8.4.3	37	2.4
Northern Basin and Range	10.1.3	20	NA
Southern Michigan/Northern Indiana Drift Plains	8.1.6	36	0.4
Canadian Rockies	6.2.4	32	1.4
Cross Timbers	9.4.5	31	0.8
Ecoregion II			
Atlantic Highlands	5.3	3960	0.85
Mixed Wood Plains	8.1	1639	1.51
Ozark/Ouachita-Appalachian Forests	8.4	5259	2.34
Southeastern USA Plains	8.3	1568	6.55
Mississippi Alluvial and Southeast USA Coastal Plains	8.5	551	5.85
Mixed Wood Shield	5.2	896	0.62
Temperate Prairies	9.2	51	1.57
Western Cordillera	6.2	2596	1.78
South Central Semi-Arid Prairies	9.4	48	0.65
Upper Gila Mountains	13.1	27	2.23
Mediterranean California	11.1	49	0.70
Marine West Coast Forest	7.1	182	4.72
Cold Deserts	10.1	46	2.83

5A.1.7 Ecoregions Sensitivity to Acidification

The CONUS areas that have been described as sensitive to aquatic acidification include the Northeast, Southeast, and upper Midwest, and to lesser extent, some areas of the Rocky Mountains, Sierra Nevada Mountains, and the Pacific Northwest (Figure 5A-7; ISA, Appendix 8, section 8.5). Area of the Appalachian Mountains (which extend from Maine to Georgia) are particularly sensitive (ISA, Appendix 8, section 8.5). Ecoregions are used here as the unit of spatial aggregation to characterize the level of acidification in sensitive areas across the CONUS. National patterns of surface water alkalinity in the conterminous U.S. based on data collected prior to 1988 (U.S. EPA, 2012) and modern ANC water quality measurements were used to define which ecoregions were considered acid sensitive. The EPA's Total Alkalinity GIS layer was developed in 1980's using water quality data to define regions of acid sensitivity, as shown in Figure 5A-7a (Omernik and Powers 1983). Additionally, over 15,000 water quality ANC measurements, collected across the CONUS for the period from 1990 to 2018 by multiple water quality networks, programs, and research groups, have been summarized in Figure 5A-7b**.**

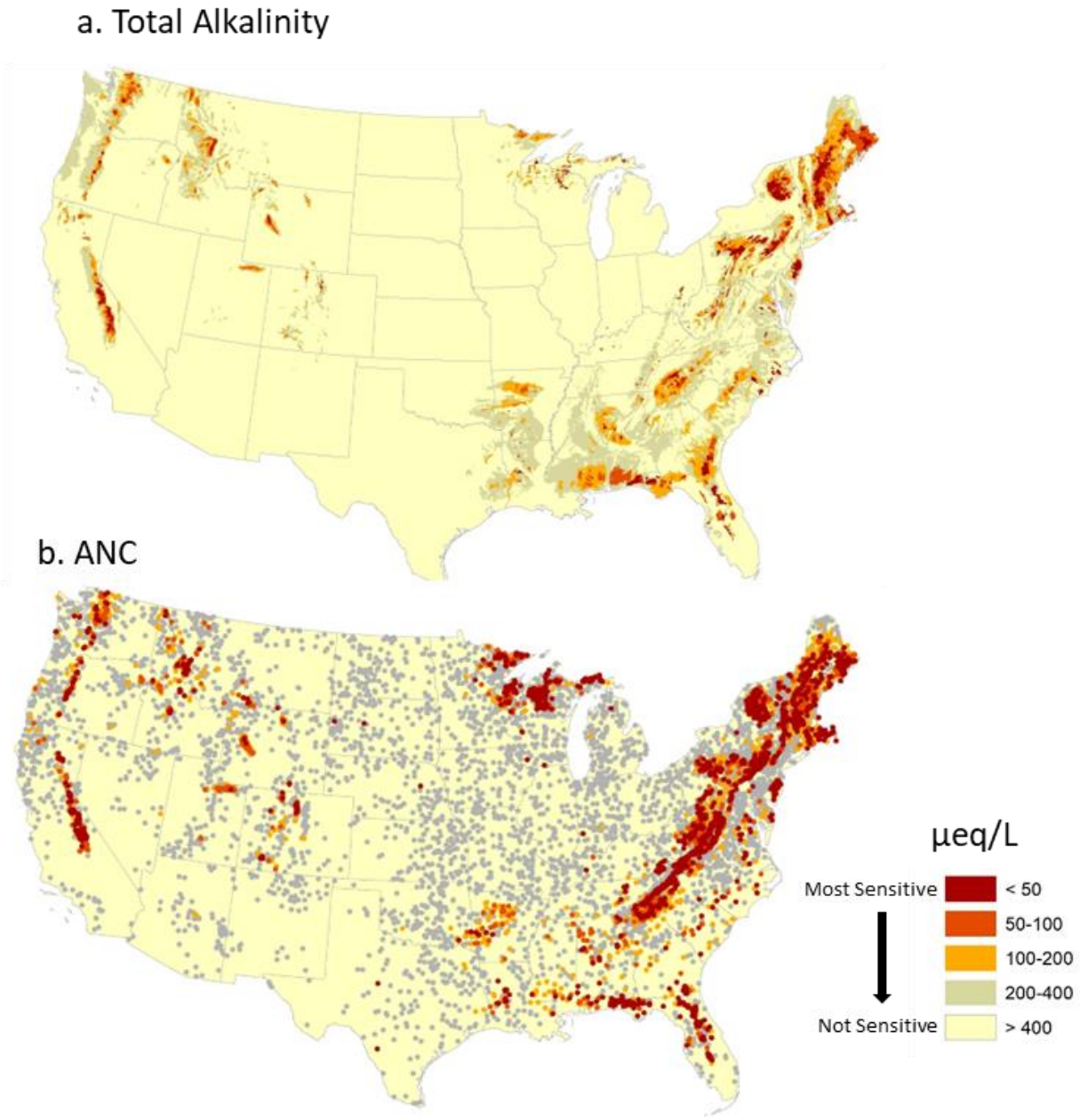

Figure 5A-7. Surface water quality alkalinity (a) and ANC (b) across the CONUS based on measurements collected prior to 1988 through 2018.

Water quality measurements of ANC and total alkalinity (Omernik and Powers, 1983) were used to classify the 84 CONUS ecoregion IIIs into four acid sensitive classes: (1) most acid sensitive (<50 µeq/L), (2) acid sensitive (50-100 µeq/L), (3) moderately acid sensitive (100-200 µeq/L), and (4) low or no acid sensitivity (>200 µeq/L). The four categories are based on what Omernik and Powers (1983) and Greaver et al. (2012) used in their assessment (Table 5A-4). A total of 24 ecoregions III were acid sensitive and another 6 ecoregions were moderately acid sensitive for a total of 30 (Table 5A-5). Fifty-four ecoregions had low or no evidence of acid

sensitivity across the CONUS (Table 5A-5 and Figure 5A-8). The acid sensitive ecoregions generally are areas with mountains, high elevation terrain or water bodies in northern latitudes (northern areas of Minnesota, Wisconsin and Michigan; and New England). The northern, non-mountainous regions share attributes similar to mountainous regions (e.g., growing season, vegetation, soils, geology) and are typically in rural areas, often in designated wilderness, park and recreation areas. Of the 30 acid sensitive ecoregions, the following three ecoregions are located on eastern coastal plain: (1) Middle Atlantic Coastal Plain (8.5.1), (2) Southern Coastal Plains (8.5.3), and (3) Atlantic Coastal Pine Barrens (8.5.4). Waterbodies in these ecoregions tend to have higher DOC values >10 mg/L, which is indicative of natural acidity (ISA, Appendix 7, section 7.1.2.5; 2008 ISA, section 3.2.4.2 and Annex B, p. B-35).

Table 5A-4. Acid sensitive categories and criteria used to define each one.

Acid Sensitive Category	Criteria
Most Acid Sensitive Ecoregions	>25 ANC values* less than 100 µeq/L, > 75 ANC values from 100-200 µeq/L and have total alkalinity areas < 50 µeq/L
Acid Sensitive Ecoregions	>10 ANC values less than 100 µeq/L, > 40 ANC values from 100-200 µeq/L and have total alkalinity areas < 100 µeq/L
Moderately Sensitive Ecoregions	>5 ANC values less than 100 µeq/L, > 20 ANC values from 100-200 µeq/L and have total alkalinity areas < 200 µeq/L
Low or Non-sensitive Ecoregions	<5 ANC values less than 100 µeq/L, < 20 ANC values from 100-200 µeq/L and have total alkalinity areas >200 µeq/L
* The four categories are based on what Omernik and Powers (1983) and Greaver et al. (2012) used in their assessment.	

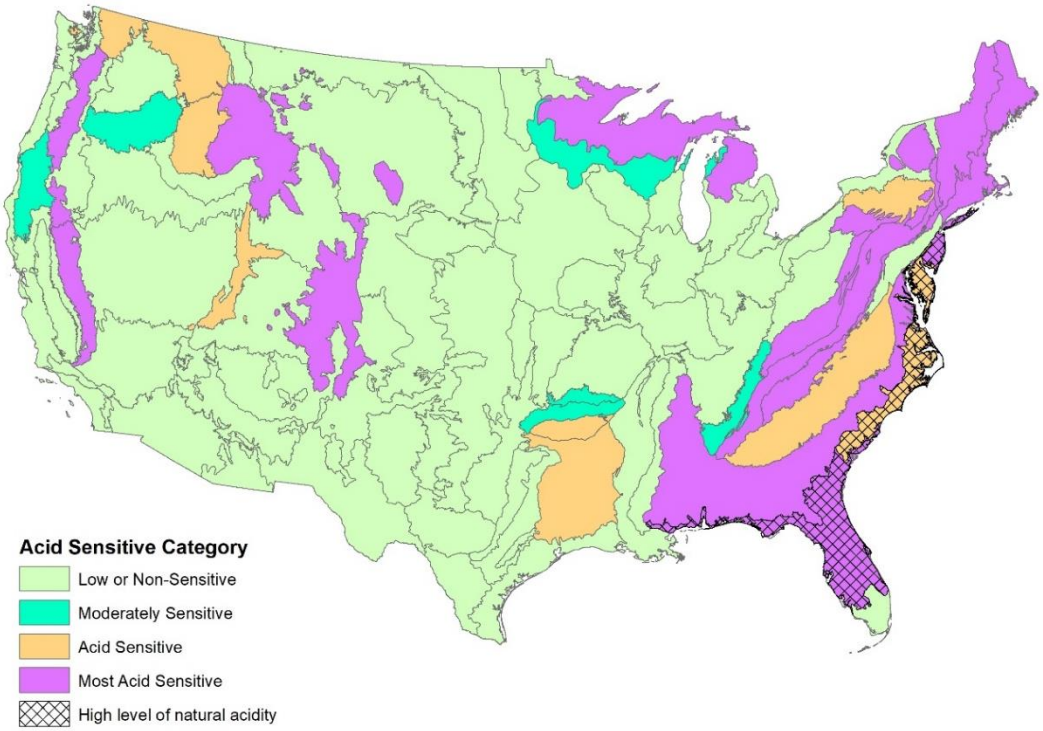

Figure 5A-8. Level III ecoregions grouped into acid sensitivity categories.

Table 5A-5. Level III ecoregion categorization for acid sensitivity.

Ecoregion III Code	No. Critical Loads	Total No. ANC Values	No. ANC values <100 µeq/L	No. ANC values <200 µeq/L	Total Alkalinity Area (µeq/L)	Acid Sensitive Category
5.3.1	2851	2053	901	1302	50	Most Acid Sensitive Ecoregion
8.4.4	1972	1136	619	916	50	Most Acid Sensitive Ecoregion
8.4.1	1292	1394	459	733	50	Most Acid Sensitive Ecoregion
5.2.1	839	1074	398	535	50	Most Acid Sensitive Ecoregion
8.1.7	565	488	88	201	50	Most Acid Sensitive Ecoregion
6.2.10	496	323	61	127	50	Most Acid Sensitive Ecoregion
8.1.8	494	492	197	316	50	Most Acid Sensitive Ecoregion
8.3.5	390	432	141	211	50	Most Acid Sensitive Ecoregion
8.4.2	372	420	229	282	50	Most Acid Sensitive Ecoregion
6.2.14	372	327	56	107	50	Most Acid Sensitive Ecoregion
6.2.12	353	359	224	279	50	Most Acid Sensitive Ecoregion
8.5.4	234	130	78	100	50	Most Acid Sensitive Ecoregion
5.3.3	216	242	113	177	50	Most Acid Sensitive Ecoregion
6.2.7	179	244	80	129	50	Most Acid Sensitive Ecoregion
8.5.3	142	228	115	132	50	Most Acid Sensitive Ecoregion
8.3.4	508	455	28	84	50	Acid Sensitive Ecoregion
8.1.3	199	223	13	42	50	Acid Sensitive Ecoregion
6.2.15	188	164	60	95	50	Acid Sensitive Ecoregion
6.2.5	162	155	40	80	50	Acid Sensitive Ecoregion
8.3.7	153	165	17	41	50	Acid Sensitive Ecoregion
6.2.13	96	139	26	61	100	Acid Sensitive Ecoregion
6.2.3	86	147	13	31	50	Acid Sensitive Ecoregion
8.4.8	42	73	17	44	50	Acid Sensitive Ecoregion
8.4.9	117	64	19	32	50	Moderately Sensitive Ecoregion
8.5.1	105	183	14	37	50	Acid Sensitive Ecoregion
8.1.4	94	162	12	21	50	Moderately Sensitive Ecoregion
8.4.7	31	59	9	25	100	Moderately Sensitive Ecoregion
8.3.1	231	211	3	6	50	Low or Non-Sensitive Ecoregion
7.1.8	115	154	4	13	200	Low or Non-Sensitive Ecoregion
8.1.1	83	97	1	2	50	Low or Non-Sensitive Ecoregion
6.2.11	81	105	5	11	50	Moderately Sensitive Ecoregion
8.3.3	71	114	0	2	200	Low or Non-Sensitive Ecoregion
6.2.9	63	91	5	16	50	Moderately Sensitive Ecoregion
8.4.5	56	111	0	0	>200	Low or Non-Sensitive Ecoregion
8.3.6	41	61	2	13	200	Low or Non-Sensitive Ecoregion
7.1.7	38	51	3	7	50	Low or Non-Sensitive Ecoregion
8.4.3	35	114	0	2	50	Low or Non-Sensitive Ecoregion

Ecoregion III Code	No. Critical Loads	Total No. ANC Values	No. ANC values <100 µeq/L	No. ANC values <200 µeq/L	Total Alkalinity Area (µeq/L)	Acid Sensitive Category
8.1.6	33	131	0	0	>200	Low or Non-Sensitive Ecoregion
6.2.4	31	42	3	5	100	Low or Non-Sensitive Ecoregion
6.2.8	27	43	0	1	50	Low or Non-Sensitive Ecoregion
9.4.5	26	96	0	0	>200	Low or Non-Sensitive Ecoregion
9.2.3	26	180	0	0	>200	Low or Non-Sensitive Ecoregion
13.1.1	25	64	0	3	>200	Low or Non-Sensitive Ecoregion
7.1.9	24	28	0	0	>200	Low or Non-Sensitive Ecoregion
8.4.6	23	31	3	20	100	Moderately Sensitive Ecoregion
11.1.3	22	19	0	0	>200	Low or Non-Sensitive Ecoregion
9.2.4	21	114	0	0	>200	Low or Non-Sensitive Ecoregion
11.1.1	21	57	0	0	>200	Low or Non-Sensitive Ecoregion
10.1.3	20	80	0	4	>200	Low or Non-Sensitive Ecoregion
8.5.2	19	91	0	0	200	Low or Non-Sensitive Ecoregion
8.3.2	18	115	5	5	50	Low or Non-Sensitive Ecoregion
9.5.1	16	36	0	0	200	Low or Non-Sensitive Ecoregion
10.1.5	16	87	0	2	200	Low or Non-Sensitive Ecoregion
8.1.5	15	80	1	1	>200	Low or Non-Sensitive Ecoregion
8.1.10	14	63	0	0	100	Low or Non-Sensitive Ecoregion
8.2.4	14	96	0	0	>200	Low or Non-Sensitive Ecoregion
8.3.8	10	27	0	0	200	Low or Non-Sensitive Ecoregion
8.2.1	10	38	1	1	>200	Low or Non-Sensitive Ecoregion
9.4.4	7	34	0	0	>200	Low or Non-Sensitive Ecoregion
9.4.2	5	144	2	3	>200	Low or Non-Sensitive Ecoregion
10.1.4	3	56	0	1	200	Low or Non-Sensitive Ecoregion
9.4.7	3	25	0	0	>200	Low or Non-Sensitive Ecoregion
5.2.2	2	26	1	1	200	Low or Non-Sensitive Ecoregion
10.1.8	2	11	0	0	200	Low or Non-Sensitive Ecoregion
11.1.2	2	14	0	0	>200	Low or Non-Sensitive Ecoregion
10.1.2	2	32	0	0	>200	Low or Non-Sensitive Ecoregion
8.2.3	2	42	0	0	>200	Low or Non-Sensitive Ecoregion
9.3.1	2	114	0	0	>200	Low or Non-Sensitive Ecoregion
10.1.6	1	51	0	0	200	Low or Non-Sensitive Ecoregion
10.2.1	0	6	0	0	>200	Low or Non-Sensitive Ecoregion
9.3.4	0	22	0	0	>200	Low or Non-Sensitive Ecoregion
9.4.6	0	19	0	0	>200	Low or Non-Sensitive Ecoregion
9.4.1	0	54	0	0	>200	Low or Non-Sensitive Ecoregion
9.2.1	0	66	0	0	>200	Low or Non-Sensitive Ecoregion
9.4.3	0	55	0	0	>200	Low or Non-Sensitive Ecoregion

Ecoregion III Code	No. Critical Loads	Total No. ANC Values	No. ANC values <100 µeq/L	No. ANC values <200 µeq/L	Total Alkalinity Area (µeq/L)	Acid Sensitive Category
10.1.7	0	21	0	0	>200	Low or Non-Sensitive Ecoregion
9.3.3	0	270	3	3	>200	Low or Non-Sensitive Ecoregion
12.1.1	0	11	0	0	>200	Low or Non-Sensitive Ecoregion
8.2.2	0	27	0	0	>200	Low or Non-Sensitive Ecoregion
15.4.1	0	5	0	0	>200	Low or Non-Sensitive Ecoregion
9.2.2	0	20	0	0	>200	Low or Non-Sensitive Ecoregion
10.2.4	0	14	0	0	>200	Low or Non-Sensitive Ecoregion
10.2.2	0	15	0	0	>200	Low or Non-Sensitive Ecoregion
9.6.1	0	7	1	1	>200	Low or Non-Sensitive Ecoregion
Note: Ecoregion III code in bold indicates the 25 ecoregions that are the focus of the ecoregion analyses described in section 5A.2.2.						

5A.2 ANALYSIS RESULTS

The aquatic acidification assessment is intended to estimate the ecological exposure and risk posed to aquatic ecosystems from the acidification effects of S and/or N deposition to sensitive regions across the CONUS. The CL itself indicates how sensitive the waterbody is to inputs of acidic deposition of S and/or N. In Figure 5A-6, a CL indicates the amount of acidic input of total S and/or N deposition that a waterbody can neutralize and still maintain an ANC of 50 µeq/L. Watersheds with CL values less than 100 meq/m^2-yr (red and orange circles) are most sensitive to surface water acidification, whereas watersheds with values greater than 100 meq/m^2-yr (yellow and green circles) are the least sensitive sites. Most sensitive waterbodies are located along the Appalachian Mountains range, the upper Mid-west, and the Rocky Mountain range in the west, which correspond to the same regions as the acid sensitive ecoregions (Figure 5A-7).

5A.2.1 Results of National Scale Assessment of Risk

A total of 13,824 unique waterbodies across the CONUS had calculated CLs available in NCLD v3.2. Table 5A-6 summarizes the percent of waterbodies with CLs that are less than 2, 6, 12, 18 kg S/ha, indicating most CLs used in this analysis are less than 18 kg S/ha. Table 5A-7 contains a summary of CL exceedances for S only and S and N combined for average annual deposition estimates for 2018-20, 2014-16, 2010-12, 2006-08, and 2001-03. An exceedance indicates that the estimated deposition for a period is greater than the amount of deposition the waterbodies are estimated to be able to neutralize and still maintain the ANC thresholds of 20, 30, and 50 µeq/L.

Table 5A-6. Percent of waterbodies with critical loads less than 2, 6, 12, and 18 kg S/ha-yr based on ANC thresholds of 20, 30, and 50 µeq/L.

Critical Load, kg/ha-yr (meq/m²-yr)	Percent of Waterbodies with CL for specified ANC threshold below 2, 6, 12 and 18 kg/ha-hr		
	20 µeq/L	30 µeq/L	50 µeq/L
<2 (12.5)	3%	5%	11%
<6 (37.5)	14%	17%	25%
<12 (75)	36%	39%	45%
<18 (112.5)	52%	55%	58%

Table 5A-7. Summary of CL exceedances, nationally, by ANC thresholds and deposition periods.

ANC Threshold	S Only CL Exceedances[A]		'S and N' CL Exceedances[A]	
	All Values[B]	CL≥0 Values Only[C]	All Values[B]	CL≥0 Values Only[C]
Deposition estimates for 2018-20				
20	2%	1%	2%	2%
30	3%	2%	4%	2%
50	9%	4%	9%	5%
50/20	7%	4%	8%	4%
Deposition estimates for 2014-16				
20	3%	3%	3%	3%
30	5%	4%	5%	4%
50	11%	6%	12%	7%
50/20	10%	6%	10%	7%
Deposition estimates for 2010-12				
20	5%	5%	6%	5%
30	8%	7%	9%	7%
50	15%	11%	16%	11%
50/20	14%	10%	15%	11%
Deposition estimates for 2006-08				
20	17%	16%	18%	17%
30	21%	19%	21%	20%
50	28%	24%	29%	25%
50/20	27%	23%	28%	24%
Deposition estimates for 2001-03				
20	22%	22%	23%	23%
30	26%	25%	27%	25%
50	33%	28%	33%	29%
50/20	31%	28%	32%	28%

[A] An exceedance is deposition above the CL and error of 3.125 meq/m2-yr.
[B] "All Values" includes all critical loads.
[C] "CL>0 Values" includes only critical loads greater than 0.

Table 5A-8 includes both numbers and percent exceedances for the CONUS for the four-deposition time periods and three ANC thresholds. Exceedance rates (e,g, percent of waterbodies that exceed the CL) are lowest for the ANC threshold of 20 μeq/L and highest for the ANC threshold of 50 μeq/L. For the most recent deposition period of 2018-20, 2%, 3%, and 9% of the modeled waterbodies received levels of total S deposition that exceeded their CL with CL thresholds of 20, 30, and 50 μeq/L, respectively. The percentage of waterbodies exceeding a CL for combined total S and N are slightly higher than S only percentages at 2%, 4%, and 9% of the modeled waterbodies for CL thresholds of 20, 30, and 50 μeq/L based on Nle. This indicates that most of the N deposition entering the watershed is retained within the watershed and/or converted to gaseous N (e.g., N_2O, N_2, etc.). For all other deposition time periods, exceedance rates are similar or only slightly higher (1-2%) when considering both N and S deposition compared to just S deposition only.

Table 5A-8. Comparison of estimated deposition to CLs nationally based on <u>all CL</u> values by ANC thresholds and deposition periods.

ANC Threshold	Class	Sulfur Only CLs		Sulfur and Nitrogen CLs	
		No.	Percent	No.	Percent
Deposition estimates for 2018-20					
20	>CL	234	2%	266	2%
	<CL	13375	97%	13333	96%
	at CL	215	2%	225	2%
30	>CL	452	3%	496	4%
	<CL	13078	95%	13033	94%
	at CL	294	2%	295	2%
50	>CL	1203	9%	1262	9%
	<CL	12218	88%	12132	88%
	at CL	403	3%	430	3%
50/20	>CL	1023	7%	1075	8%
	<CL	12416	90%	12344	89%
	at CL	385	3%	405	3%
Deposition estimates for 2014-16					
20	>CL	423	3%	465	3%
	<CL	13137	95%	13089	95%
	at CL*	264	2%	270	2%
30	>CL	680	5%	724	5%
	<CL	12807	93%	12730	92%
	at CL*	337	2%	370	3%
50	>CL	1512	11%	1591	12%
	<CL	11859	86%	11750	85%
	at CL*	453	3%	483	3%
50/20	>CL	1324	10%	1400	10%
	<CL	12060	87%	11959	87%
	at CL*	440	3%	465	3%

ANC Threshold	Class	Sulfur Only CLs		Sulfur and Nitrogen CLs	
		No.	Percent	No.	Percent
Deposition estimates for 2010-12					
20	>CL	748	5%	798	6%
	<CL	12731	92%	12670	92%
	at CL*	345	2%	356	3%
30	>CL	1122	8%	1192	9%
	<CL	12271	89%	12190	88%
	at CL*	431	3%	442	3%
50	>CL	2114	15%	2215	16%
	<CL	11206	81%	11099	80%
	at CL*	504	4%	510	4%
50/20	>CL	1918	14%	2013	15%
	<CL	11424	83%	11324	82%
	at CL*	482	3%	487	4%
Deposition estimates for 2006-08					
20	>CL	2328	17%	2433	18%
	<CL	10994	80%	10871	79%
	at CL*	502	4%	520	4%
30	>CL	2845	21%	2962	21%
	<CL	10450	76%	10322	75%
	at CL*	529	4%	540	4%
50	>CL	3911	28%	4035	29%
	<CL	9384	68%	9266	67%
	at CL*	529	4%	523	4%
50/20	>CL	3710	27%	3825	28%
	<CL	9609	70%	9492	69%
	at CL*	505	4%	57	4%
Deposition estimates for 2001-03					
20	>CL	3064	22%	3191	23%
	<CL	10271	74%	10156	73%
	at CL*	489	4%	477	3%
30	>CL	3587	26%	3694	27%
	<CL	9784	71%	9683	70%
	at CL*	453	3%	447	3%
50	>CL	4504	33%	4611	33%
	<CL	8905	64%	8807	64%
	at CL*	415	3%	406	3%
50/20	>CL	4313	31%	4410	32%
	<CL	9124	66%	9030	65%
	at CL*	387	3%	384	3%

An exceedance (>CL) is where deposition for the modeled waterbodies is above the CL and error of 3.125 meq/m^2-yr.
"at CL" indicates estimated deposition is within 3.125 meq/m^2-yr of the CL.
This summary includes CLs below zero.

Table 5A-9 includes both numbers of waterbodies and percent exceedances for the CONUS for the four-deposition time periods and four ANC thresholds where CLs less than or

equal to zero were removed from the exceedance counts and percentages. Sites with CLs less than or equal to zero are very sensitive waterbodies that naturally could not meet the ANC threshold at any level of deposition. When zero and negative CLs are excluded, in the most recent deposition period of 2018-2020, 1%, 2%, 4%, and 4% of the modeled waterbodies received levels of total S deposition that exceeded CLs for ANC thresholds of 20, 30, 50, and 50/20 μeq/L, respectively (Table 5A-9). The percent exceedances for combined total S and/or N CLs are only slightly higher than for S only CLs at 2%, 2%, 5%, and 4% of the modeled waterbodies for ANC thresholds of 20, 30, 50, and 50/20 μeq/L based on Nle (Table 5A-9). For the deposition period of 2001-2003, exceedance percentages for Sulfur only were much higher than in the 2018-2020 period, at 22%, 25%, 29, and 29% for ANC thresholds of 20, 30, 50, and 50/20 μeq/L (Table 5A-9). The percent of modeled waterbodies with negative CLs is the lowest for an ANC threshold of 20 μeq/L at 0.4% and the highest for an ANC threshold of 50 μeq/L at 4.6% (Figure 5A-9).

Table 5A-9. National aquatic CL exceedances based on <u>CLs greater than 0</u> by ANC thresholds and deposition periods.

ANC Threshold	Class	Sulfur Only CLs		Sulfur and Nitrogen CLs	
		No.	Percent	No.	Percent
Deposition estimates for 2018-20					
20	>CL	182	1%	214	2%
	<CL	13375	97%	13333	97%
	at CL*	215	2%	225	2%
30	>CL	279	2%	323	2%
	<CL	13078	96%	13033	95%
	at CL*	293	2%	294	2%
50	>CL	566	4%	624	5%
	<CL	12218	93%	12132	92%
	at CL*	401	3%	429	3%
5020	>CL	544	4%	596	4%
	<CL	12416	93%	12344	92%
	at CL*	385	3%	405	3%
Deposition estimates for 2014-16					
20	>CL	371	3%	413	3%
	<CL	13137	95%	13089	95%
	at CL*	264	2%	270	2%
30	>CL	506	4%	550	4%
	<CL	12807	94%	12730	93%
	at CL*	337	2%	370	3%
50	>CL	873	7%	952	7%
	<CL	11859	90%	11750	89%
	at CL*	453	3%	483	4%
5020	>CL	845	6%	921	7%
	<CL	12060	90%	11959	90%
	at CL*	440	3%	465	3%

ANC Threshold	Class	Sulfur Only CLs		Sulfur and Nitrogen CLs	
		No.	Percent	No.	Percent
Deposition estimates for 2010-12					
20	>CL	696	5%	746	5%
	<CL	12731	92%	12670	92%
	at CL*	345	3%	356	3%
30	>CL	948	8%	1018	7%
	<CL	12271	89%	12190	89%
	at CL*	431	3%	442	3%
50	>CL	1475	15%	1576	12%
	<CL	11206	81%	11099	84%
	at CL*	504	4%	510	4%
5020	>CL	1439	14%	1534	11%
	<CL	11424	83%	11324	85%
	at CL*	482	3%	487	4%
Deposition estimates for 2006-08					
20	>CL	2276	17%	2381	17%
	<CL	10994	80%	10871	79%
	at CL*	502	4%	520	4%
30	>CL	2671	20%	2788	20%
	<CL	10450	77%	10322	76%
	at CL*	529	4%	540	4%
50	>CL	3272	25%	3396	26%
	<CL	9384	71%	9266	70%
	at CL*	529	4%	523	4%
50/20	>CL	3231	24%	3346	25%
	<CL	9609	72%	9492	71%
	at CL*	505	4%	507	4%
Deposition estimates for 2001-03					
20	>CL	3012	22%	3139	23%
	<CL	10271	75%	10156	74%
	at CL*	489	4%	477	3%
30	>CL	3413	25%	3520	26%
	<CL	9784	72%	9683	71%
	at CL*	453	3%	447	3%
50	>CL	3865	29%	3972	30%
	<CL	8905	68%	8807	67%
	at CL*	415	3%	406	3%
50/20	>CL	3834	29%	3931	29%
	<CL	9124	68%	9030	68%
	at CL*	387	3%	384	3%

An exceedance (>CL) is where deposition for the modeled waterbodies is above the CL and error of 3.125 meq/m^2-yr.

* "at CL" indicates that estimated deposition is within 3.125 meq/m^2-yr of the waterbody CL

Zero and negative CLs were excluded from this summary.

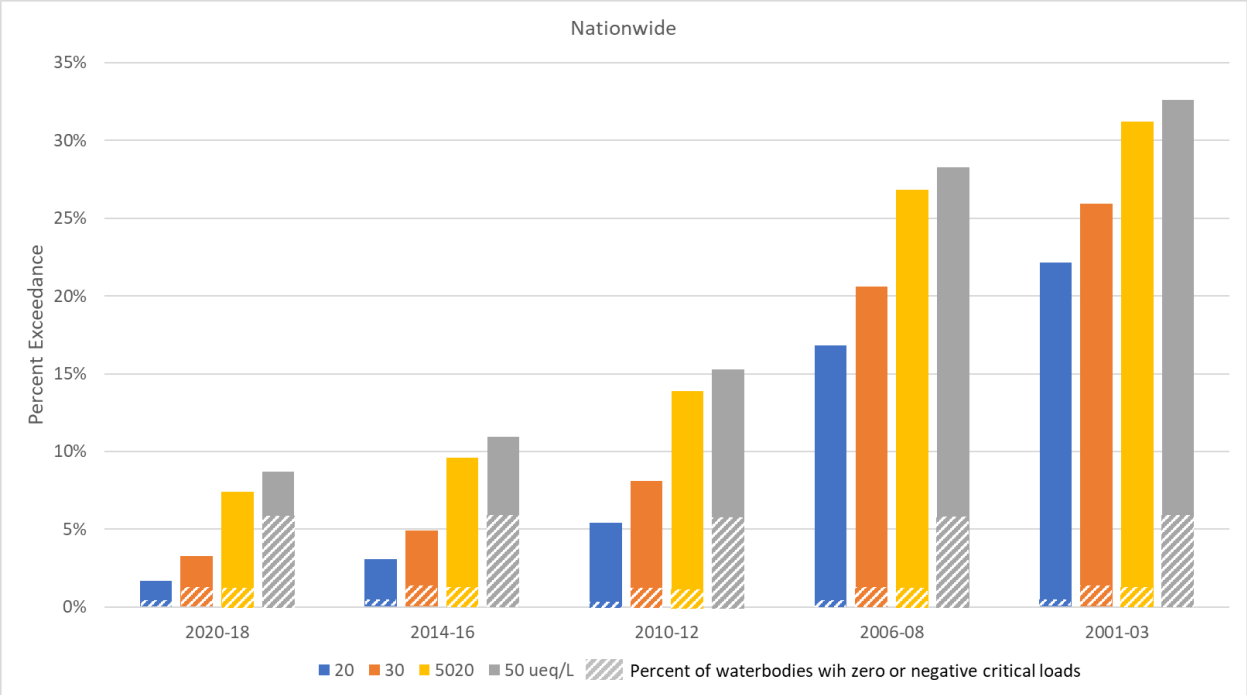

Figure 5A-9. Percent CL exceedances by ANC thresholds and deposition periods.

Figures 5A-10 to 5A-29 show locations of estimated CL exceedances across the CONUS for S only and for ANC thresholds of 20, 30, 50, and 50/20 μeq/L for positive CLs only. Figure 5A-30 highlights the locations of waterbodies that have calculated negative CLs (grey dots). These are waterbodies that are highly sensitive to acidification and likely naturally acidic as indicated by their zero or negative CL. These waterbodies exceed the calculated CL at any deposition amount. For these reasons, these sites have been removed from the assessment. Exceedance maps for S and/or N combined are not included here because they show the same pattern of exceedances as for S only and because exceedance rates are only slightly higher for combined N and/or S deposition. Most exceedances occur in New England, the Adirondacks, the Appalachian Mountain range (New England to Georgia), the upper Midwest, Florida, and the Sierra Nevada mountains in California. Waterbodies in Florida and other coastal plain ecoregions that exceed the CL are likely not related to deposition of S, but instead are related to high levels of natural acidity in these drainage waters. These drainage waters tend to be naturally high in dissolved organic carbon, causing these systems to be acidic (2008 ISA, section 3.2.4.2).

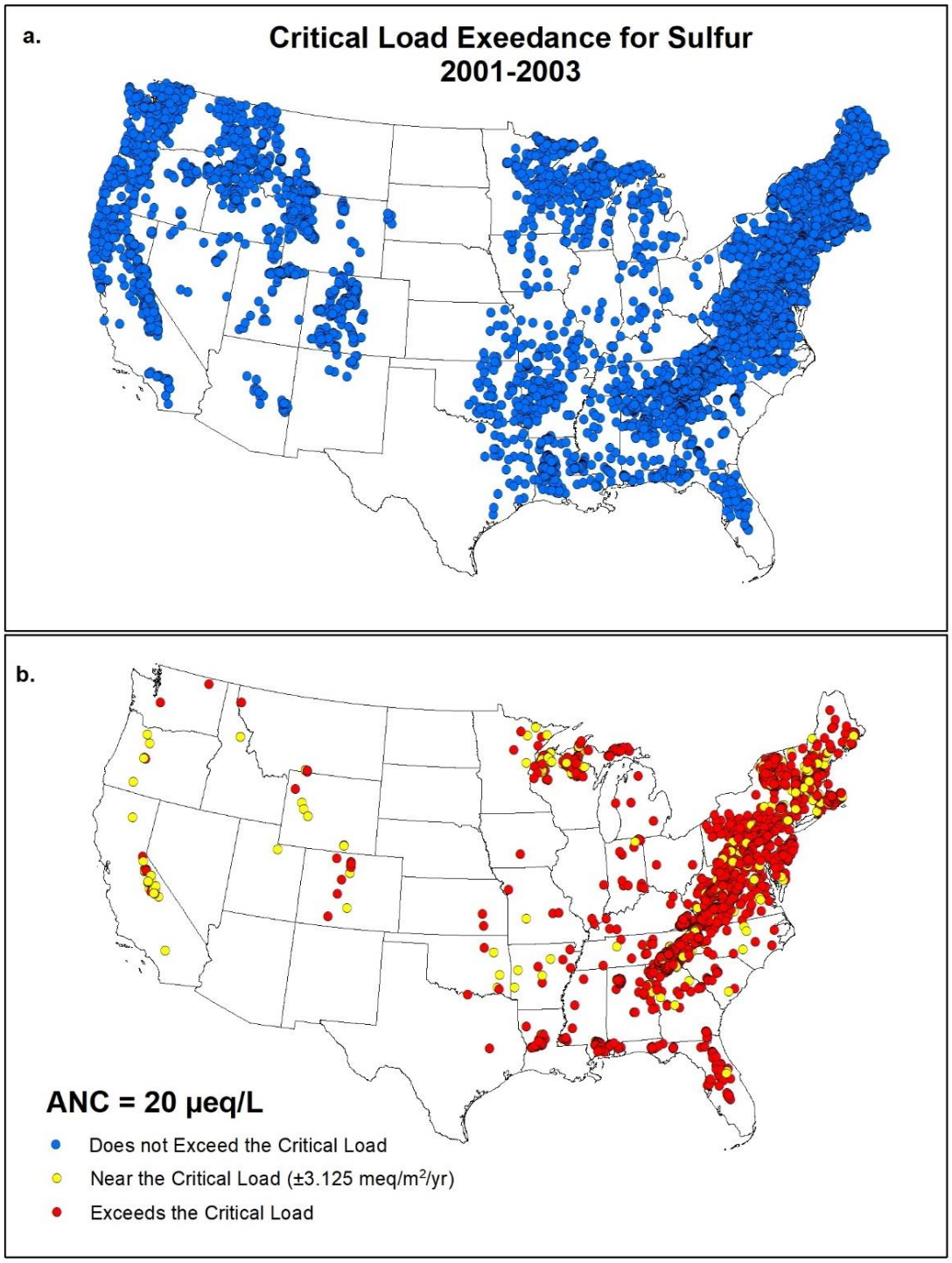

Figure 5A-10. Critical load exceedance (Ex) for S only total deposition from 2001-03 for an ANC threshold of 20 µeq/L: a) waterbodies with sulfur deposition below the CL and uncertainty (Ex < -3.125 meq/m^2-yr), and b) waterbodies with sulfur deposition above or near the CL.

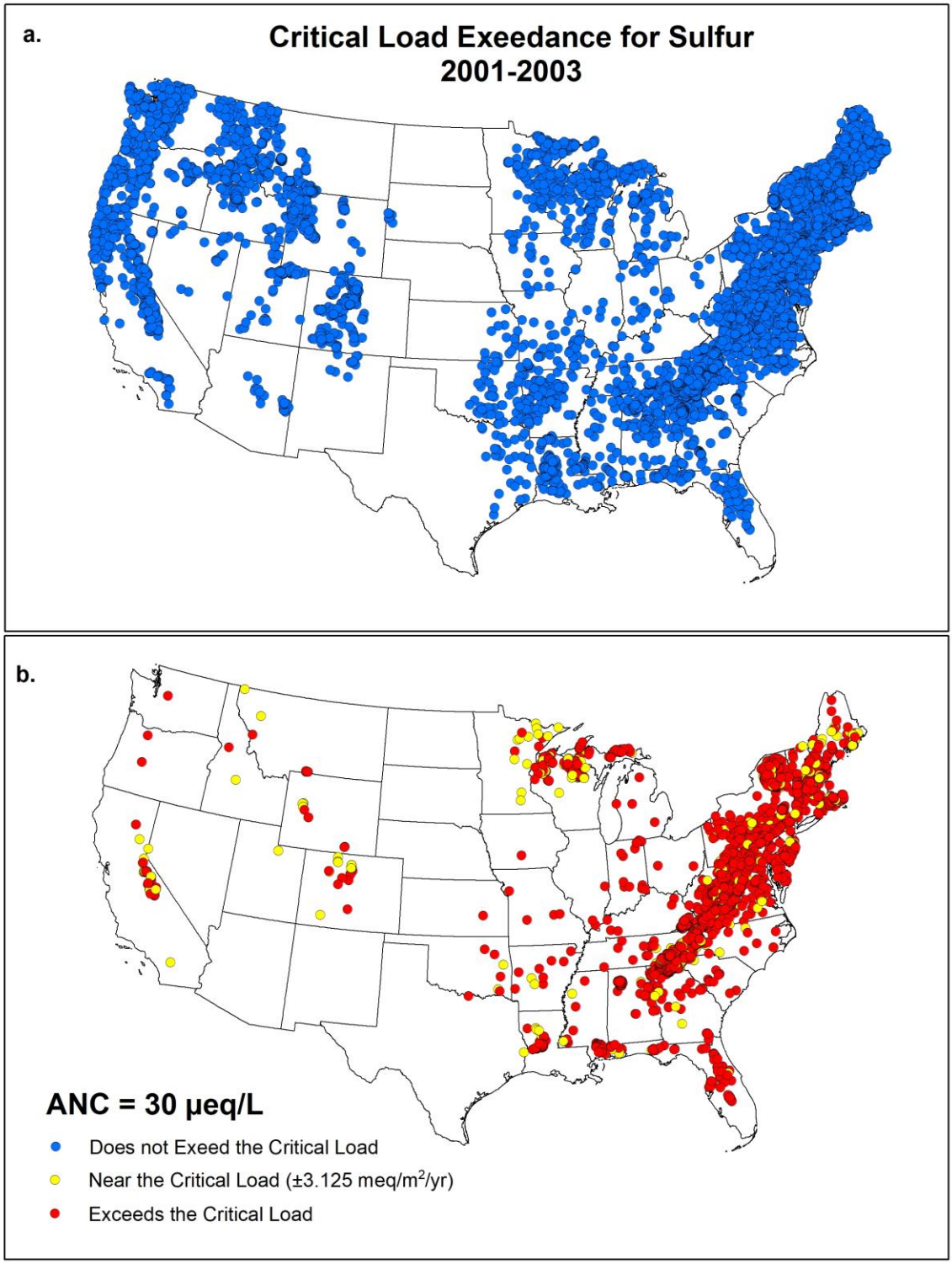

Figure 5A-11. Critical load exceedance (Ex) for S only total deposition from 2001-03 for an ANC threshold of 30 µeq/L: a) waterbodies with sulfur deposition below the CL and uncertainty (Ex < -3.125 meq/m^2-yr), and b) waterbodies with sulfur deposition above or near the CL.

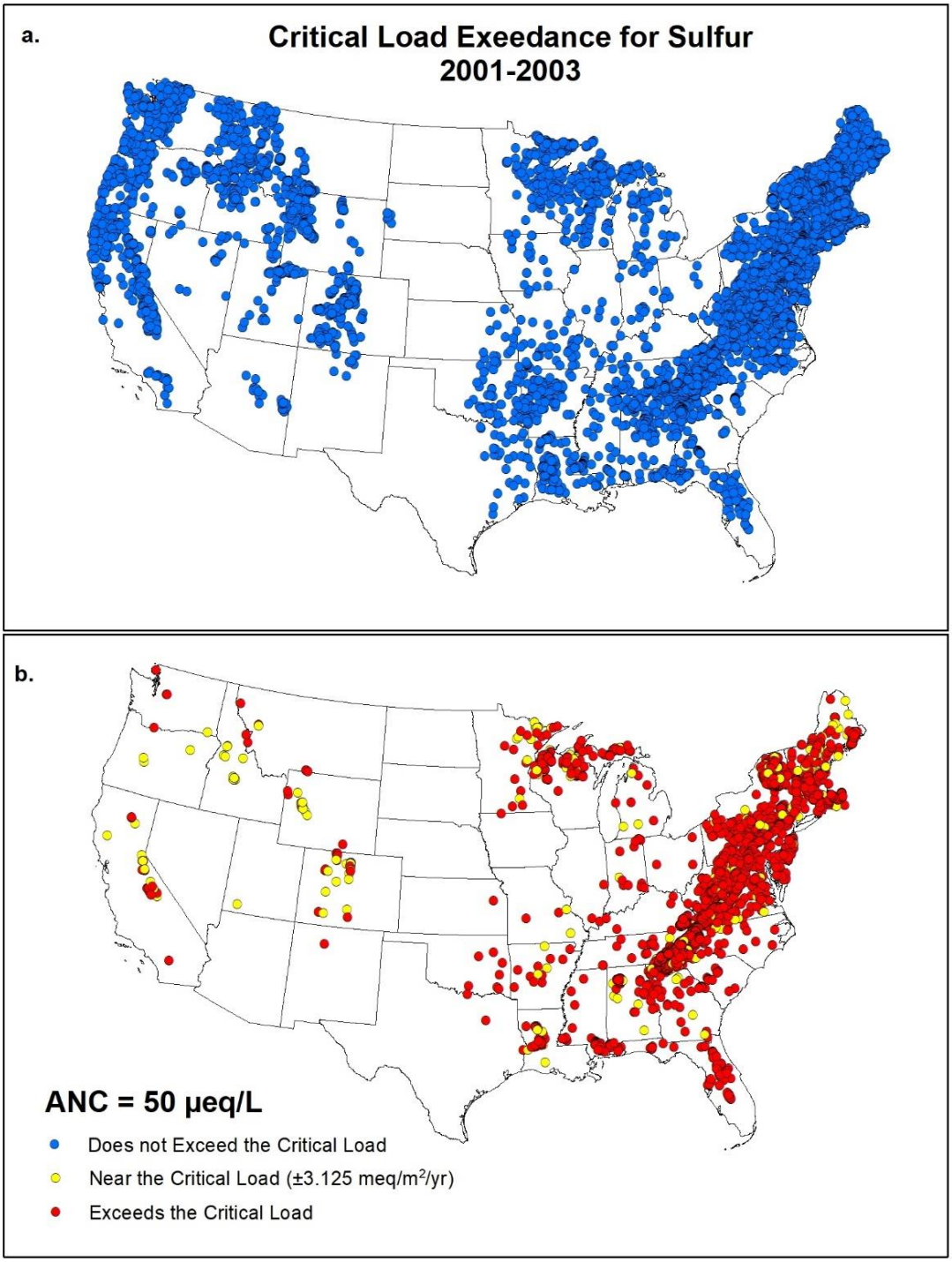

Figure 5A-12. Critical load exceedance (Ex) for S only total deposition from 2001-03 for an ANC threshold of 50 µeq/L: a) waterbodies with sulfur deposition below the CL and uncertainty (Ex < -3.125 meq/m^2-yr), and b) waterbodies with sulfur deposition above or near the CL.

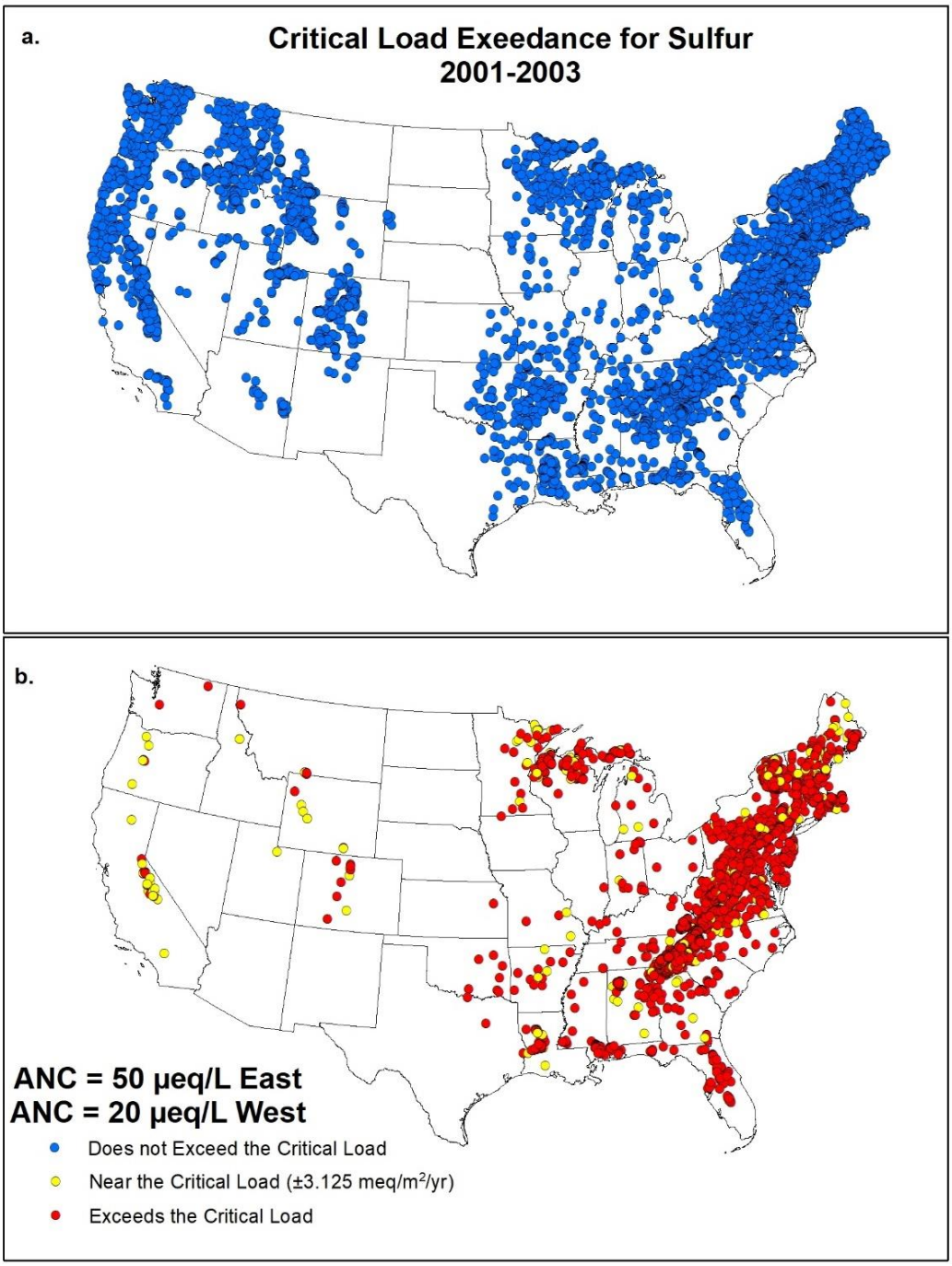

Figure 5A-13. Critical load exceedance (Ex) for S only total deposition from 2001-03 for an ANC threshold of 50 for the eastern and 20 µeq/L for Western CONUS: a) waterbodies with sulfur deposition below the CL and uncertainty (Ex < -3.125 meq/m^2-yr), and b) waterbodies with sulfur deposition above or near the CL.

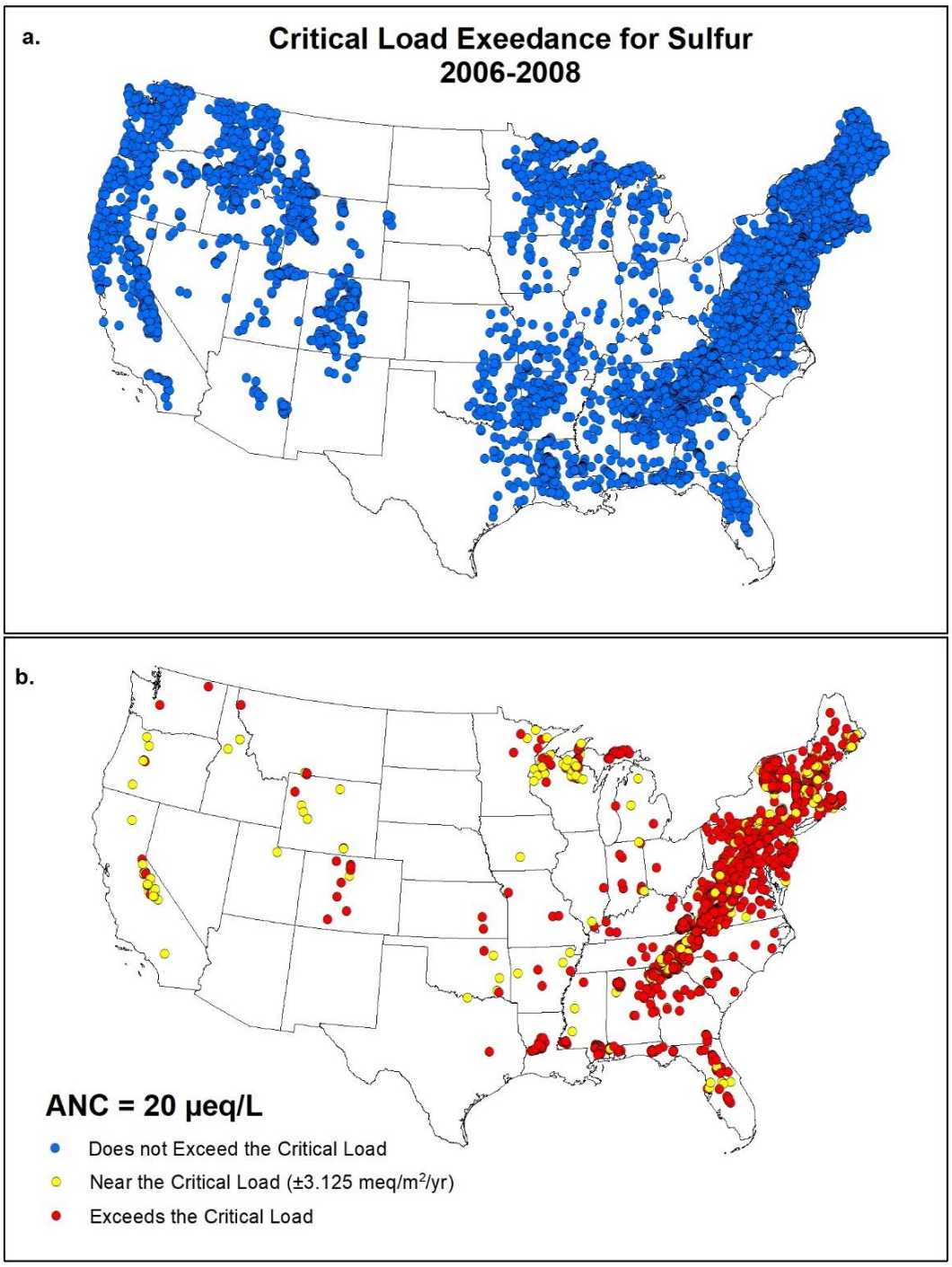

Figure 5A-14. Critical load exceedance (Ex) for S only total deposition from 2006-08 for an ANC threshold of 20 µeq/L: a) waterbodies with sulfur deposition below the CL and uncertainty (Ex < -3.125 meq/m^2-yr), and b) waterbodies with sulfur deposition above or near the CL.

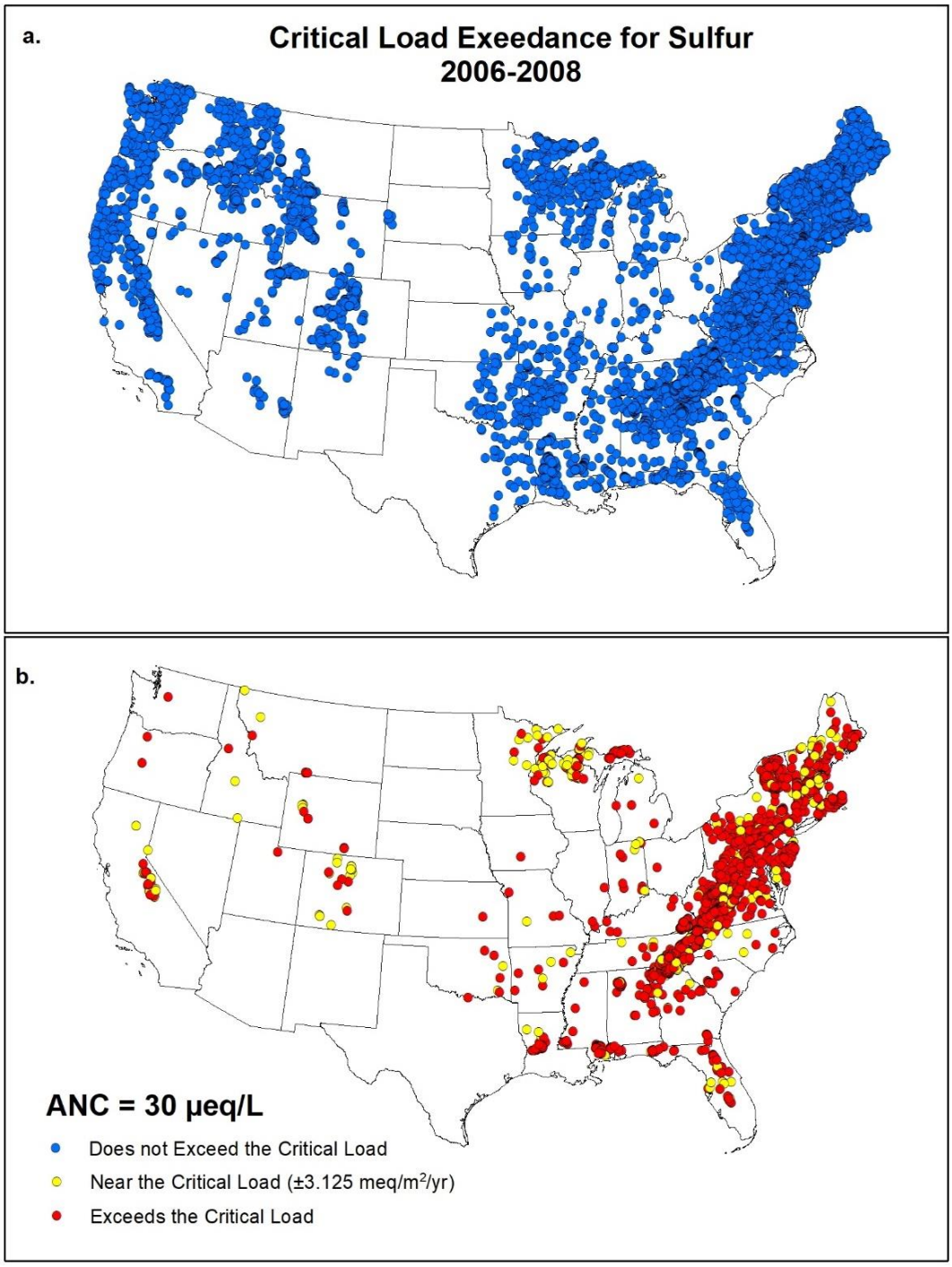

Figure 5A-15. Critical load exceedance (Ex) for S only total deposition from 2006-08 for an ANC threshold of 30 µeq/L: a) waterbodies with sulfur deposition below the CL and uncertainty (Ex < -3.125 meq/m^2-yr), and b) waterbodies with sulfur deposition above or near the CL.

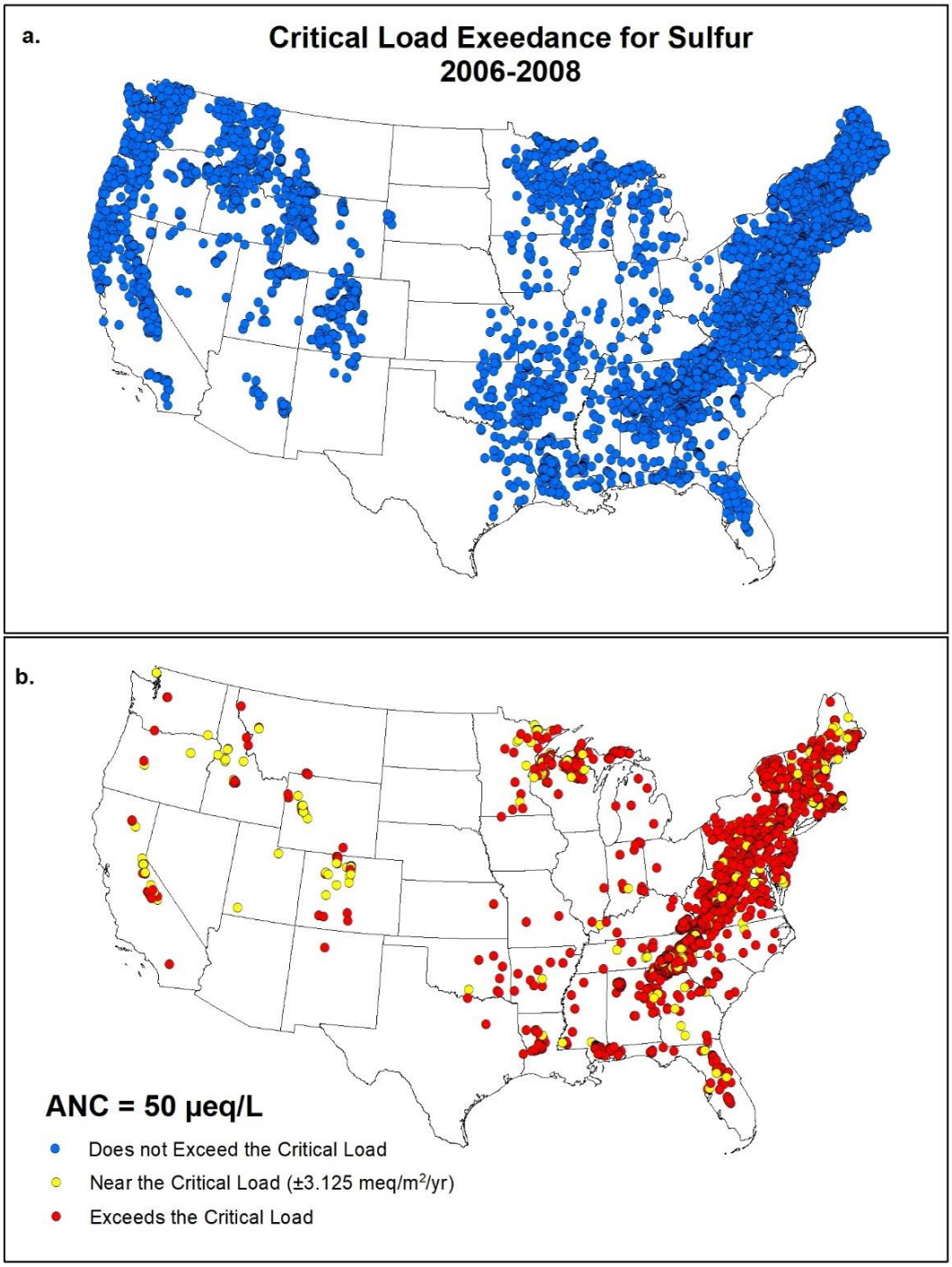

Figure 5A-16. Critical load exceedance (Ex) for S only total deposition from 2006-08 for an ANC threshold of 50 µeq/L: a) waterbodies with sulfur deposition below the CL and uncertainty (Ex < -3.125 meq/m^2-yr), and b) waterbodies with sulfur deposition above or near the CL.

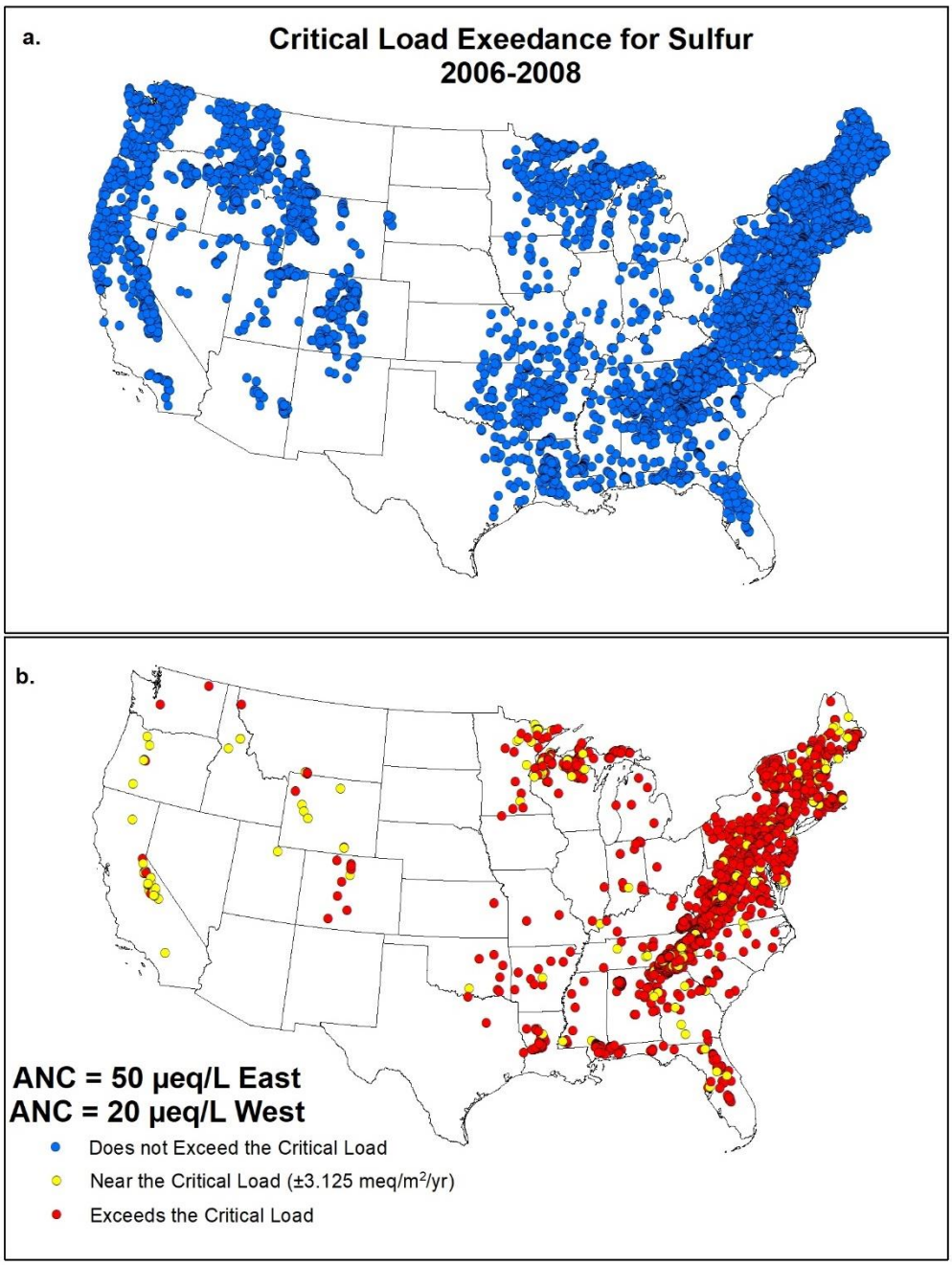

Figure 5A-17. Critical load exceedance (Ex) for S only total deposition from 2006-08 for an ANC threshold of 50 for the eastern and 20 µeq/L for Western CONUS: a) waterbodies with sulfur deposition below the CL and uncertainty (Ex < -3.125 meq/m^2-yr), and b) waterbodies with sulfur deposition above or near the CL.

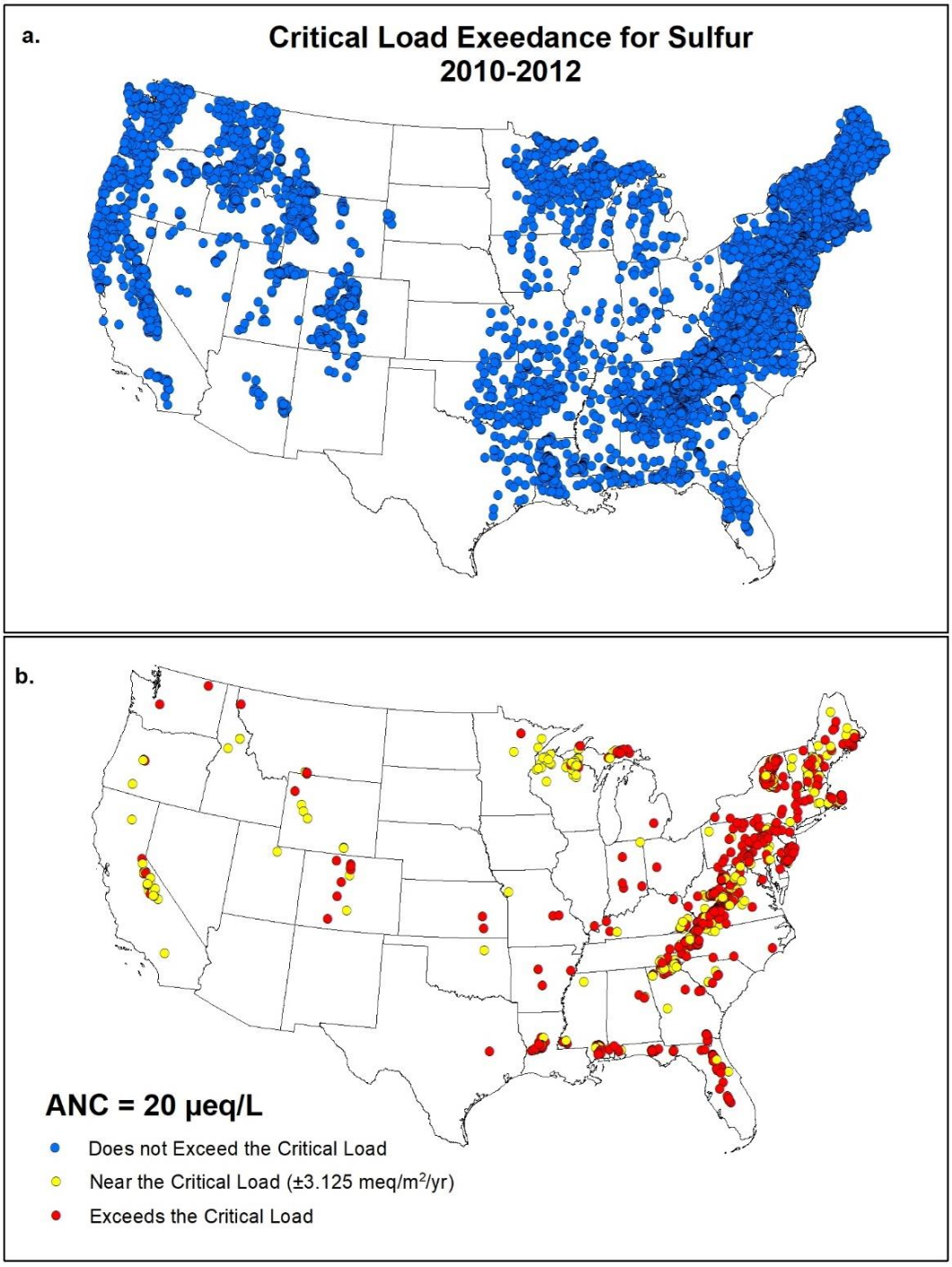

Figure 5A-18. Critical load exceedance (Ex) for S only total deposition from 2010-12 for an ANC threshold of 20 µeq/L: a) waterbodies with sulfur deposition below the CL and uncertainty (Ex < -3.125 meq/m^2-yr), and b) waterbodies with sulfur deposition above or near the CL.

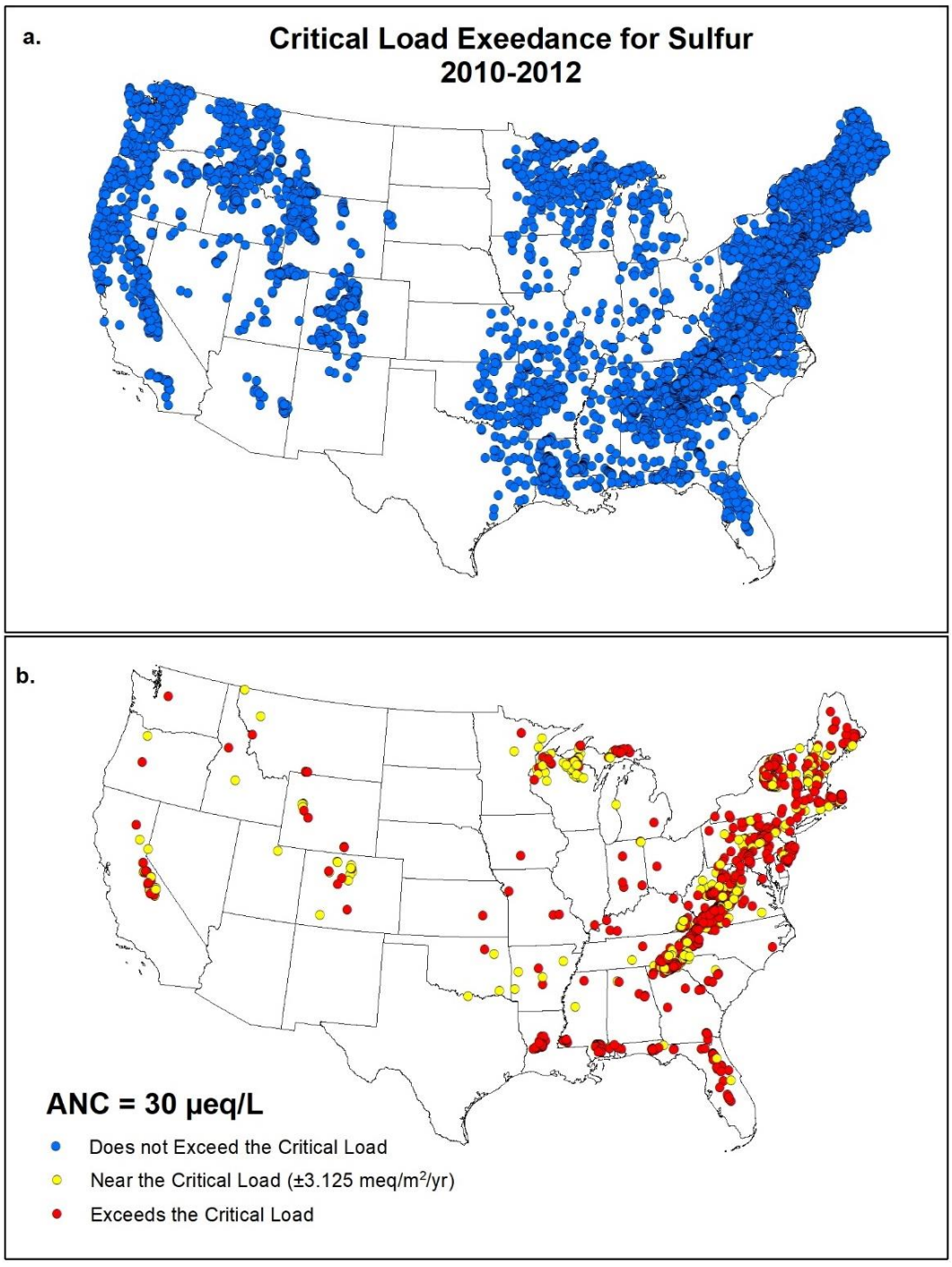

Figure 5A-19. Critical load exceedance (Ex) for S only total deposition from 2010-12 for an ANC threshold of 30 µeq/L: a) waterbodies with sulfur deposition below the CL and uncertainty (Ex < -3.125 meq/m^2-yr), and b) waterbodies with sulfur deposition above or near the CL.

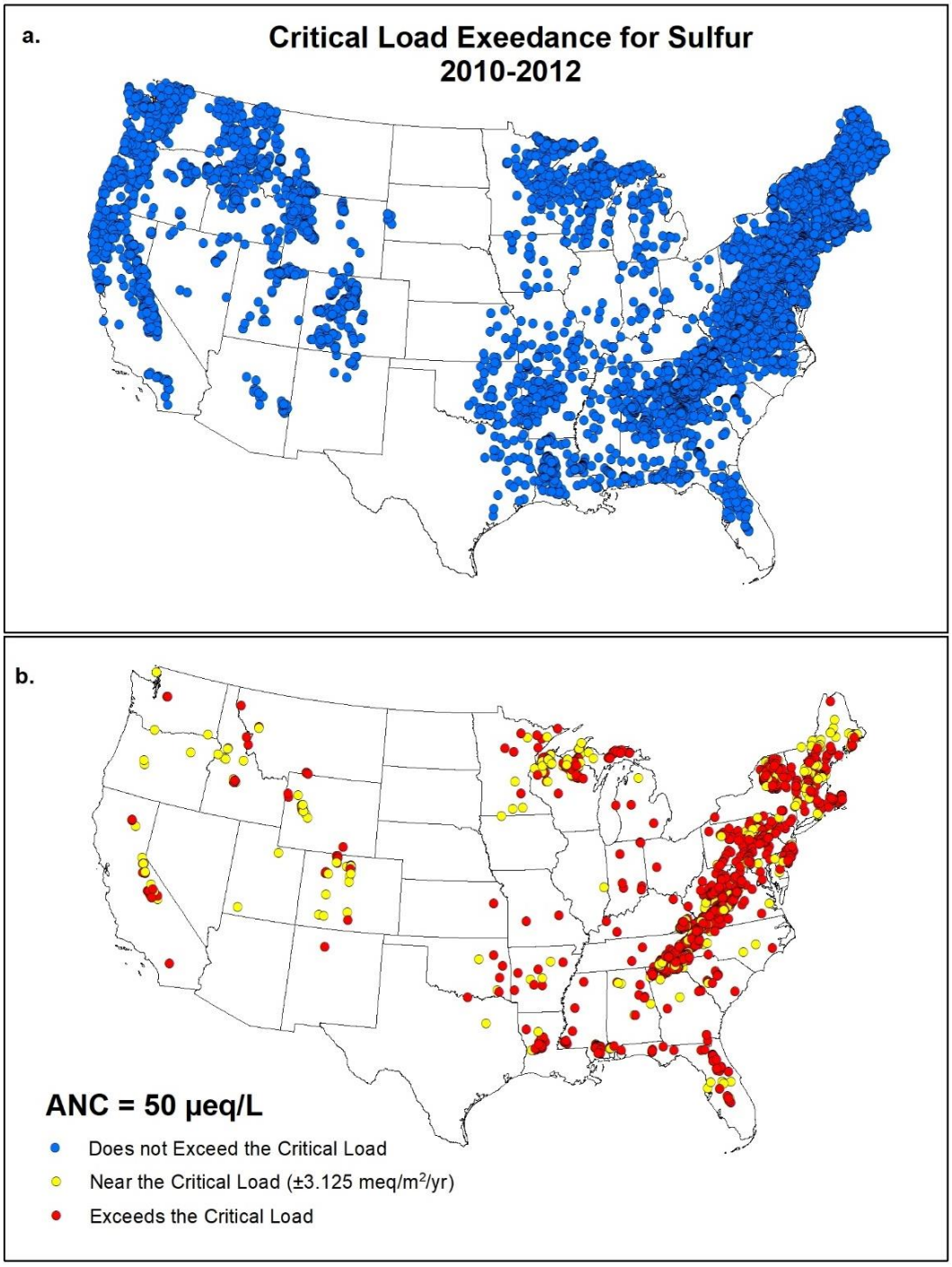

Figure 5A-20. Critical load exceedance (Ex) for S only total deposition from 2010-12 for an ANC threshold of 50 µeq/L: a) waterbodies with sulfur deposition below the CL and uncertainty (Ex < -3.125 meq/m^2-yr), and b) waterbodies with sulfur deposition above or near the CL.

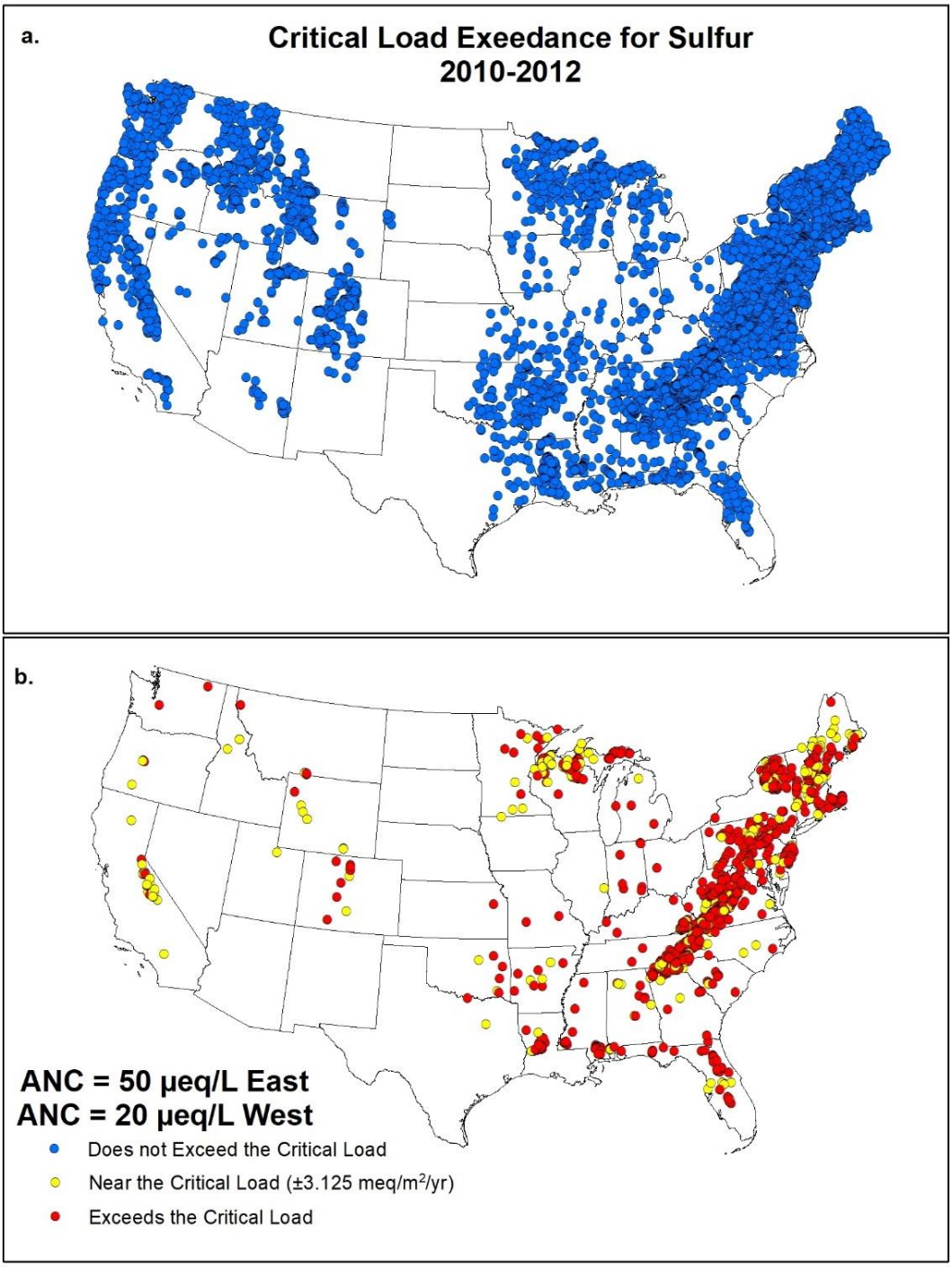

Figure 5A-21. Critical load exceedance (Ex) for S only total deposition from 2010-12 for an ANC threshold of 50 for the eastern and 20 µeq/L for Western CONUS: a) waterbodies with sulfur deposition below the CL and uncertainty (Ex < -3.125 meq/m^2-yr), and b) waterbodies with sulfur deposition above or near the CL.

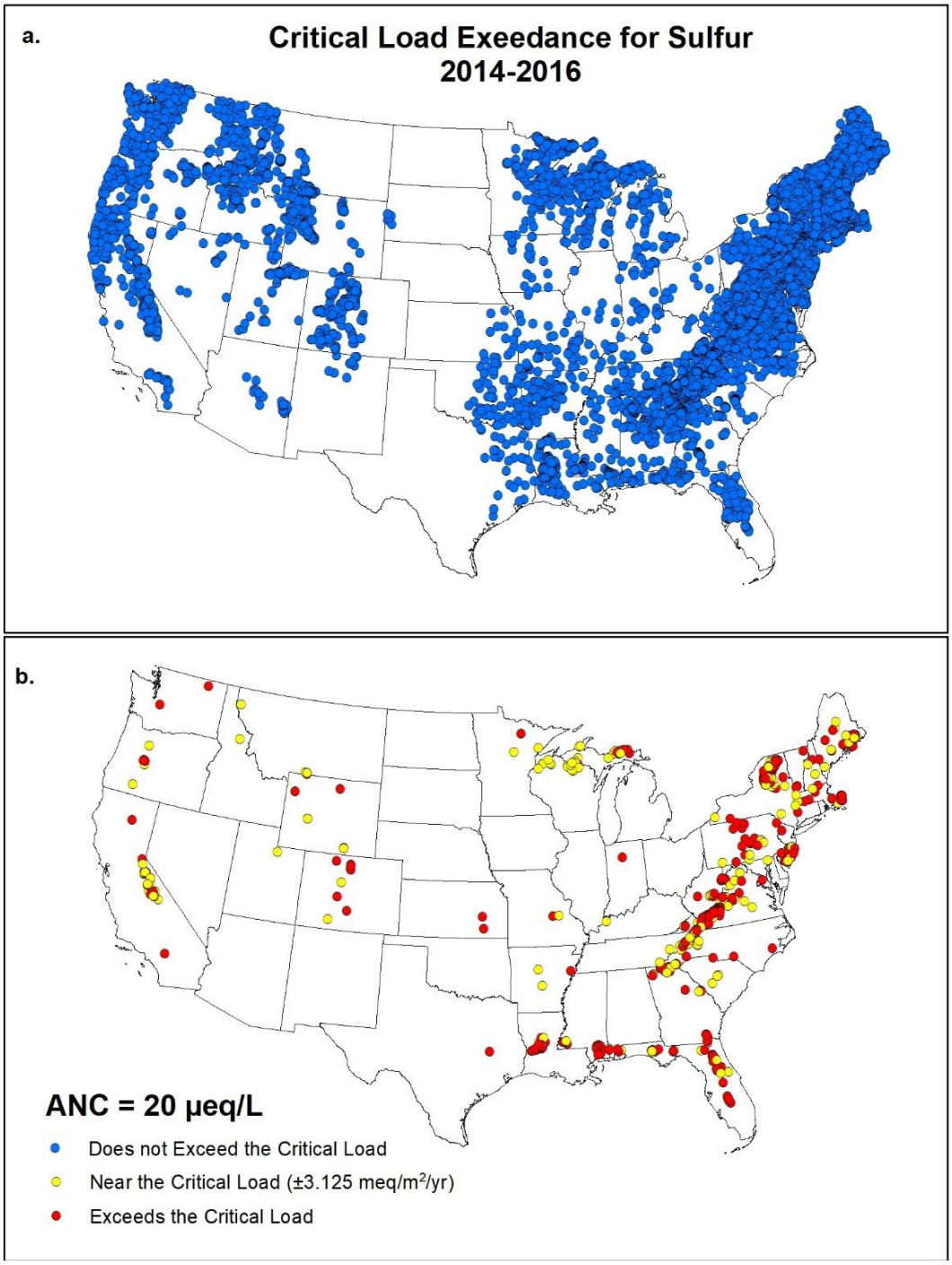

Figure 5A-22. Critical load exceedance (Ex) for S only total deposition from 2014-16 for an ANC threshold of 20 µeq/L: a) waterbodies with sulfur deposition below the CL and uncertainty (Ex < -3.125 meq/m^2-yr), and b) waterbodies with sulfur deposition above or near the CL.

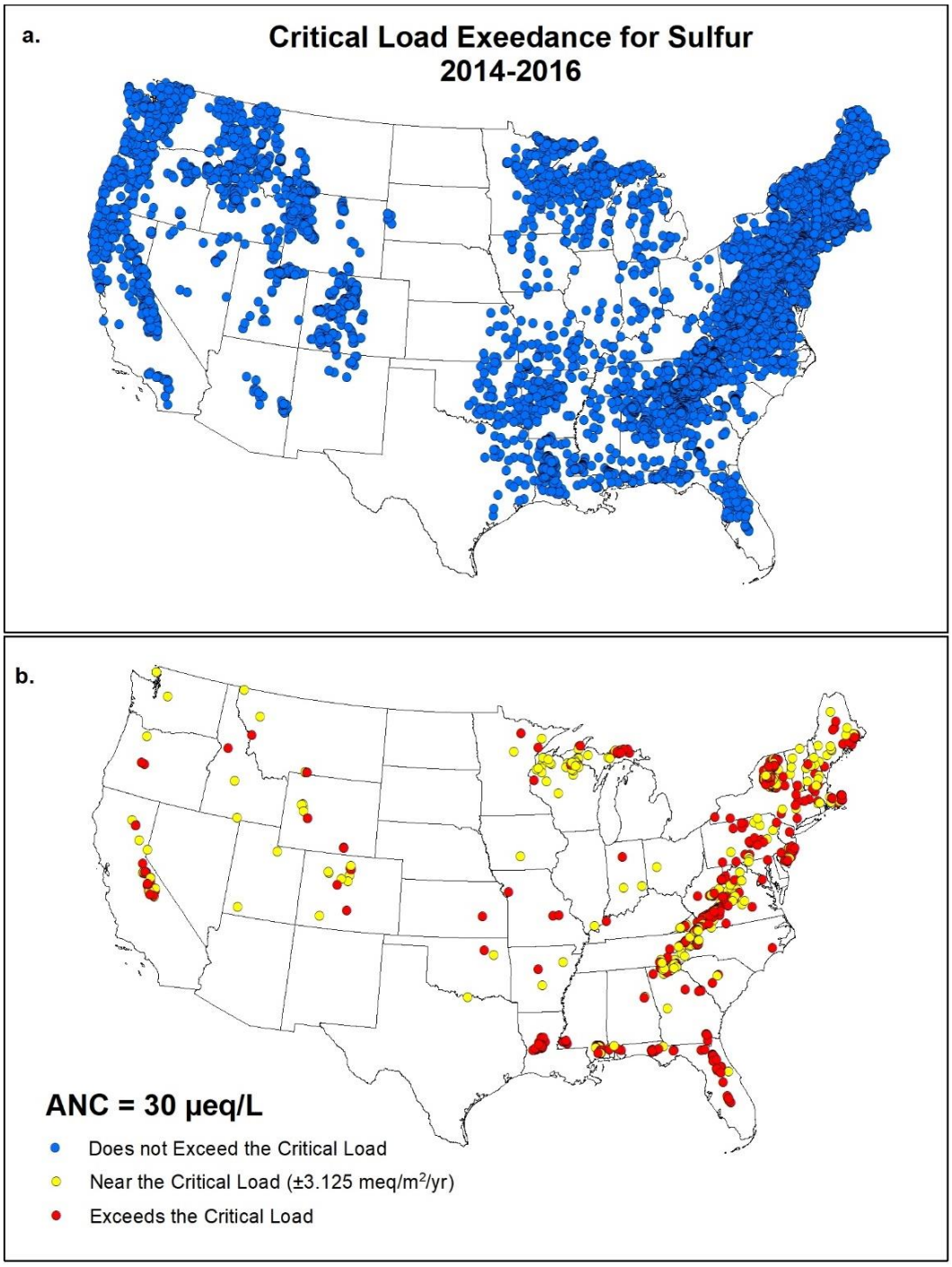

Figure 5A-23. Critical load exceedance (Ex) for S only total deposition from 2014-16 for an ANC threshold of 30 µeq/L: a) waterbodies with sulfur deposition below the CL and uncertainty (Ex < -3.125 meq/m^2-yr), and b) waterbodies with sulfur deposition above or near the CL.

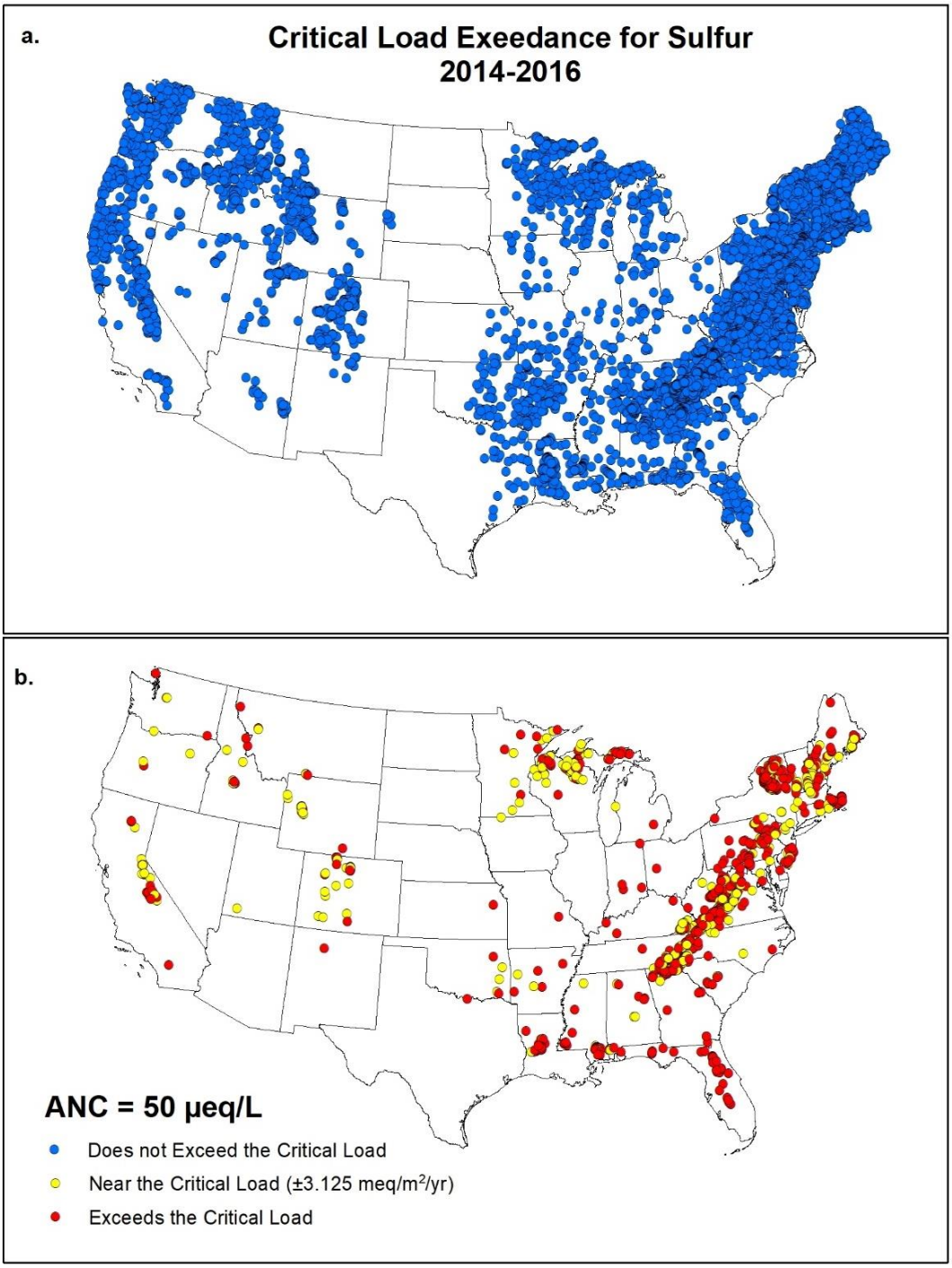

Figure 5A-24. Critical load exceedance (Ex) for S only total deposition from 2014-16 for an ANC threshold of 50 µeq/L: a) waterbodies with sulfur deposition below the CL and uncertainty (Ex < -3.125 meq/m^2-yr), and b) waterbodies with sulfur deposition above or near the CL.

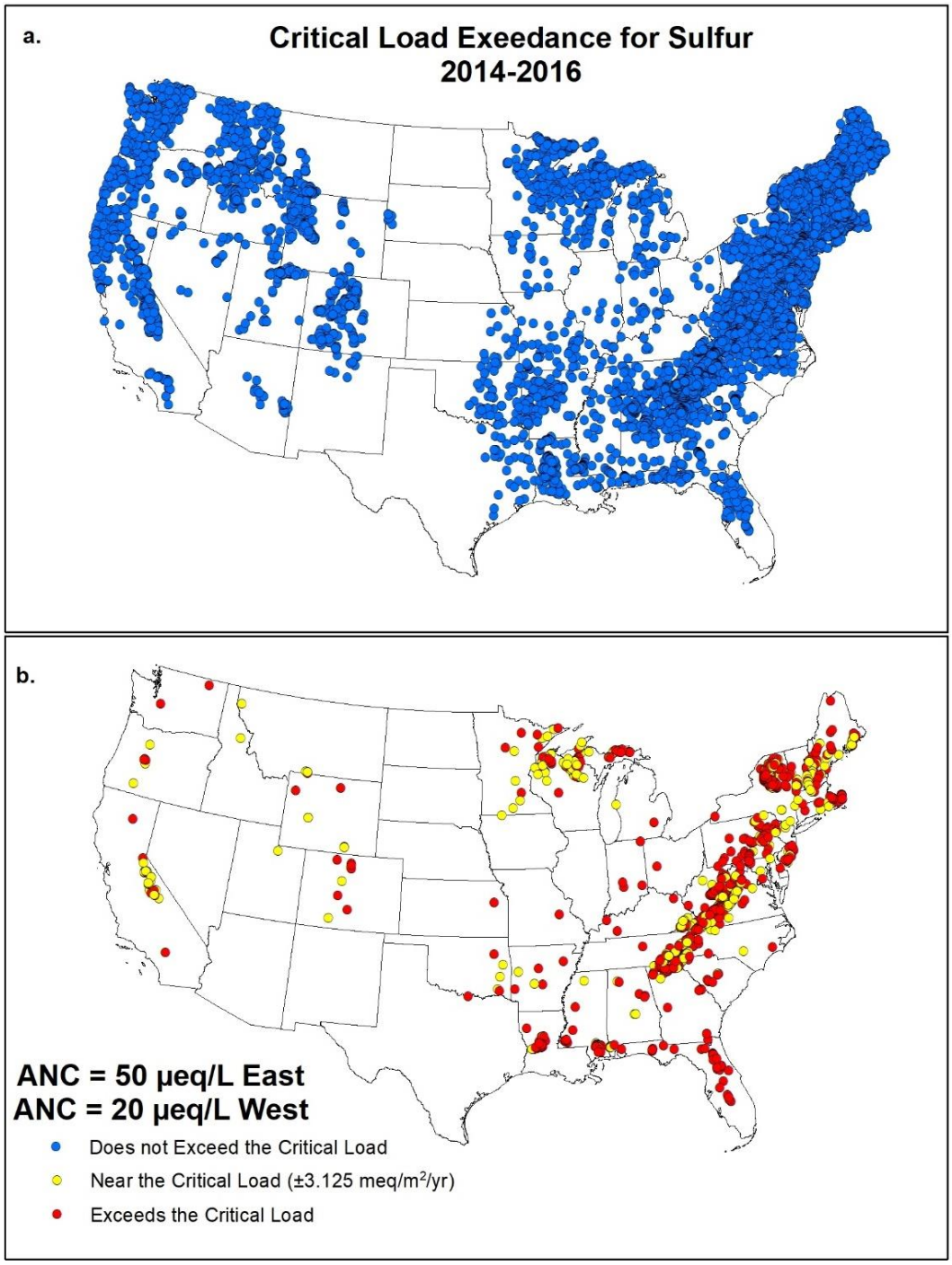

Figure 5A-25. Critical load exceedance (Ex) for S only total deposition from 2014-16 for an ANC threshold of 50 for the eastern and 20 µeq/L for Western CONUS: a) waterbodies with sulfur deposition below the CL and uncertainty (Ex < -3.125 meq/m^2-yr), and b) waterbodies with sulfur deposition above or near the CL.

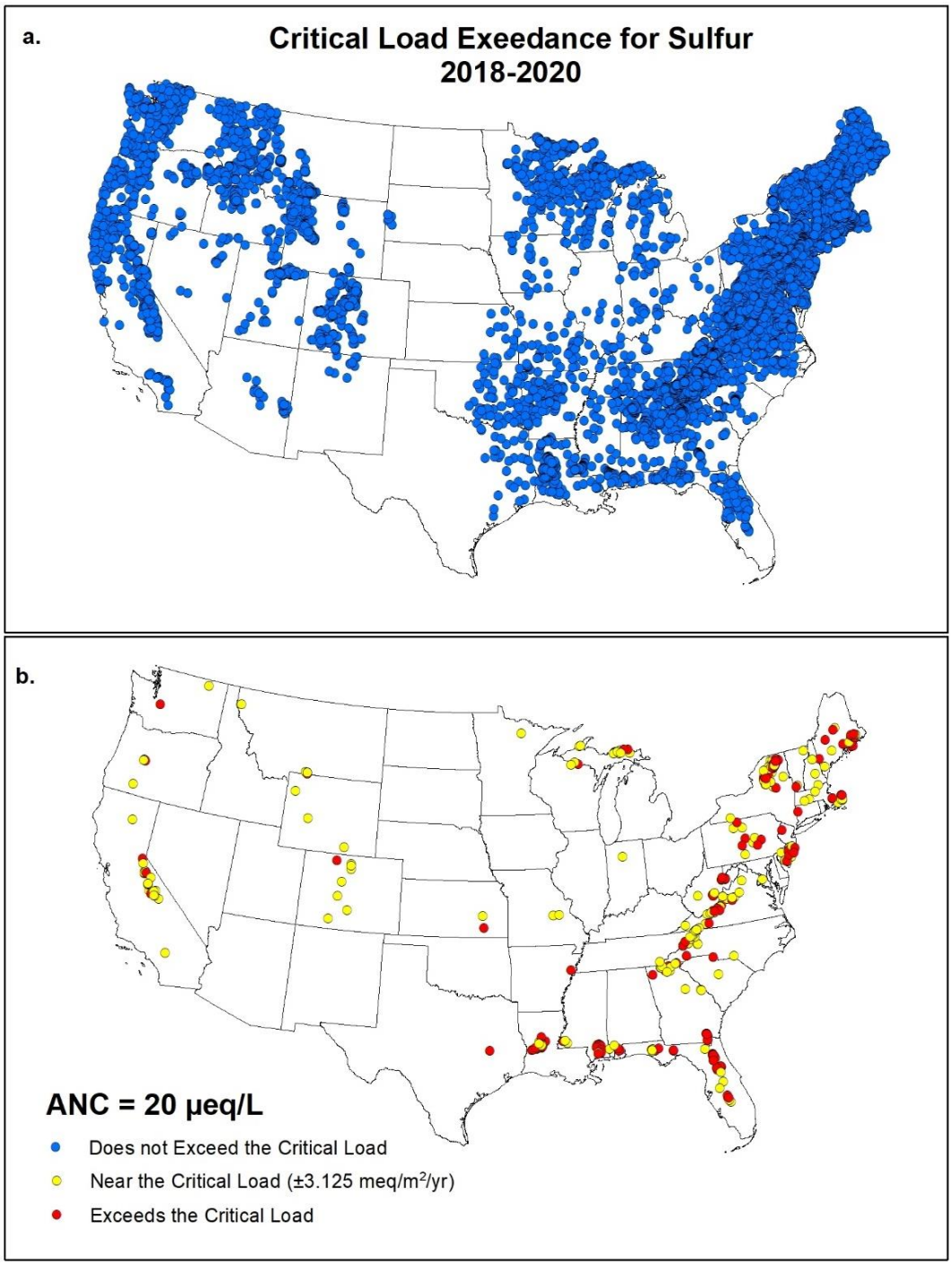

Figure 5A-26. Critical load exceedance (Ex) for S only total deposition from 2018-20 for an ANC threshold of 20 µeq/L: a) waterbodies with sulfur deposition below the CL and uncertainty (Ex < -3.125 meq/m^2-yr), and b) waterbodies with sulfur deposition above or near the CL.

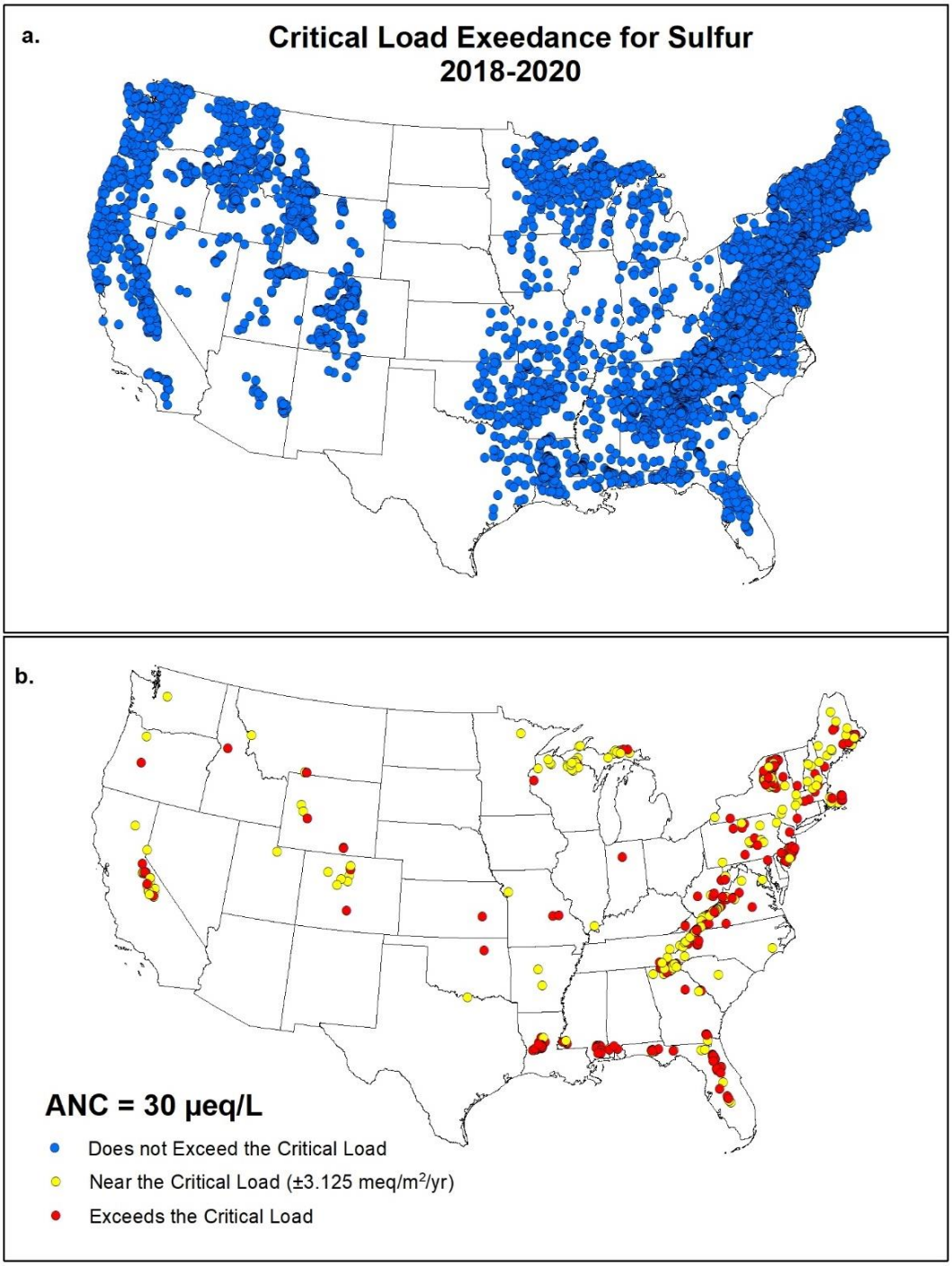

Figure 5A-27. Critical load exceedance (Ex) for S only total deposition from 2018-20 for an ANC threshold of 30 µeq/L: a) waterbodies with sulfur deposition below the CL and uncertainty (Ex < -3.125 meq/m^2-yr), and b) waterbodies with sulfur deposition above or near the CL.

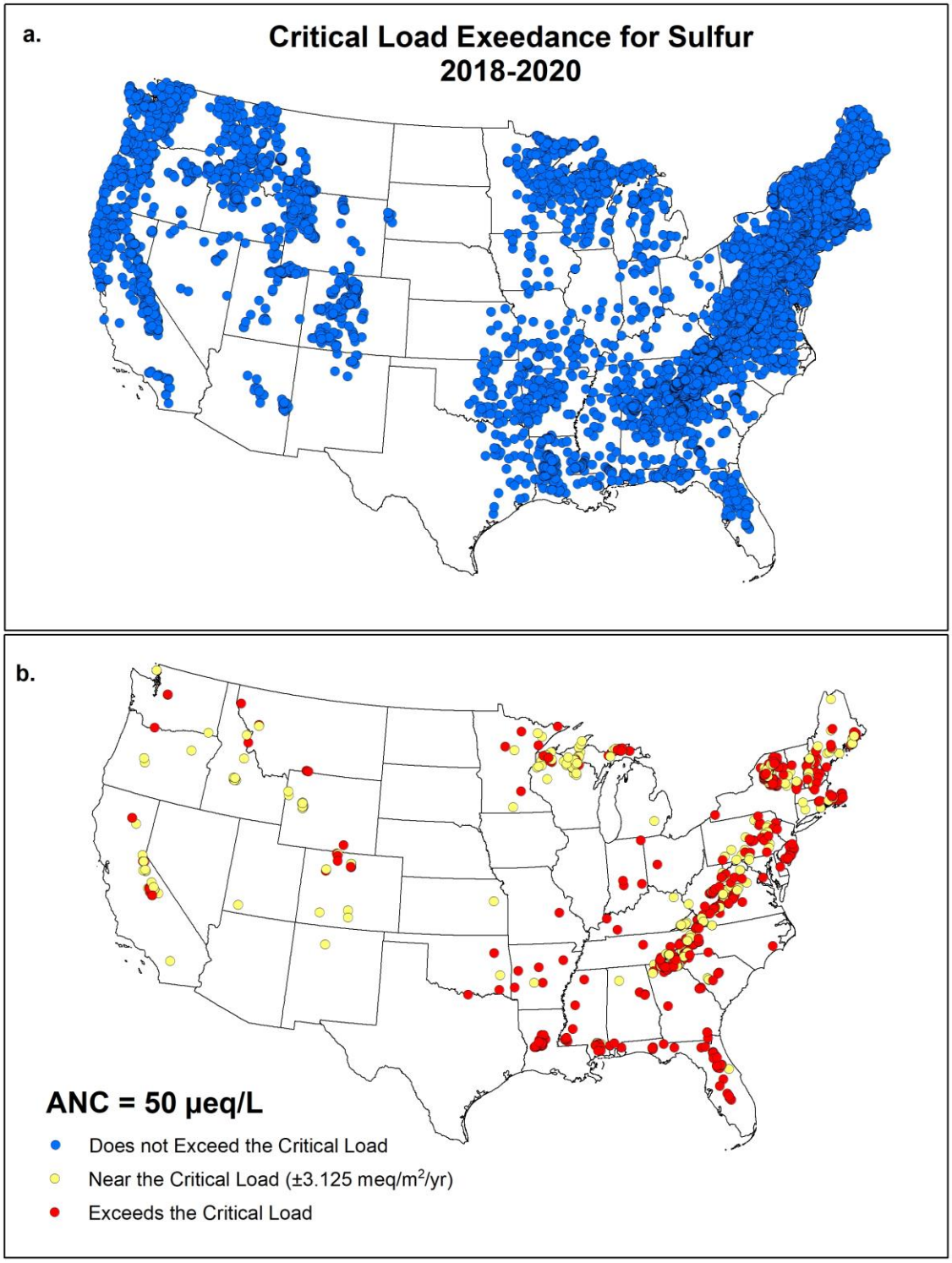

Figure 5A-28. Critical load exceedance (Ex) for S only total deposition from 2018-20 for an ANC threshold of 50 µeq/L: a) waterbodies with sulfur deposition below the CL and uncertainty (Ex < -3.125 meq/m^2-yr), and b) waterbodies with sulfur deposition above or near the CL.

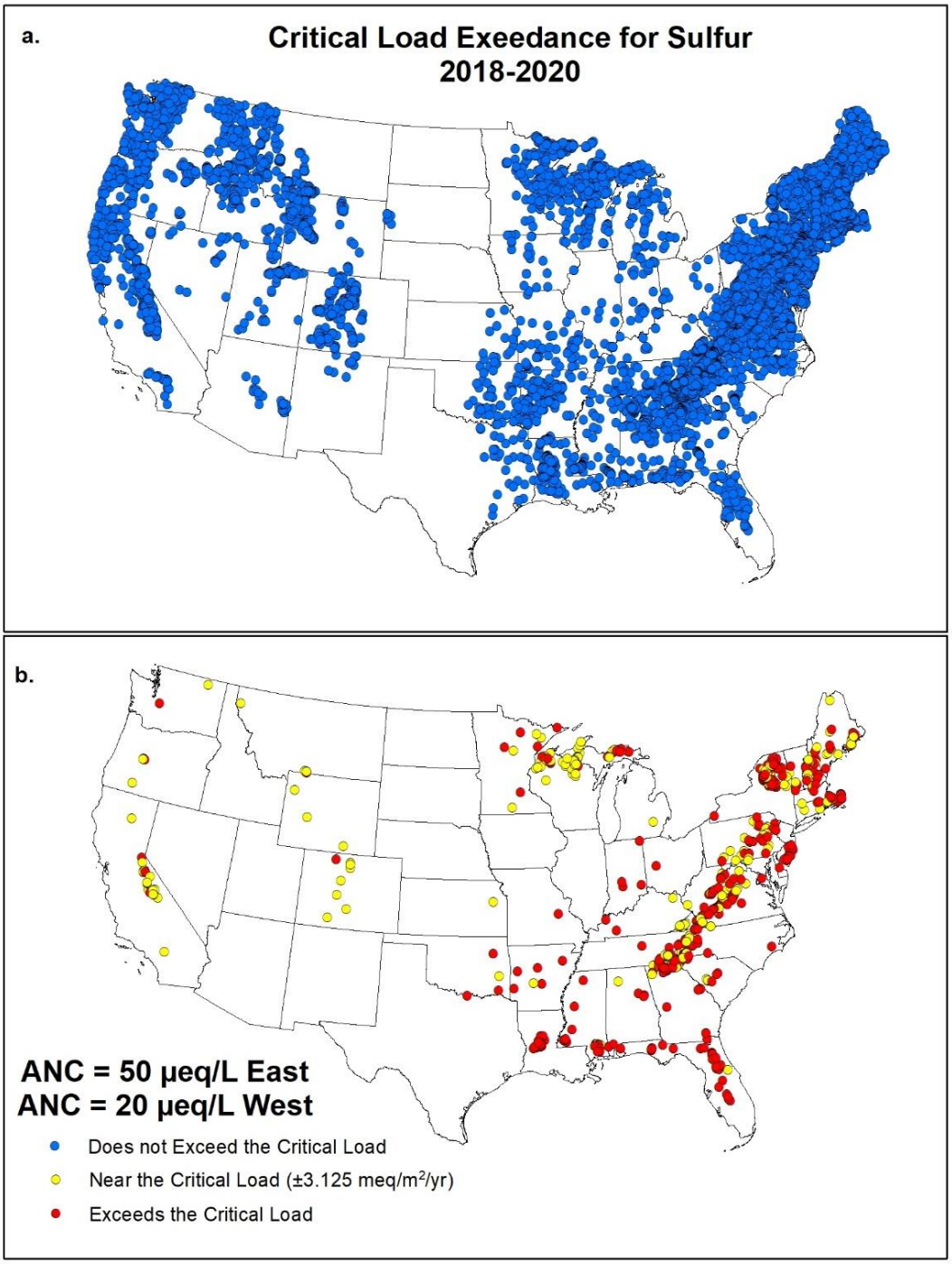

Figure 5A-29. Critical load exceedance (Ex) for S only total deposition from 2018-20 for an ANC threshold of 50 for the eastern and 20 µeq/L for Western CONUS: a) waterbodies with sulfur deposition below the CL and uncertainty (Ex < -3.125 meq/m^2-yr), and b) waterbodies with sulfur deposition above or near the CL.

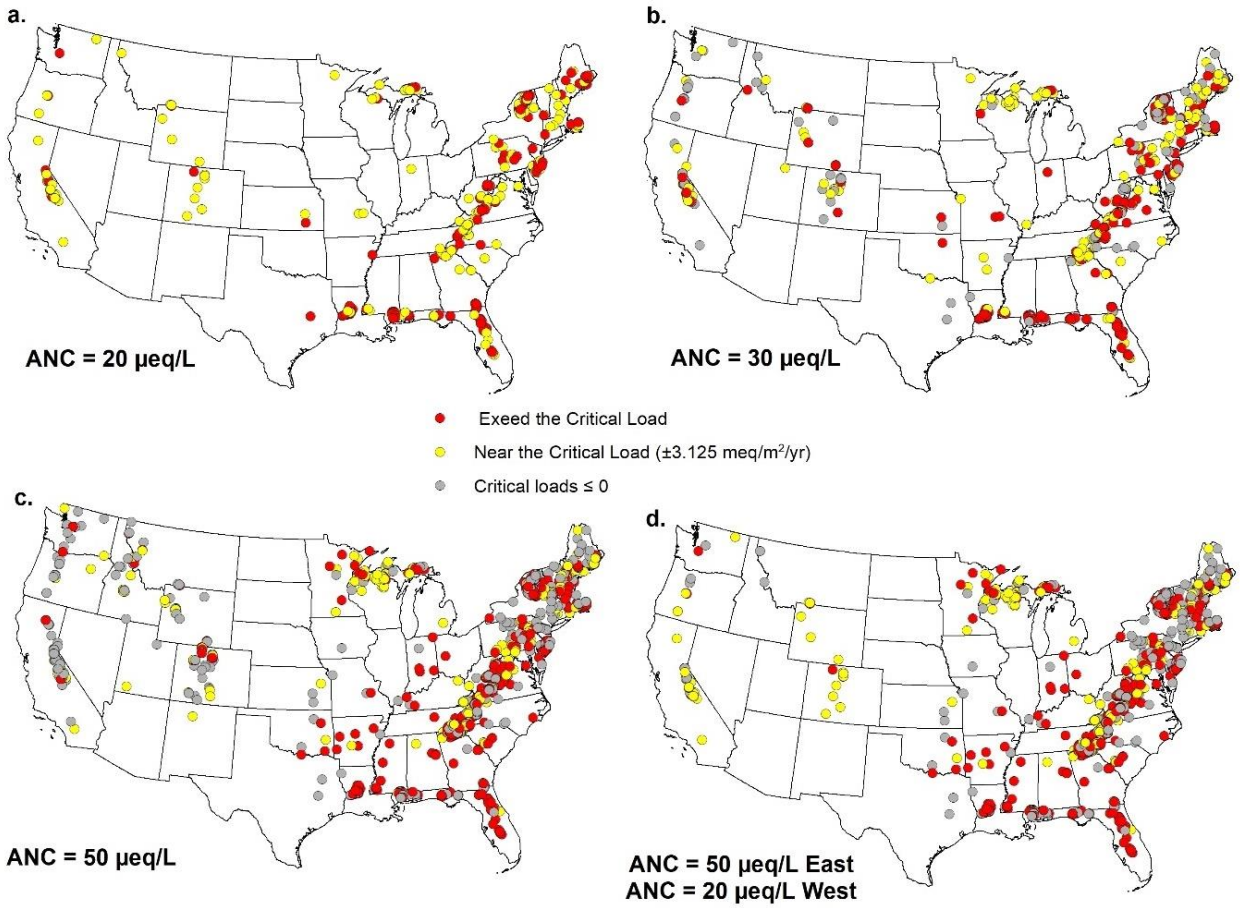

Figure 5A-30. Critical load exceedance for S only deposition from 2018-20 for four ANC thresholds: a. 20, b. 30, c. 50, d. 50/20 µeq/L.

5A.2.2 Ecoregion Analyses

There are 84 level III ecoregions across the CONUS. As seen in Tables 5A-10 and 5A-11 below, S deposition has declined in all of them since the 2001-03 time period.

Table 5A-10. Summary of median deposition estimates during five time periods for the 84 ecoregions in the CONUS. Deposition based on TDEP; median determined by GIS zonal statistic.

Total Sulfur Deposition	Number of ecoregions with median deposition within specified range				
kg S/ha-yr	2001-03	2006-08	2010-12	2014-16	2018-20
>10	16	11	0	0	0
7-10	10	10	5	0	0
5-7	11	14	10	0	0
2-5	13	14	31	45	33
<2	34	35	38	39	51

Table 5A-11. Median sulfur deposition for the 84 ecoregions in the CONUS determined by GIS zonal statistic based on TDEP estimates.

	Ecoregion III		Median Total Sulfur Deposition (kg S/ha-yr)				
Code	Name	E/W	2001-03	2006-08	2010-12	2014-16	2018-20
10.1.2	Columbia Plateau	W	0.46	0.42	0.43	0.50	0.29
10.1.3	Northern Basin and Range	W	0.34	0.37	0.53	0.48	0.29
10.1.4	Wyoming Basin	W	0.64	0.67	0.52	0.56	0.42
10.1.5	Central Basin and Range	W	0.49	0.45	0.47	0.52	0.34
10.1.6	Colorado Plateaus	W	0.74	0.74	0.56	0.61	0.32
10.1.7	Arizona/New Mexico Plateau	W	0.82	0.80	0.64	0.57	0.33
10.1.8	Snake River Plain	W	0.48	0.66	0.59	0.59	0.38
10.2.1	Mojave Basin and Range	W	0.58	0.41	0.42	0.41	0.30
10.2.10	Chihuahuan Deserts (also 10.2.4)	W	1.21	1.12	1.11	1.22	0.86
10.2.2	Sonoran Basin and Range	W	0.54	0.46	0.45	0.44	0.29
11.1.1	Southern and Central California Chaparral and Oak Woodlands	W	1.12	0.95	0.94	0.84	0.74
11.1.2	Central California Valley	W	1.09	0.92	0.82	0.80	0.66
11.1.3	Southern California Mountains	W	1.23	1.08	1.07	0.98	0.83
12.1.1	Madrean Archipelago	W	1.16	1.14	0.92	0.94	0.49
13.1.1	Arizona/New Mexico Mountains	W	1.43	1.41	1.17	1.03	0.60
15.4.1	Southern Florida Coastal Plain	E	5.96	5.16	4.20	4.34	3.76
5.2.1	Northern Lakes and Forests	E	4.29	3.24	2.44	1.89	1.33
5.2.2	Northern Minnesota Wetlands	E	2.28	2.12	1.45	1.13	0.86
5.3.1	Northern Appalachian and Atlantic Maritime Highlands	E	6.46	5.78	3.01	1.99	1.34
5.3.3	North Central Appalachians	E	18.08	15.05	7.24	4.09	2.40
6.2.10	Middle Rockies	W	1.04	1.14	0.93	0.86	0.71
6.2.11	Klamath Mountains	W	0.90	1.05	1.02	1.07	0.93
6.2.12	Sierra Nevada	W	1.32	1.14	1.24	1.14	0.98
6.2.13	Wasatch and Uinta Mountains	W	1.36	1.38	1.18	1.27	0.77
6.2.14	Southern Rockies	W	1.14	1.18	0.92	0.85	0.54
6.2.15	Idaho Batholith	W	0.90	1.16	1.10	0.93	0.60
6.2.3	Northern Rockies	W	0.90	0.98	0.83	0.79	0.52
6.2.4	Canadian Rockies	W	1.22	1.35	0.97	0.97	0.78
6.2.5	North Cascades	W	1.64	1.55	1.28	1.39	1.09
6.2.7	Cascades	W	1.69	1.66	1.41	1.51	1.24
6.2.8	Eastern Cascades Slopes and Foothills	W	0.44	0.49	0.47	0.55	0.47
6.2.9	Blue Mountains	W	0.46	0.50	0.52	0.61	0.36
7.1.7	Puget Lowland	W	2.13	1.63	1.37	2.11	1.25
7.1.8	Coast Range	W	2.39	2.14	2.00	2.03	1.50
7.1.9	Willamette Valley	W	1.61	1.48	1.43	1.71	1.08
8.1.1	Eastern Great Lakes Lowlands	E	10.97	8.82	4.04	2.71	1.64
8.1.10	Erie Drift Plain	E	18.39	15.10	8.07	4.99	2.81

	Ecoregion III		Median Total Sulfur Deposition (kg S/ha-yr)				
Code	Name	E/W	2001-03	2006-08	2010-12	2014-16	2018-20
8.1.3	Northern Allegheny Plateau	E	11.92	10.24	4.81	2.79	1.68
8.1.4	North Central Hardwood Forests	E	4.57	3.42	2.63	2.01	1.39
8.1.5	Driftless Area	E	5.39	5.00	3.37	2.61	1.95
8.1.6	Southern Michigan/Northern Indiana Drift Plains	E	9.62	8.34	5.32	3.25	2.16
8.1.7	Northeastern Coastal Zone	E	9.57	8.42	3.82	2.40	1.87
8.1.8	Acadian Plains and Hills	E	4.46	4.61	2.38	1.65	1.22
8.2.1	Southeastern Wisconsin Till Plains	E	7.02	6.37	3.98	2.74	2.02
8.2.2	Huron/Erie Lake Plains	E	9.86	8.59	5.22	3.15	2.11
8.2.3	Central Corn Belt Plains	E	9.78	8.96	5.42	4.11	2.45
8.2.4	Eastern Corn Belt Plains	E	14.84	11.98	7.08	4.11	2.59
8.3.1	Northern Piedmont	E	14.94	12.58	5.30	3.32	2.12
8.3.2	Interior River Valleys and Hills	E	10.55	9.30	6.20	4.29	3.03
8.3.3	Interior Plateau	E	13.52	10.96	6.24	4.16	2.73
8.3.4	Piedmont	E	11.71	9.58	4.34	2.62	1.89
8.3.5	Southeastern Plains	E	9.68	8.05	4.34	3.48	2.63
8.3.6	Mississippi Valley Loess Plains	E	8.64	6.69	4.60	3.96	3.18
8.3.7	South Central Plains	E	7.34	6.78	4.91	4.70	3.64
8.3.8	East Central Texas Plains	E	6.41	5.14	3.82	4.45	3.62
8.4.1	Ridge and Valley	E	14.10	11.86	5.31	3.23	2.14
8.4.2	Central Appalachians	E	16.20	13.28	7.05	4.12	2.32
8.4.3	Western Allegheny Plateau	E	20.35	16.36	8.26	4.76	2.89
8.4.4	Blue Ridge	E	11.12	9.26	4.41	2.61	1.95
8.4.5	Ozark Highlands	E	6.31	5.84	4.65	3.19	2.59
8.4.6	Boston Mountains	E	5.98	5.72	4.48	3.33	2.79
8.4.7	Arkansas Valley	E	5.54	5.20	4.15	3.38	2.97
8.4.8	Ouachita Mountains	E	6.20	5.82	4.67	4.09	3.52
8.4.9	Southwestern Appalachians	E	14.71	11.56	5.47	3.46	2.61
8.5.1	Middle Atlantic Coastal Plain	E	10.52	9.34	5.09	3.43	2.36
8.5.2	Mississippi Alluvial Plain	E	7.37	6.06	4.22	3.91	3.17
8.5.3	Southern Coastal Plain	E	7.94	6.02	4.43	3.95	3.23
8.5.4	Atlantic Coastal Pine Barrens	E	14.03	12.27	5.61	3.80	2.75
9.2.1	Northern Glaciated Plains	W	2.04	2.08	1.74	1.33	1.22
9.2.2	Lake Agassiz Plain	W	1.97	1.99	1.44	1.19	1.07
9.2.3	Western Corn Belt Plains	E	4.52	4.25	2.98	2.56	1.93
9.2.4	Central Irregular Plains	E	5.81	5.34	4.13	2.98	2.27
9.3.1	Northwestern Glaciated Plains	W	1.57	1.62	1.38	1.20	1.09
9.3.3	Northwestern Great Plains	W	1.20	1.33	1.01	0.88	0.82
9.3.4	Nebraska Sand Hills	W	1.67	1.99	1.48	1.36	1.36
9.4.1	High Plains	W	1.60	1.52	1.27	1.33	0.98
9.4.2	Central Great Plains	E	3.06	2.99	2.16	2.19	1.84

	Ecoregion III		Median Total Sulfur Deposition (kg S/ha-yr)				
Code	Name	E/W	2001-03	2006-08	2010-12	2014-16	2018-20
9.4.3	Southwestern Tablelands	W	1.30	1.24	0.99	1.12	0.65
9.4.4	Flint Hills	E	4.44	4.03	2.85	2.46	1.93
9.4.5	Cross Timbers	E	4.58	3.96	3.02	3.05	2.61
9.4.6	Edwards Plateau	E	3.07	2.76	2.21	2.54	2.10
9.4.7	Texas Blackland Prairies	E	6.15	4.87	3.85	4.02	3.39
9.5.1	Western Gulf Coastal Plain	E	6.95	5.64	4.31	4.74	4.33
9.6.1	Southern Texas Plains	E	3.72	3.03	2.54	3.09	2.36

Acidification of waterbodies is controlled by local factors such as geology, hydrology, etc. For this reason, aquatic CLs for acidification are unique to the waterbody itself and information about the waterbody, like water quality, is needed to determine its CL. Unfortunately, not all waterbodies within an ecoregion have sufficient data to calculate a CL. This is the case for many level III ecoregions (from this point on level III ecoregions will be referred to as ecoregions), except for ones that historically are known to be in acid sensitive areas. Acid sensitive areas typically have been heavily sampled, and, hence, contain many waterbody sites with estimated CLs (see Figure 5A-31). These areas tend to be in the eastern CONUS in such ecoregions as Central Appalachians (8.4.2), Northern Appalachian and Atlantic Maritime Highlands (5.3.1), and the Blue Ridge (8.4.4). Areas in the Rockies and Sierra Nevada also have been sampled extensively and contain many CLs.

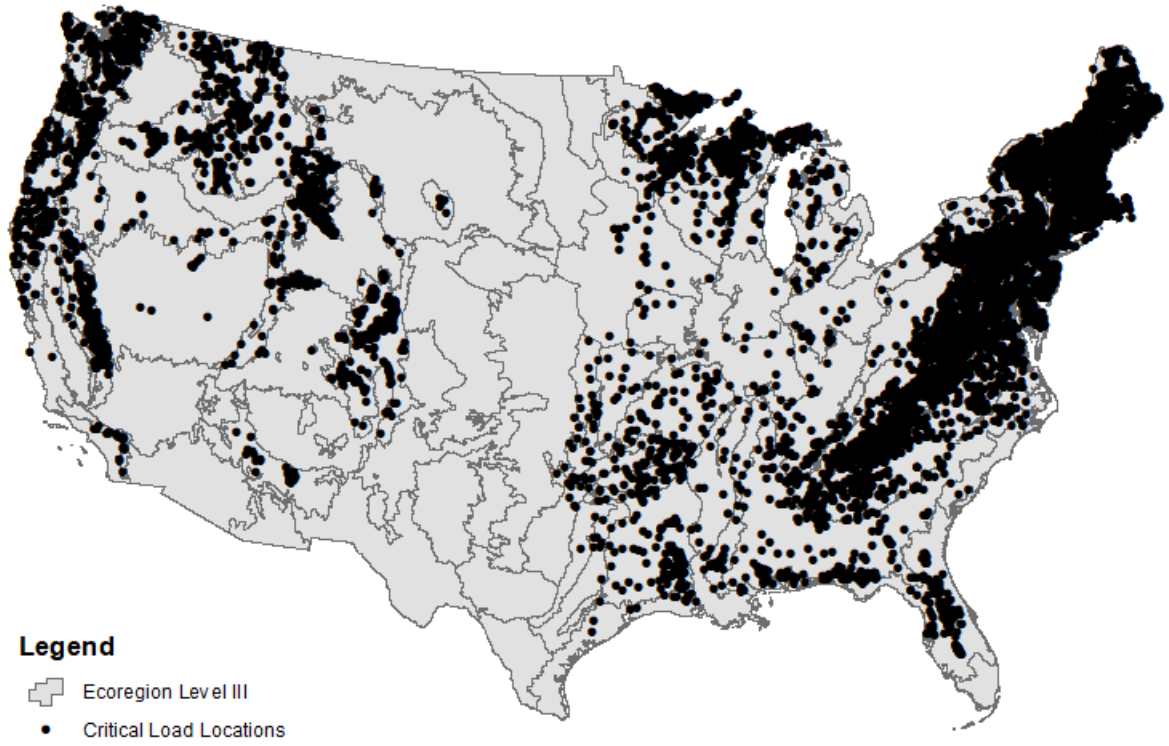

Figure 5A-31. Locations of aquatic critical loads mapped across level III ecoregions.

More CLs in an ecoregion helps to capture the spatial variability of acid sensitive areas across the landscape and provide a more accurate measurement of the impact of deposition driven acidification. Ecoregions with few CLs, however, fail to capture the spatial variability of acid sensitive areas, which in turn reduces the accuracy of the percentile CL value and limits our confidence in the estimated percent of exceedances. For this reason, although CL exceedances were derived for all ecoregions with a CL in NCLD, ecoregions containing greater than 50 CLs were the primary focus of the ecoregion-scale assessment summary (section 5A.2.2.2).

There are 84 ecoregions across the CONUS, 69 of which had at least one CL available. The Northern Appalachian and Atlantic Maritime Highlands ecoregion had the most CLs at 2,851 (see Table 5A-10**)**. Eleven ecoregions had 9 or fewer CLs and 58 ecoregions had 10 or more. Of the 58 ecoregions only 32 had 50 or more CLs. Three of the 32 are recognized to have acidity heavily influenced by natural acids (see section 5A.2.2.1).

The 10^{th} to 30^{th} percentile S only CL estimates for an ecoregion varied greatly among ecoregions from 1.2 to 136.1 kg/ha-yr (7.4 to 850.6 meq/m^2-yr) with an ANC threshold of 20 µeq/L to 0.1 to 134.9 kg/ha-yr (0.625 to 843.1 meq/m^2-yr) with an ANC threshold of 50 µeq/L (Tables 5A-12 and 5A-13). The lower values indicate ecoregions of higher sensitivity, in terms of risk of exceeding CLs based on the ANC threshold of 50 µeq/L. The most sensitive ecoregions include Sierra Nevada, Southern Coastal Plain, Idaho Batholith, Atlantic Coastal Pine Barrens, Blue Ridge, Middle Rockies, Wasatch and Uinta Mountains, Southern Rockies, and Central Appalachian and Atlantic Maritime Highlands. See Tables 5A-12 and 5A-13 for 10^{th}, 30^{th} percentile, minimum S only CL estimates for each of the 58 ecoregions with at least 10 CLs.

Table 5A-12. Summary of sulfur only CLs (kg S/ha-yr) for ANC thresholds of 20 and 30 µeq/L for ecoregions with at least 10 CL values.

Ecoregion III			ANC of 20 µeq/L			ANC of 30 µeq/L		
Name	Code	No. Sites	30th	10th	Min.	30th	10th	Min.
Northern Appalachian and Atlantic Maritime Highlands	5.3.1	2851	9.7	4.8	0.0	8.7	3.6	0.0
Ridge and Valley	8.4.1	1292	11.5	5.9	0.0	10.7	5.0	0.0
Blue Ridge	8.4.4	1972	9.1	5.3	0.0	7.6	4.0	0.0
Northern Lakes and Forests	5.2.1	839	5.1	3.0	0.0	4.7	2.6	0.0
Northeastern Coastal Zone	8.1.7	565	16.3	8.1	0.0	15.2	7.1	0.0
Middle Rockies	6.2.10	496	9.2	5.2	0.5	8.1	4.1	0.0
Acadian Plains and Hills	8.1.8	494	11.2	5.2	0.0	10.3	4.2	0.0
Piedmont	8.3.4	508	16.0	8.7	0.9	14.9	7.7	0.0
Southern Rockies	6.2.14	372	7.4	3.8	0.0	6.2	2.7	0.0
Central Appalachians	8.4.2	372	8.4	5.0	0.0	7.3	3.8	0.0
Sierra Nevada	6.2.12	353	4.7	1.6	0.0	3.4	0.1	0.0
Southeastern Plains	8.3.5	390	13.9	4.4	0.0	13.1	3.3	0.0
Atlantic Coastal Pine Barrens	8.5.4	234	6.2	2.0	0.0	5.4	1.3	0.0
Northern Piedmont	8.3.1	231	40.0	16.8	1.5	39.2	15.8	1.0

Ecoregion III			ANC of 20 µeq/L			ANC of 30 µeq/L		
Name	**Code**	**No. Sites**	**30th**	**10th**	**Min.**	**30th**	**10th**	**Min.**
North Central Appalachians	5.3.3	216	14.6	8.3	1.9	13.5	7.2	0.8
Northern Allegheny Plateau	8.1.3	199	22.2	11.8	0.2	21.1	10.9	0.0
Idaho Batholith	6.2.15	188	10.3	5.6	0.0	8.9	4.1	0.0
Cascades	6.2.7	179	13.4	3.3	0.0	12.2	1.9	0.0
North Cascades	6.2.5	162	26.3	11.5	0.0	23.9	9.9	0.0
Southern Coastal Plain	8.5.3	142	4.2	1.5	0.0	3.8	1.1	0.0
Coast Range	7.1.8	115	48.6	15.7	6.1	47.0	15.0	5.9
Middle Atlantic Coastal Plain	8.5.1	105	15.3	8.1	0.0	14.5	7.2	0.0
Wasatch and Uinta Mountains	6.2.13	96	11.0	7.7	2.1	10.4	6.7	1.6
North Central Hardwood Forests	8.1.4	94	23.0	5.8	2.8	22.0	4.8	0.1
Columbia Mountains/Northern Rockies	6.2.3	86	19.5	6.9	0.0	18.6	6.1	0.0
Eastern Great Lakes Lowlands	8.1.1	83	50.5	17.5	0.0	50.1	16.1	0.0
Klamath Mountains	6.2.11	81	27.6	12.4	7.3	26.5	11.6	6.2
Interior Plateau	8.3.3	71	66.8	10.9	5.3	65.8	9.8	3.2
Blue Mountains	6.2.9	63	18.1	8.6	3.6	16.7	7.5	2.5
South Central Plains	8.3.7	153	10.9	3.9	0.0	9.9	2.9	0.0
Ozark Highlands	8.4.5	56	48.3	13.5	2.8	47.4	12.5	1.7
Southwestern Appalachians	8.4.9	117	14.3	10.3	6.4	13.2	9.2	5.3
Ouachita Mountains	8.4.8	42	13.1	7.2	6.3	12.2	6.3	4.5
Strait of Georgia/Puget Lowland	7.1.7	38	28.9	10.5	4.6	28.3	9.2	3.6
Western Allegheny Plateau	8.4.3	35	18.7	8.2	5.0	17.7	7.0	4.3
Southern Michigan/Northern Indiana Drift Plains	8.1.6	33	11.5	5.8	2.1	10.8	4.4	1.3
Arkansas Valley	8.4.7	31	14.9	6.3	3.4	14.1	5.4	2.6
Canadian Rockies	6.2.4	31	41.8	8.3	3.5	40.4	7.8	1.6
Western Corn Belt Plains	9.2.3	26	14.8	5.8	4.6	14.0	4.6	2.9
Cross Timbers	9.4.5	26	11.3	7.1	2.9	10.0	5.2	0.8
Eastern Cascades Slopes and Foothills	6.2.8	27	21.5	6.6	3.6	20.9	5.9	2.5
Arizona/New Mexico Mountains	13.1.1	25	20.3	11.7	10.1	19.7	10.8	9.4
Willamette Valley	7.1.9	24	65.2	25.5	8.3	63.2	24.9	7.4
Boston Mountains	8.4.6	23	20.3	9.3	6.4	19.5	8.6	5.4
Southern & Baja California Pine-Oak Mtns	11.1.3	22	25.2	3.4	1.1	24.5	2.7	0.0
Central Irregular Plains	9.2.4	21	14.2	5.4	4.5	13.0	4.1	2.5
California Coastal Sage, Chaparral, and Oak Woodlands	11.1.1	21	34.9	4.6	2.7	34.4	3.8	2.3
Northern Basin and Range	10.1.3	20	19.1	10.1	3.2	18.7	8.9	1.7
Mississippi Alluvial Plain	8.5.2	19	12.5	4.7	0.6	11.1	3.5	0.0
Interior River Valleys and Hills	8.3.2	18	39.1	5.8	5.8	37.5	4.7	4.4
Driftless Area	8.1.5	15	54.5	25.0	17.8	54.2	24.2	17.0
Western Gulf Coastal Plain	9.5.1	16	52.2	20.6	10.3	51.6	19.7	9.4
Central Basin and Range	10.1.5	16	45.4	21.5	8.7	44.4	20.3	6.4
Eastern Corn Belt Plains	8.2.4	14	14.4	4.5	3.8	13.2	3.6	3.1
Erie Drift Plain	8.1.10	14	18.6	5.8	4.1	17.6	4.8	2.8
Mississippi Valley Loess Plains	8.3.6	41	14.5	4.2	1.6	13.4	3.1	0.5
East Central Texas Plains	8.3.8	10	16.6	1.2	0.3	15.4	0.8	0.0
Southeastern Wisconsin Till Plains	8.2.1	10	136.1	16.7	15.0	135.7	14.8	13.6

Table 5A-13. Summary of sulfur only CLs (kg S/ha-yr) for ANC thresholds of 50 and 50/20 µeq/L for ecoregions with at least 10 CL values.

Ecoregion III			ANC of 50 µeq/L			ANC of 50/20 µeq/L		
Name	Code	No. Sites	30th	10th	Min.	30th	10th	Min.
Northern Appalachian and Atlantic Maritime Highlands	5.3.1	2851	6.5	1.1	0.0	6.5	1.1	0.0
Ridge and Valley	8.4.1	1292	8.9	3.3	0.0	8.9	3.3	0.0
Blue Ridge	8.4.4	1972	4.7	1.1	0.0	4.7	1.1	0.0
Northern Lakes and Forests	5.2.1	839	3.8	1.5	0.0	3.8	1.5	0.0
Northeastern Coastal Zone	8.1.7	565	13.4	5.3	0.0	13.4	5.3	0.0
Middle Rockies	6.2.10	496	6.1	2.4	0.0	9.2	5.2	0.5
Acadian Plains and Hills	8.1.8	494	8.5	2.2	0.0	8.5	2.2	0.0
Piedmont	8.3.4	508	12.7	5.4	0.0	12.7	5.4	0.0
Southern Rockies	6.2.14	372	3.9	0.6	0.0	7.4	3.8	0.0
Central Appalachians	8.4.2	372	5.2	1.3	0.0	5.2	1.3	0.0
Sierra Nevada	6.2.12	353	0.7	0.0	0.0	4.7	1.6	0.0
Southeastern Plains	8.3.5	390	11.5	1.8	0.0	11.5	1.8	0.0
Atlantic Coastal Pine Barrens	8.5.4	234	3.8	0.0	0.0	3.8	0.0	0.0
Northern Piedmont	8.3.1	231	37.6	13.2	0.0	37.6	13.2	0.0
North Central Appalachians	5.3.3	216	11.5	4.9	0.0	11.5	4.9	0.0
Northern Allegheny Plateau	8.1.3	199	19.1	8.7	0.0	19.1	8.7	0.0
Idaho Batholith	6.2.15	188	7.2	1.0	0.0	10.3	5.6	0.0
Cascades	6.2.7	179	9.8	0.0	0.0	13.4	3.3	0.0
North Cascades	6.2.5	162	21.8	6.1	0.0	26.3	11.5	0.0
Southern Coastal Plain	8.5.3	142	2.9	0.2	0.0	2.9	0.2	0.0
Coast Range	7.1.8	115	42.5	14.0	4.9	48.6	15.7	6.1
Middle Atlantic Coastal Plain	8.5.1	105	13.2	5.3	0.0	13.2	5.3	0.0
Wasatch and Uinta Mountains	6.2.13	96	8.8	5.0	0.0	11.0	7.7	2.1
North Central Hardwood Forests	8.1.4	94	20.1	3.5	0.0	20.1	3.5	0.0
Columbia Mountains/Northern Rockies	6.2.3	86	16.8	4.0	0.0	19.5	6.9	0.0
Eastern Great Lakes Lowlands	8.1.1	83	49.7	13.1	0.0	49.7	13.1	0.0
Klamath Mountains	6.2.11	81	24.2	10.0	4.1	27.6	12.4	7.3
Interior Plateau	8.3.3	71	63.6	7.7	0.0	63.6	7.7	0.0
Blue Mountains	6.2.9	63	14.5	6.2	0.3	18.1	8.6	3.6
South Central Plains	8.3.7	153	8.2	1.0	0.0	8.2	1.0	0.0
Ozark Highlands	8.4.5	56	45.6	10.5	0.0	45.6	10.5	0.0
Southwestern Appalachians	8.4.9	117	10.9	7.0	3.1	10.9	7.0	3.1
Ouachita Mountains	8.4.8	42	10.2	4.8	0.4	10.2	4.8	0.4
Strait of Georgia/Puget Lowland	7.1.7	38	26.9	7.1	0.0	28.9	10.5	4.6
Western Allegheny Plateau	8.4.3	35	15.7	5.1	3.0	15.7	5.1	3.0
Southern Michigan/Northern Indiana Drift Plains	8.1.6	33	9.1	2.4	0.0	9.1	2.4	0.0
Arkansas Valley	8.4.7	31	12.5	4.7	1.2	12.5	4.7	1.2
Canadian Rockies	6.2.4	31	37.8	6.5	0.0	41.8	8.3	3.5
Western Corn Belt Plains	9.2.3	26	12.4	2.6	0.0	12.4	2.6	0.0
Cross Timbers	9.4.5	26	7.1	1.0	0.0	7.1	1.0	0.0
Eastern Cascades Slopes and Foothills	6.2.8	27	19.7	4.7	0.5	21.5	6.6	3.6

Ecoregion III			ANC of 50 µeq/L			ANC of 50/20 µeq/L		
Name	Code	No. Sites	30th	10th	Min.	30th	10th	Min.
Arizona/New Mexico Mountains	13.1.1	25	18.4	9.1	7.8	20.3	11.7	10.1
Willamette Valley	7.1.9	24	59.2	23.5	5.5	65.2	25.5	8.3
Boston Mountains	8.4.6	23	15.9	7.1	3.4	15.9	7.1	3.4
Southern & Baja California Pine-Oak Mtns	11.1.3	22	23.2	1.3	0.0	25.2	3.4	1.1
Central Irregular Plains	9.2.4	21	11.0	1.5	0.0	11.0	1.5	0.0
California Coastal Sage, Chaparral, and Oak Woodlands	11.1.1	21	33.4	2.4	1.4	34.9	4.6	2.7
Northern Basin and Range	10.1.3	20	18.0	6.4	0.0	19.1	10.1	3.2
Mississippi Alluvial Plain	8.5.2	19	8.2	0.4	0.0	8.2	0.4	0.0
Interior River Valleys and Hills	8.3.2	18	35.3	2.4	1.7	35.3	2.4	1.7
Driftless Area	8.1.5	15	53.5	22.5	15.4	53.5	22.5	15.4
Western Gulf Coastal Plain	9.5.1	16	50.3	17.9	7.6	50.3	17.9	7.6
Central Basin and Range	10.1.5	16	42.4	17.9	1.9	45.4	21.5	8.7
Eastern Corn Belt Plains	8.2.4	14	10.9	1.7	1.7	10.9	1.7	1.7
Erie Drift Plain	8.1.10	14	15.4	2.7	0.3	15.4	2.7	0.3
Mississippi Valley Loess Plains	8.3.6	41	11.3	1.0	0.0	11.3	1.0	0.0
East Central Texas Plains	8.3.8	10	12.9	0.6	0.0	12.9	0.6	0.0
Southeastern Wisconsin Till Plains	8.2.1	10	134.9	11.0	10.7	134.9	11.0	10.7

For the 69 ecoregions with at least one CL, the minimum, maximum and average total S deposition in each of the five deposition periods at each of the CL locations are summarized in Table 5A-14. The minimum to maximum range of total S deposition across these locations was 0.32 – 32.20 kg S/ha-yr for 2001-2003 and 0.27 – 7.59 kg S/ha-yr for 2018 – 2020. Average values ranged from 1.77 to 8.63 kg S/ha-yr for 2018-2020 to 2001-2003, respectively (Table 5A-14).

Table 5A-14. Summary of total S deposition (kg S/ha-yr) estimates (based on TDEP) at CL locations for 69 ecoregions with at least one CL.

	Total Sulfur Deposition (kg S/ha-yr)				
	2001-03	2006-08	2010-12	2014-16	2018-20
Minimum	0.32	0.31	0.36	0.52	0.27
Maximum	32.20	25.97	12.75	9.38	7.59
Average	8.63	7.39	3.76	2.55	1.77

The medians of the TDEP deposition estimates at all CL locations in each ecoregion with any CLs are presented in Table 5A-15. Ecoregions with the highest median total S deposition estimates were Western Allegheny Plateau (8.4.3), Erie Drift Plain (8.1.10), North Central Appalachians (5.3.3), Central Appalachians (8.4.2), Northern Piedmont (8.3.1), Eastern Corn Belt Plains (8.2.4), Southwestern Appalachians (8.4.9), and Ridge and Valley (8.4.1), all in the Mid-Atlantic region of the eastern U.S (5A-15).

Table 5A-15. Median total sulfur deposition (based on TDEP estimates at CL locations) for the 69 ecoregions with at least one CL.

Ecoregion Name	Code	E/W	No. CLs	2001-03 (kg/ha-yr)	2006-08 (kg/ha-yr)	2010-12 (kg/ha-yr)	2014-16 (kg/ha-yr)	2018-20 (kg/ha-yr)
Columbia Plateau	10.1.2	W	2	0.84	0.83	0.76	0.63	0.38
Northern Basin and Range	10.1.3	W	20	0.93	1.05	1.02	1.04	0.75
Wyoming Basin	10.1.4	W	3	0.77	0.76	0.70	0.68	0.59
Central Basin and Range	10.1.5	W	16	0.86	0.66	0.67	0.76	0.57
Colorado Plateaus	10.1.6	W	1	1.32	1.44	1.25	1.33	0.84
Snake River Plain	10.1.8	W	2	0.80	0.93	0.98	0.79	0.55
Southern and Central California Chaparral and Oak Woodlands	11.1.1	W	21	1.65	1.20	1.26	0.98	1.06
Central California Valley	11.1.2	W	2	2.17	1.70	1.54	1.46	1.19
Southern California Mountains	11.1.3	W	22	1.45	1.21	1.24	1.04	0.86
Arizona/New Mexico Mountains	13.1.1	W	25	2.07	2.58	1.96	1.47	0.81
Northern Lakes and Forests	5.2.1	E	839	4.01	3.10	2.34	1.84	1.31
Northern Minnesota Wetlands	5.2.2	E	2	2.19	2.21	1.51	1.20	0.91
Northern Appalachian and Atlantic Maritime Highlands	5.3.1	E	2851	7.29	6.12	3.12	2.22	1.48
North Central Appalachians	5.3.3	E	216	15.73	13.37	5.83	3.17	2.17
Middle Rockies	6.2.10	W	496	1.48	1.53	1.33	1.06	0.87
Klamath Mountains	6.2.11	W	81	0.92	1.07	1.06	0.99	0.84
Sierra Nevada	6.2.12	W	353	1.40	1.24	1.27	1.17	1.01
Wasatch and Uinta Mountains	6.2.13	W	96	1.75	1.92	1.64	1.72	1.11
Southern Rockies	6.2.14	W	372	1.63	1.70	1.29	1.10	0.74
Idaho Batholith	6.2.15	W	188	1.21	1.52	1.39	1.14	0.72
Northern Rockies	6.2.3	W	86	1.18	1.22	1.02	0.93	0.62
Canadian Rockies	6.2.4	W	31	1.27	1.43	1.08	0.99	0.79
North Cascades	6.2.5	W	162	1.94	1.83	1.47	1.48	1.19
Cascades	6.2.7	W	179	1.25	1.51	1.25	1.23	1.07
Eastern Cascades Slopes and Foothills	6.2.8	W	27	0.66	0.75	0.73	0.74	0.62
Blue Mountains	6.2.9	W	63	0.63	0.68	0.72	0.85	0.46
Puget Lowland	7.1.7	W	38	2.28	1.94	1.55	2.25	1.36
Coast Range	7.1.8	W	115	2.49	2.31	2.07	2.09	1.52
Willamette Valley	7.1.9	W	24	1.71	1.44	1.45	1.76	1.08
Eastern Great Lakes Lowlands	8.1.1	E	83	8.04	6.50	3.26	2.16	1.44
Erie Drift Plain	8.1.10	E	14	18.62	15.49	7.83	5.14	2.84
Northern Allegheny Plateau	8.1.3	E	199	11.69	10.45	4.69	2.70	1.73
North Central Hardwood Forests	8.1.4	E	94	5.30	3.72	2.86	2.12	1.48
Driftless Area	8.1.5	E	15	6.16	5.34	3.56	2.76	2.11
Southern Michigan/Northern Indiana Drift Plains	8.1.6	E	33	10.36	8.99	5.41	3.35	2.37
Northeastern Coastal Zone	8.1.7	E	565	9.29	8.28	3.71	2.30	1.91
Acadian Plains and Hills	8.1.8	E	494	4.98	5.42	2.83	1.95	1.44

Ecoregion Name	Code	E/W	No. CLs	2001-03 (kg/ha-yr)	2006-08 (kg/ha-yr)	2010-12 (kg/ha-yr)	2014-16 (kg/ha-yr)	2018-20 (kg/ha-yr)
Southeastern Wisconsin Till Plains	8.2.1	E	10	6.94	5.71	3.93	2.74	1.96
Central Corn Belt Plains	8.2.3	E	2	10.64	9.79	5.98	4.44	2.50
Eastern Corn Belt Plains	8.2.4	E	14	17.43	13.48	7.90	4.76	2.87
Northern Piedmont	8.3.1	E	231	15.18	12.94	5.63	3.33	2.21
Interior River Valleys and Hills	8.3.2	E	18	12.59	11.03	6.54	4.25	2.94
Interior Plateau	8.3.3	E	71	13.11	9.84	5.58	4.01	2.74
Piedmont	8.3.4	E	508	12.26	10.14	4.24	2.69	2.03
Southeastern Plains	8.3.5	E	390	10.88	9.14	4.83	3.49	2.41
Mississippi Valley Loess Plains	8.3.6	E	41	9.40	7.66	4.72	4.44	3.57
South Central Plains	8.3.7	E	153	7.77	7.15	5.03	4.69	3.88
East Central Texas Plains	8.3.8	E	10	6.36	6.37	4.65	4.78	3.79
Ridge and Valley	8.4.1	E	1292	14.18	11.93	5.71	3.33	1.94
Central Appalachians	8.4.2	E	372	17.03	13.98	7.25	4.09	2.43
Western Allegheny Plateau	8.4.3	E	35	17.08	14.12	7.59	4.19	2.56
Blue Ridge	8.4.4	E	1972	11.29	9.58	4.41	2.70	2.06
Ozark Highlands	8.4.5	E	56	6.95	6.18	4.87	3.24	2.66
Boston Mountains	8.4.6	E	23	6.25	5.90	4.60	3.43	2.78
Arkansas Valley	8.4.7	E	31	5.70	5.38	4.24	3.35	2.91
Ouachita Mountains	8.4.8	E	42	6.09	5.71	4.65	4.05	3.58
Southwestern Appalachians	8.4.9	E	117	17.27	14.44	5.59	4.17	2.93
Middle Atlantic Coastal Plain	8.5.1	E	105	14.10	12.07	5.35	3.58	2.41
Mississippi Alluvial Plain	8.5.2	E	19	7.02	5.45	4.06	3.67	3.05
Southern Coastal Plain	8.5.3	E	142	8.70	5.92	4.56	4.18	3.35
Atlantic Coastal Pine Barrens	8.5.4	E	234	13.88	12.01	5.40	3.89	2.84
Western Corn Belt Plains	9.2.3	E	26	4.72	4.01	2.85	2.35	1.99
Central Irregular Plains	9.2.4	E	21	5.55	5.12	3.99	2.95	2.29
Northwestern Glaciated Plains	9.3.1	E	2	0.67	0.74	0.54	0.56	0.46
Central Great Plains	9.4.2	E	5	4.32	4.67	2.86	2.73	2.44
Flint Hills	9.4.4	E	7	4.45	4.36	2.91	2.57	2.27
Cross Timbers	9.4.5	E	26	4.89	4.47	3.25	3.17	2.72
Texas Blackland Prairies	9.4.7	E	3	6.51	5.95	4.47	4.37	3.66
Western Gulf Coastal Plain	9.5.1	E	16	7.59	6.99	4.92	5.31	4.34

5A.2.2.1 Ecoregion Critical Load Exceedances – Sulfur Only

Of the 69 ecoregions that had at least one CL, 58 ecoregions had 10 or more values. We evaluated CL exceedances and summarize sites with CLs in each ecoregion based on two categories of CLs: (1) all CLs and (2) only CLs with positive values. Exceedances were evaluated with respect to 2001-2003, 2006-2008, 2012-2014, 2014-2016, and 2018-2020 TDEP deposition estimates for S only. Exceedances were calculated for ANC thresholds of 20, 30, 50 µeq/L and combined 50 µeq/L in the East and 20 µeq/L in the West (denoted as 50/20 µeq/L). See section 5A.1.6 above for a description of how exceedances were calculated. Information

about S only exceedances in the 58 ecoregions with 10 or more CL sites are summarized in Table 5A-16 for each ANC threshold and time period.

Results of S only exceedances per ecoregion for the 69 ecoregions with at least a single CL estimate included in Tables 5A-17 through 5A-24. For ANC thresholds of 20 and 30 µeq/L, and the most recent years (2018-2020 and 2014-2016), there were few exceedances in either the 58 ecoregions with at least 10 CLs or the 69 ecoregions with at least a single CL estimate. Of the 69 ecoregions (and focusing on the CLs greater than zero), 48 and 40 had no exceedances for an ANC threshold of 20 µeq/L for 2018-2020 and 2014-2016, respectively. Of the remaining 21 and 29 ecoregions, only 6 and 9 had greater than 5% exceedance and 3 and 5 had greater than 10% for ANC thresholds of 20 µeq/L for the two deposition periods. For the following three deposition periods 2010-2012, 2006-2008, and 2001-2003, the number of ecoregions without an exceedance decreased to 35, 31, and 29 while the number with greater than 10% exceedance increased to 8, 21, and 23, respectively (Tables 5A-17 and 5A-18). There were slightly more exceedances for CLs based on an ANC threshold of 30 µeq/L across all deposition periods (Tables 5A-19 and 5A-20). Critical loads determined for ANC thresholds of 50 and 50/20 µeq/L were exceeded in more sites within ecoregions and there were more ecoregions with exceedances particularly for the early deposition periods of 2010-2012, 2006-2008, and 2001-2003 (Tables 5A-21 to 5A-24 and Figures 5A-38 to 5A-43). For CLs using an ANC threshold of 50 µeq/L, 31, 25, 21, 21, and 21 of the 58 ecoregions had no CL exceedances for the 5 deposition periods 2018-2020, 2014-2016, 2010-2012, 2006-2008, and 2001-2003. Of the remaining ecoregions, 13,17, 36, 43, and 44 had greater than 5% exceedances and 8, 9, 25, 33, and 35 ecoregions had exceedance percentage greater than 10%.

The Southeastern Plains (code 8.3.5), Southern Coastal Plain (code 8.5.3), and Atlantic Coastal Pine Barrens (code 8.5.4) are ecoregions known to have naturally acidic surface waters and the high exceedances calculated for these ecoregions are likely not driven by air pollution deposition but instead by natural acidity linked to DOC, hydrology, and natural biogeochemical processes (2008 ISA, section 3.2.4.2; Baker et al., 1991; Herlihy et al., 1991). Central Appalachians (8.4.2), Acadian Plains and Hills (8.1.8), and Northern Appalachian and Atlantic Maritime Highlands (5.3.1) are ecoregions know to be acid sensitive (Table 5A-5).

Table 5A-16. Summary of CL values for those that have been exceeded for each ANC threshold and time period for the 58 ecoregions with 10 or more values.

	CL Value *	S Deposition estimates for CLs that exceed**	Average percentage of sites/ecoregion exceeding their CLs	Number of ecoregions with more than the specified percentage of sites exceeding their CLs			
Time Period	Average (5^{th} - 95^{th} percentile) kg S/ha-yr	Average (5^{th} - 95^{th} percentile) kg S/ha-yr		# >5% EX	# >10% EX	# >15% EX	#>25% EA
ANC Threshold = 50/20 µeq/L							
2018-2020	1 (0.1-2.4)	2.1 (1.2-3.6)	3.1	12	7	2	0
2014-2016	1.5 (0.2-3.4)	3 (1.7-5)	4.0	16	8	3	0
2010-2012	2.4 (0.2-5.4)	2.4 (0.2-5.4)	6.1	24	16	7	3
2006-2008	4.9 (0.5-11.2)	9.5 (3.3-15)	12.1	33	24	20	10
2001-2003	5.7 (0.5-13)	11.4 (4.3-17.9)	14.4	33	27	20	13
ANC Threshold = 50 µeq/L							
2018-2020	1 (0.1-2.4)	2.1 (1-3.6)	3.2	12	7	0	0
2014-2016	1.5 (0.2-3.4)	3 (1.5-5)	4.3	16	8	3	0
2010-2012	2.3 (0.2-5.4)	4.5 (2-7.7)	6.3	25	16	7	3
2006-2008	4.9 (0.4-11.1)	9.4 (3.1-15)	12.4	33	24	20	10
2001-2003	5.6 (0.5-12.9)	11.3 (4-17.9)	14.6	33	27	20	13
ANC Threshold = 30 µeq/L							
2018-2020	1.3 (0.1-3)	2.2 (0.9-3.4)	1.9	5	4	1	0
2014-2016	1.7 (0.2-3.9)	3.1 (1.3-5.3)	3.0	11	5	2	1
2010-2012	2.7 (0.3-5.7)	4.6 (1.7-7.8)	4.7	20	11	4	1
2006-2008	5.6 (0.8-11.5)	9.7 (3.2-15.1)	10.3	30	23	17	8
2001-2003	6.6 (0.9-13.7)	11.5 (4.2-17.9)	13.1	31	24	20	12
ANC Threshold = 20 µeq/L							
2018-2020	1.4 (0.2-3.2)	2.3 (0.9-4.3)	1.5	6	3	1	0
2014-2016	1.9 (0.3-4.1)	3.4 (1.5-5.4)	2.7	8	4	3	2
2010-2012	2.9 (0.5-6.1)	4.8 (2.1-8.1)	4.2	17	7	3	2
2006-2008	6.1 (1.1-11.8)	9.8 (4.1-15.2)	9.4	27	20	17	8
2001-2003	7.1 (1.3-14.1)	11.8 (4.5-18.1)	12.3	28	22	21	11

* This summarizes the magnitude of the CL values of those that were exceeded by the deposition estimated in these time periods. This summary is based on CL values greater than zero.
** This summarizes the magnitude of deposition estimates that yielded CL exceedances in these time periods.

Table 5A-17. Percent exceedances of aquatic CLs for S only and ANC threshold of 20 µeq/L for deposition years of 2018-20 and 2014-16 in 69 ecoregions.

Ecoregion (n=69)		Sulfur only - ANC = 20 ueq/L						
		Number of CLs			% Exceedances			
		Total Number	CL≤0		2018-20		2014-16	
Name	Code		n	% of total	All	CL>0	All	CL>0
Northern Appalachian and Atlantic Maritime Highlands	5.3.1	2851	11	0.4	1.6	1.2	2.7	2.4
Blue Ridge	8.4.4	1972	3	0.2	0.9	0.7	1.8	1.6
Ridge and Valley	8.4.1	1292	2	0.2	0.9	0.8	4.7	4.6
Northern Lakes and Forests	5.2.1	839	1	0.1	0.5	0.4	1.2	1.1
Northeastern Coastal Zone	8.1.7	565	1	0.2	0.2	0.0	0.9	0.7
Piedmont	8.3.4	508	0	0.0	0.2	0.2	0.6	0.6
Middle Rockies	6.2.10	496	0	0.0	0.2	0.2	0.6	0.6
Acadian Plains and Hills	8.1.8	494	2	0.4	2.0	1.6	2.6	2.2
Southeastern Plains	8.3.5	390	3	0.8	4.9	4.1	6.9	6.2
Central Appalachians	8.4.2	372	4	1.1	3.8	2.7	5.6	4.6
Southern Rockies	6.2.14	372	1	0.3	0.5	0.3	1.9	1.6
Sierra Nevada	6.2.12	353	11	3.1	4.2	1.1	5.9	2.8
Atlantic Coastal Pine Barrens	8.5.4	234	1	0.4	12.4	12.0	17.9	17.5
Northern Piedmont	8.3.1	231	0	0.0	0.0	0.0	0.4	0.4
North Central Appalachians	5.3.3	216	0	0.0	0.0	0.0	2.3	2.3
Northern Allegheny Plateau	8.1.3	199	0	0.0	0.5	0.5	1.0	1.0
Idaho Batholith	6.2.15	188	1	0.5	0.5	0.0	0.5	0.0
Cascades	6.2.7	179	4	2.2	3.9	1.7	5.0	2.8
North Cascades	6.2.5	162	1	0.6	0.6	0.0	0.6	0.0
South Central Plains	8.3.7	153	2	1.3	9.8	8.5	12.4	11.1
Southern Coastal Plain	8.5.3	142	1	0.7	20.4	19.7	27.5	26.8
Southwestern Appalachians	8.4.9	117	0	0.0	0.0	0.0	0.0	0.0
Coast Range	7.1.8	115	0	0.0	0.0	0.0	0.0	0.0
Middle Atlantic Coastal Plain	8.5.1	105	1	1.0	2.9	1.9	4.8	3.8
Wasatch and Uinta Mountains	6.2.13	96	0	0.0	0.0	0.0	0.0	0.0
North Central Hardwood Forests	8.1.4	94	0	0.0	0.0	0.0	0.0	0.0
Columbia Mountains/Northern Rockies	6.2.3	86	1	1.2	1.2	0.0	2.3	1.2
Eastern Great Lakes Lowlands	8.1.1	83	1	1.2	1.2	0.0	1.2	0.0
Klamath Mountains	6.2.11	81	0	0.0	0.0	0.0	0.0	0.0
Interior Plateau	8.3.3	71	0	0.0	0.0	0.0	0.0	0.0
Blue Mountains	6.2.9	63	0	0.0	0.0	0.0	0.0	0.0
Ozark Highlands	8.4.5	56	0	0.0	0.0	0.0	1.8	1.8
Ouachita Mountains	8.4.8	42	0	0.0	0.0	0.0	0.0	0.0
Mississippi Valley Loess Plains	8.3.6	41	0	0.0	2.4	2.4	9.8	9.8
Strait of Georgia/Puget Lowland	7.1.7	38	0	0.0	0.0	0.0	0.0	0.0
Western Allegheny Plateau	8.4.3	35	0	0.0	0.0	0.0	0.0	0.0

Ecoregion (n=69)		Sulfur only - ANC = 20 ueq/L						
		Number of CLs			% Exceedances			
		Total Number	CL≤0		2018-20		2014-16	
Name	Code		n	% of total	All	CL>0	All	CL>0
Southern Michigan/Northern Indiana Drift Plains	8.1.6	33	0	0.0	0.0	0.0	3.0	3.0
Arkansas Valley	8.4.7	31	0	0.0	0.0	0.0	0.0	0.0
Canadian Rockies	6.2.4	31	0	0.0	0.0	0.0	0.0	0.0
Eastern Cascades Slopes and Foothills	6.2.8	27	0	0.0	0.0	0.0	0.0	0.0
Cross Timbers	9.4.5	26	0	0.0	0.0	0.0	0.0	0.0
Western Corn Belt Plains	9.2.3	26	0	0.0	0.0	0.0	0.0	0.0
Arizona/New Mexico Mountains	13.1.1	25	0	0.0	0.0	0.0	0.0	0.0
Willamette Valley	7.1.9	24	0	0.0	0.0	0.0	0.0	0.0
Boston Mountains	8.4.6	23	0	0.0	0.0	0.0	0.0	0.0
Southern and Baja California Pine-Oak Mountains	11.1.3	22	0	0.0	0.0	0.0	4.5	4.5
Central Irregular Plains	9.2.4	21	0	0.0	0.0	0.0	0.0	0.0
California Coastal Sage, Chaparral, and Oak Woodlands	11.1.1	21	0	0.0	0.0	0.0	0.0	0.0
Northern Basin and Range	10.1.3	20	0	0.0	0.0	0.0	0.0	0.0
Mississippi Alluvial Plain	8.5.2	19	0	0.0	5.3	5.3	5.3	5.3
Interior River Valleys and Hills	8.3.2	18	0	0.0	0.0	0.0	0.0	0.0
Western Gulf Coastal Plain	9.5.1	16	0	0.0	0.0	0.0	0.0	0.0
Central Basin and Range	10.1.5	16	0	0.0	0.0	0.0	0.0	0.0
Driftless Area	8.1.5	15	0	0.0	0.0	0.0	0.0	0.0
Erie Drift Plain	8.1.10	14	0	0.0	0.0	0.0	0.0	0.0
Eastern Corn Belt Plains	8.2.4	14	0	0.0	0.0	0.0	0.0	0.0
East Central Texas Plains	8.3.8	10	0	0.0	10.0	10.0	10.0	10.0
Southeastern Wisconsin Till Plains	8.2.1	10	0	0.0	0.0	0.0	0.0	0.0
Flint Hills	9.4.4	7	0	0.0	14.3	14.3	28.6	28.6
Central Great Plains	9.4.2	5	0	0.0	0.0	0.0	0.0	0.0
Wyoming Basin	10.1.4	3	0	0.0	0.0	0.0	0.0	0.0
Texas Blackland Prairies	9.4.7	3	0	0.0	0.0	0.0	0.0	0.0
Northern Minnesota Wetlands	5.2.2	2	0	0.0	0.0	0.0	50.0	50.0
Snake River Plain	10.1.8	2	0	0.0	0.0	0.0	0.0	0.0
Central California Valley	11.1.2	2	0	0.0	0.0	0.0	0.0	0.0
Columbia Plateau	10.1.2	2	0	0.0	0.0	0.0	0.0	0.0
Central Corn Belt Plains	8.2.3	2	0	0.0	0.0	0.0	0.0	0.0
Northwestern Glaciated Plains	9.3.1	2	0	0.0	0.0	0.0	0.0	0.0
Colorado Plateaus	10.1.6	1	0	0.0	0.0	0.0	0.0	0.0

Table 5A-18. Percent exceedances of CLs for S only and ANC threshold of 20 µeq/L for deposition years of 2010-12, 2006-08 and 2001-03 in 69 ecoregions.

Ecoregion (n=69)		Sulfur only - ANC = 20 ueq/L								
		Number of CLs			% Exceedances					
		Total Number	CL≤0		2010-2012		2006-2008		2001-2003	
Name	Code		n	% of total	All	CL>0	All	CL>0	All	CL>0
Northern Appalachian and Atlantic Maritime Highlands	5.3.1	2851	11	0.4	4.5	4.1	14.9	14.5	19.9	19.5
Blue Ridge	8.4.4	1972	3	0.2	6.1	5.9	32.3	32.2	43.7	43.5
Ridge and Valley	8.4.1	1292	2	0.2	9.8	9.6	29.3	29.2	38.5	38.3
Northern Lakes and Forests	5.2.1	839	1	0.1	2.9	2.7	8.8	8.7	16.0	15.9
Northeastern Coastal Zone	8.1.7	565	1	0.2	2.1	1.9	8.3	8.1	10.1	9.9
Piedmont	8.3.4	508	0	0.0	1.8	1.8	11.8	11.8	16.1	16.1
Middle Rockies	6.2.10	496	0	0.0	0.8	0.8	0.8	0.8	0.8	0.8
Acadian Plains and Hills	8.1.8	494	2	0.4	4.7	4.3	10.5	10.1	10.1	9.7
Southeastern Plains	8.3.5	390	3	0.8	9.5	8.7	20.5	19.7	24.1	23.3
Central Appalachians	8.4.2	372	4	1.1	15.9	14.8	44.4	43.3	53.8	52.7
Southern Rockies	6.2.14	372	1	0.3	2.2	1.9	2.7	2.4	2.7	2.4
Sierra Nevada	6.2.12	353	11	3.1	5.1	2.0	5.1	2.0	6.2	3.1
Atlantic Coastal Pine Barrens	8.5.4	234	1	0.4	23.9	23.5	45.3	44.9	53.8	53.4
Northern Piedmont	8.3.1	231	0	0.0	1.7	1.7	6.5	6.5	7.8	7.8
North Central Appalachians	5.3.3	216	0	0.0	5.6	5.6	24.1	24.1	31.5	31.5
Northern Allegheny Plateau	8.1.3	199	0	0.0	2.0	2.0	7.5	7.5	9.5	9.5
Idaho Batholith	6.2.15	188	1	0.5	0.5	0.0	0.5	0.0	0.5	0.0
Cascades	6.2.7	179	4	2.2	3.9	1.7	3.9	1.7	3.9	1.7
North Cascades	6.2.5	162	1	0.6	0.6	0.0	0.6	0.0	0.6	0.0
South Central Plains	8.3.7	153	2	1.3	13.7	12.4	19.0	17.6	20.3	19.0
Southern Coastal Plain	8.5.3	142	1	0.7	28.9	28.2	35.2	34.5	49.3	48.6
Southwestern Appalachians	8.4.9	117	0	0.0	0.0	0.0	25.6	25.6	41.0	41.0
Coast Range	7.1.8	115	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Middle Atlantic Coastal Plain	8.5.1	105	1	1.0	5.7	4.8	20.0	19.0	24.8	23.8
Wasatch and Uinta Mountains	6.2.13	96	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
North Central Hardwood Forests	8.1.4	94	0	0.0	0.0	0.0	0.0	0.0	3.2	3.2
Columbia Mountains/Northern Rockies	6.2.3	86	1	1.2	3.5	2.3	3.5	2.3	3.5	2.3
Eastern Great Lakes Lowlands	8.1.1	83	1	1.2	1.2	0.0	2.4	1.2	6.0	4.8
Klamath Mountains	6.2.11	81	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Interior Plateau	8.3.3	71	0	0.0	5.6	5.6	8.5	8.5	12.7	12.7
Blue Mountains	6.2.9	63	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Ozark Highlands	8.4.5	56	0	0.0	3.6	3.6	3.6	3.6	3.6	3.6
Ouachita Mountains	8.4.8	42	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Mississippi Valley Loess Plains	8.3.6	41	0	0.0	12.2	12.2	17.1	17.1	19.5	19.5

Ecoregion (n=69)		Sulfur only - ANC = 20 ueq/L								
		Number of CLs			% Exceedances					
		Total Number	CL≤0		2010-2012		2006-2008		2001-2003	
Name	Code		n	% of total	All	CL>0	All	CL>0	All	CL>0
Strait of Georgia/Puget Lowland	7.1.7	38	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Western Allegheny Plateau	8.4.3	35	0	0.0	2.9	2.9	20.0	20.0	28.6	28.6
Southern Michigan/Northern Indiana Drift Plains	8.1.6	33	0	0.0	6.1	6.1	15.2	15.2	21.2	21.2
Arkansas Valley	8.4.7	31	0	0.0	3.2	3.2	3.2	3.2	3.2	3.2
Canadian Rockies	6.2.4	31	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Eastern Cascades Slopes and Foothills	6.2.8	27	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Cross Timbers	9.4.5	26	0	0.0	0.0	0.0	3.8	3.8	7.7	7.7
Western Corn Belt Plains	9.2.3	26	0	0.0	0.0	0.0	0.0	0.0	3.8	3.8
Arizona/New Mexico Mountains	13.1.1	25	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Willamette Valley	7.1.9	24	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Boston Mountains	8.4.6	23	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Southern and Baja California Pine-Oak Mountains	11.1.3	22	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Central Irregular Plains	9.2.4	21	0	0.0	0.0	0.0	4.8	4.8	4.8	4.8
California Coastal Sage, Chaparral, and Oak Woodlands	11.1.1	21	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Northern Basin and Range	10.1.3	20	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Mississippi Alluvial Plain	8.5.2	19	0	0.0	5.3	5.3	5.3	5.3	21.1	21.1
Interior River Valleys and Hills	8.3.2	18	0	0.0	5.6	5.6	16.7	16.7	22.2	22.2
Western Gulf Coastal Plain	9.5.1	16	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Central Basin and Range	10.1.5	16	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Driftless Area	8.1.5	15	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Erie Drift Plain	8.1.10	14	0	0.0	7.1	7.1	21.4	21.4	28.6	28.6
Eastern Corn Belt Plains	8.2.4	14	0	0.0	14.3	14.3	28.6	28.6	28.6	28.6
East Central Texas Plains	8.3.8	10	0	0.0	10.0	10.0	10.0	10.0	10.0	10.0
Southeastern Wisconsin Till Plains	8.2.1	10	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Flint Hills	9.4.4	7	0	0.0	28.6	28.6	28.6	28.6	28.6	28.6
Central Great Plains	9.4.2	5	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Wyoming Basin	10.1.4	3	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Texas Blackland Prairies	9.4.7	3	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Northern Minnesota Wetlands	5.2.2	2	0	0.0	50.0	50.0	50.0	50.0	50.0	50.0
Snake River Plain	10.1.8	2	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Central California Valley	11.1.2	2	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Columbia Plateau	10.1.2	2	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Central Corn Belt Plains	8.2.3	2	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Northwestern Glaciated Plains	9.3.1	2	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Colorado Plateaus	10.1.6	1	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0

Table 5A-19. Percent exceedances of aquatic CLs for S only and ANC threshold of 30 µeq/L for deposition years of 2018-20 and 2014-16 in 69 ecoregions.

Ecoregion (n=69)		Sulfur only - ANC = 30 ueq/L						
		Number of CLs			% Exceedances			
		Total Number	CL≤0		2018-2020		2014-2016	
Name	Code		n	% of total	All	CL>0	All	CL>0
Northern Appalachian and Atlantic Maritime Highlands	5.3.1	2851	40	1.4	3.0	1.6	4.9	3.5
Blue Ridge	8.4.4	1972	13	0.7	2.3	1.6	3.9	3.2
Ridge and Valley	8.4.1	1292	8	0.6	2.4	1.8	5.8	5.2
Northern Lakes and Forests	5.2.1	839	1	0.1	1.0	0.8	2.4	2.3
Northeastern Coastal Zone	8.1.7	565	1	0.2	1.2	1.1	1.6	1.4
Piedmont	8.3.4	508	1	0.2	1.0	0.8	1.2	1.0
Middle Rockies	6.2.10	496	4	0.8	1.4	0.6	1.6	0.8
Acadian Plains and Hills	8.1.8	494	9	1.8	3.8	2.0	4.7	2.8
Southeastern Plains	8.3.5	390	9	2.3	7.2	4.9	9.5	7.2
Central Appalachians	8.4.2	372	10	2.7	5.6	3.0	8.3	5.6
Southern Rockies	6.2.14	372	8	2.2	3.0	0.8	4.0	1.9
Sierra Nevada	6.2.12	353	29	8.2	11.9	3.7	16.4	8.2
Atlantic Coastal Pine Barrens	8.5.4	234	9	3.8	17.9	14.1	22.2	18.4
Northern Piedmont	8.3.1	231	0	0.0	1.3	1.3	1.3	1.3
North Central Appalachians	5.3.3	216	0	0.0	1.4	1.4	2.3	2.3
Northern Allegheny Plateau	8.1.3	199	1	0.5	1.0	0.5	1.5	1.0
Idaho Batholith	6.2.15	188	3	1.6	2.1	0.5	2.7	1.1
Cascades	6.2.7	179	11	6.1	6.7	0.6	7.3	1.1
North Cascades	6.2.5	162	1	0.6	0.6	0.0	0.6	0.0
South Central Plains	8.3.7	153	3	2.0	14.4	12.4	16.3	14.4
Southern Coastal Plain	8.5.3	142	4	2.8	22.5	19.7	30.3	27.5
Southwestern Appalachians	8.4.9	117	0	0.0	0.0	0.0	0.0	0.0
Coast Range	7.1.8	115	0	0.0	0.0	0.0	0.0	0.0
Middle Atlantic Coastal Plain	8.5.1	105	1	1.0	2.9	1.9	4.8	3.8
Wasatch and Uinta Mountains	6.2.13	96	0	0.0	0.0	0.0	0.0	0.0
North Central Hardwood Forests	8.1.4	94	0	0.0	1.1	1.1	1.1	1.1
Columbia Mountains/Northern Rockies	6.2.3	86	3	3.5	3.5	0.0	3.5	0.0
Eastern Great Lakes Lowlands	8.1.1	83	1	1.2	1.2	0.0	1.2	0.0
Klamath Mountains	6.2.11	81	0	0.0	0.0	0.0	0.0	0.0
Interior Plateau	8.3.3	71	0	0.0	0.0	0.0	0.0	0.0
Blue Mountains	6.2.9	63	0	0.0	0.0	0.0	0.0	0.0
Ozark Highlands	8.4.5	56	0	0.0	3.6	3.6	3.6	3.6
Ouachita Mountains	8.4.8	42	0	0.0	0.0	0.0	0.0	0.0
Mississippi Valley Loess Plains	8.3.6	41	0	0.0	9.8	9.8	14.6	14.6
Strait of Georgia/Puget Lowland	7.1.7	38	0	0.0	0.0	0.0	0.0	0.0

Ecoregion (n=69)		Sulfur only - ANC = 30 ueq/L						
		Number of CLs			% Exceedances			
		Total Number	CL≤0		2018-2020		2014-2016	
Name	Code		n	% of total	All	CL>0	All	CL>0
Western Allegheny Plateau	8.4.3	35	0	0.0	0.0	0.0	0.0	0.0
Southern Michigan/Northern Indiana Drift Plains	8.1.6	33	0	0.0	3.0	3.0	3.0	3.0
Arkansas Valley	8.4.7	31	0	0.0	0.0	0.0	3.2	3.2
Canadian Rockies	6.2.4	31	0	0.0	0.0	0.0	0.0	0.0
Eastern Cascades Slopes and Foothills	6.2.8	27	0	0.0	0.0	0.0	3.7	3.7
Cross Timbers	9.4.5	26	0	0.0	3.8	3.8	3.8	3.8
Western Corn Belt Plains	9.2.3	26	0	0.0	0.0	0.0	0.0	0.0
Arizona/New Mexico Mountains	13.1.1	25	0	0.0	0.0	0.0	0.0	0.0
Willamette Valley	7.1.9	24	0	0.0	0.0	0.0	0.0	0.0
Boston Mountains	8.4.6	23	0	0.0	0.0	0.0	0.0	0.0
Southern and Baja California Pine-Oak Mountains	11.1.3	22	1	4.5	4.5	0.0	4.5	0.0
Central Irregular Plains	9.2.4	21	0	0.0	0.0	0.0	4.8	4.8
California Coastal Sage, Chaparral, and Oak Woodlands	11.1.1	21	0	0.0	0.0	0.0	0.0	0.0
Northern Basin and Range	10.1.3	20	0	0.0	0.0	0.0	0.0	0.0
Mississippi Alluvial Plain	8.5.2	19	1	5.3	5.3	0.0	5.3	0.0
Interior River Valleys and Hills	8.3.2	18	0	0.0	0.0	0.0	5.6	5.6
Western Gulf Coastal Plain	9.5.1	16	0	0.0	0.0	0.0	0.0	0.0
Central Basin and Range	10.1.5	16	0	0.0	0.0	0.0	0.0	0.0
Driftless Area	8.1.5	15	0	0.0	0.0	0.0	0.0	0.0
Erie Drift Plain	8.1.10	14	0	0.0	0.0	0.0	7.1	7.1
Eastern Corn Belt Plains	8.2.4	14	0	0.0	0.0	0.0	0.0	0.0
East Central Texas Plains	8.3.8	10	1	10.0	10.0	0.0	10.0	0.0
Southeastern Wisconsin Till Plains	8.2.1	10	0	0.0	0.0	0.0	0.0	0.0
Flint Hills	9.4.4	7	1	14.3	28.6	14.3	28.6	14.3
Central Great Plains	9.4.2	5	0	0.0	0.0	0.0	0.0	0.0
Wyoming Basin	10.1.4	3	0	0.0	0.0	0.0	0.0	0.0
Texas Blackland Prairies	9.4.7	3	0	0.0	0.0	0.0	0.0	0.0
Northern Minnesota Wetlands	5.2.2	2	0	0.0	0.0	0.0	50.0	50.0
Snake River Plain	10.1.8	2	0	0.0	0.0	0.0	0.0	0.0
Central California Valley	11.1.2	2	0	0.0	0.0	0.0	0.0	0.0
Columbia Plateau	10.1.2	2	0	0.0	0.0	0.0	0.0	0.0
Central Corn Belt Plains	8.2.3	2	0	0.0	0.0	0.0	0.0	0.0
Northwestern Glaciated Plains	9.3.1	2	0	0.0	0.0	0.0	0.0	0.0
Colorado Plateaus	10.1.6	1	0	0.0	0.0	0.0	0.0	0.0

Table 5A-20. Percent exceedances of CLs for S only and ANC threshold of 30 µeq/L for deposition years of 2010-12, 2006-08 and 2001-03 in 69 ecoregions.

Ecoregion (n=69)		Sulfur only - ANC = 30 ueq/L								
		Number of CLs			% Exceedances					
		Total Number	CL≤0		2010-2012		2006-2008		2001-2003	
Name	Code		n	%	All	CL>0	All	CL>0	All	CL>0
Northern Appalachian and Atlantic Maritime Highlands	5.3.1	2851	40	1.4	7.5	6.1	18.7	17.3	24.2	22.8
Blue Ridge	8.4.4	1972	13	0.7	10.7	10.0	40.6	40.0	51.1	50.5
Ridge and Valley	8.4.1	1292	8	0.6	11.4	10.8	33.3	32.7	42.4	41.8
Northern Lakes and Forests	5.2.1	839	1	0.1	5.6	5.5	11.3	11.2	20.0	19.9
Northeastern Coastal Zone	8.1.7	565	1	0.2	3.0	2.8	10.6	10.4	12.9	12.7
Piedmont	8.3.4	508	1	0.2	3.3	3.1	14.4	14.2	19.7	19.5
Middle Rockies	6.2.10	496	4	0.8	2.2	1.4	2.4	1.6	2.4	1.6
Acadian Plains and Hills	8.1.8	494	9	1.8	6.9	5.1	13.6	11.7	11.7	9.9
Southeastern Plains	8.3.5	390	9	2.3	11.8	9.5	22.1	19.7	25.1	22.8
Central Appalachians	8.4.2	372	10	2.7	20.7	18.0	48.9	46.2	57.8	55.1
Southern Rockies	6.2.14	372	8	2.2	4.6	2.4	5.4	3.2	5.4	3.2
Sierra Nevada	6.2.12	353	29	8.2	16.4	8.2	16.1	7.9	17.3	9.1
Atlantic Coastal Pine Barrens	8.5.4	234	9	3.8	26.9	23.1	47.4	43.6	55.6	51.7
Northern Piedmont	8.3.1	231	0	0.0	3.0	3.0	7.4	7.4	8.7	8.7
North Central Appalachians	5.3.3	216	0	0.0	7.4	7.4	26.4	26.4	33.3	33.3
Northern Allegheny Plateau	8.1.3	199	1	0.5	2.5	2.0	9.0	8.5	10.6	10.1
Idaho Batholith	6.2.15	188	3	1.6	2.7	1.1	2.7	1.1	2.7	1.1
Cascades	6.2.7	179	11	6.1	7.3	1.1	7.3	1.1	7.8	1.7
North Cascades	6.2.5	162	1	0.6	1.2	0.6	1.2	0.6	1.2	0.6
South Central Plains	8.3.7	153	3	2.0	17.0	15.0	19.6	17.6	20.9	19.0
Southern Coastal Plain	8.5.3	142	4	2.8	31.0	28.2	35.2	32.4	51.4	48.6
Southwestern Appalachians	8.4.9	117	0	0.0	0.9	0.9	30.8	30.8	47.0	47.0
Coast Range	7.1.8	115	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Middle Atlantic Coastal Plain	8.5.1	105	1	1.0	6.7	5.7	21.9	21.0	28.6	27.6
Wasatch and Uinta Mountains	6.2.13	96	0	0.0	0.0	0.0	1.0	1.0	0.0	0.0
North Central Hardwood Forests	8.1.4	94	0	0.0	1.1	1.1	2.1	2.1	3.2	3.2
Columbia Mountains/Northern Rockies	6.2.3	86	3	3.5	3.5	0.0	3.5	0.0	3.5	0.0
Eastern Great Lakes Lowlands	8.1.1	83	1	1.2	1.2	0.0	4.8	3.6	6.0	4.8
Klamath Mountains	6.2.11	81	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Interior Plateau	8.3.3	71	0	0.0	7.0	7.0	8.5	8.5	14.1	14.1
Blue Mountains	6.2.9	63	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Ozark Highlands	8.4.5	56	0	0.0	3.6	3.6	3.6	3.6	5.4	5.4
Ouachita Mountains	8.4.8	42	0	0.0	0.0	0.0	4.8	4.8	4.8	4.8
Mississippi Valley Loess Plains	8.3.6	41	0	0.0	14.6	14.6	19.5	19.5	19.5	19.5
Strait of Georgia/Puget Lowland	7.1.7	38	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0

Western Allegheny Plateau	8.4.3	35	0	0.0	2.9	2.9	20.0	20.0	28.6	28.6
Southern Michigan/Northern Indiana Drift Plains	8.1.6	33	0	0.0	9.1	9.1	15.2	15.2	27.3	27.3
Arkansas Valley	8.4.7	31	0	0.0	3.2	3.2	6.5	6.5	9.7	9.7
Canadian Rockies	6.2.4	31	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Eastern Cascades Slopes and Foothills	6.2.8	27	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Cross Timbers	9.4.5	26	0	0.0	3.8	3.8	7.7	7.7	7.7	7.7
Western Corn Belt Plains	9.2.3	26	0	0.0	3.8	3.8	3.8	3.8	3.8	3.8
Arizona/New Mexico Mountains	13.1.1	25	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Willamette Valley	7.1.9	24	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Boston Mountains	8.4.6	23	0	0.0	0.0	0.0	0.0	0.0	4.3	4.3
Southern and Baja California Pine-Oak Mountains	11.1.3	22	1	4.5	4.5	0.0	4.5	0.0	4.5	0.0
Central Irregular Plains	9.2.4	21	0	0.0	4.8	4.8	9.5	9.5	9.5	9.5
California Coastal Sage, Chaparral, and Oak Woodlands	11.1.1	21	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Northern Basin and Range	10.1.3	20	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Mississippi Alluvial Plain	8.5.2	19	1	5.3	5.3	0.0	15.8	10.5	21.1	15.8
Interior River Valleys and Hills	8.3.2	18	0	0.0	11.1	11.1	22.2	22.2	22.2	22.2
Western Gulf Coastal Plain	9.5.1	16	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Central Basin and Range	10.1.5	16	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Driftless Area	8.1.5	15	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Erie Drift Plain	8.1.10	14	0	0.0	14.3	14.3	21.4	21.4	28.6	28.6
Eastern Corn Belt Plains	8.2.4	14	0	0.0	14.3	14.3	28.6	28.6	28.6	28.6
East Central Texas Plains	8.3.8	10	1	10.0	10.0	0.0	10.0	0.0	10.0	0.0
Southeastern Wisconsin Till Plains	8.2.1	10	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Flint Hills	9.4.4	7	1	14.3	28.6	14.3	28.6	14.3	28.6	14.3
Central Great Plains	9.4.2	5	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Wyoming Basin	10.1.4	3	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Texas Blackland Prairies	9.4.7	3	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Northern Minnesota Wetlands	5.2.2	2	0	0.0	50.0	50.0	50.0	50.0	50.0	50.0
Snake River Plain	10.1.8	2	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Central California Valley	11.1.2	2	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Columbia Plateau	10.1.2	2	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Central Corn Belt Plains	8.2.3	2	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Northwestern Glaciated Plains	9.3.1	2	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Colorado Plateaus	10.1.6	1	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0

Table 5A-21. Percent exceedances of aquatic CLs for S only and ANC threshold of 50 µeq/L for deposition years of 2018-20 and 2014-16 in 69 ecoregions.

Ecoregion (n=69)		Sulfur only - ANC = 50 ueq/L						
		Number of CLs			% Exceedances			
		Total Number	CL≤0		2018-2020		2014-2016	
Name	Code		No.	%	All	CL>0	All	CL>0
Northern Appalachian and Atlantic Maritime Highlands	5.3.1	2851	153	5.4	9.7	4.3	11.9	6.6
Blue Ridge	8.4.4	1972	103	5.2	12.7	7.5	15.1	9.8
Ridge and Valley	8.4.1	1292	28	2.2	5.3	3.1	10.1	8.0
Northern Lakes and Forests	5.2.1	839	11	1.3	4.9	3.6	9.3	8.0
Northeastern Coastal Zone	8.1.7	565	9	1.6	2.8	1.2	3.4	1.8
Piedmont	8.3.4	508	6	1.2	3.5	2.4	4.5	3.3
Middle Rockies	6.2.10	496	16	3.2	4.4	1.2	5.0	1.8
Acadian Plains and Hills	8.1.8	494	29	5.9	7.9	2.0	8.9	3.0
Southeastern Plains	8.3.5	390	21	5.4	12.1	6.7	13.8	8.5
Central Appalachians	8.4.2	372	22	5.9	11.6	5.6	19.6	13.7
Southern Rockies	6.2.14	372	30	8.1	9.4	1.3	11.3	3.2
Sierra Nevada	6.2.12	353	90	25.5	28.3	2.8	30.0	4.5
Atlantic Coastal Pine Barrens	8.5.4	234	28	12.0	24.8	12.8	27.8	15.8
Northern Piedmont	8.3.1	231	3	1.3	1.7	0.4	2.6	1.3
North Central Appalachians	5.3.3	216	5	2.3	4.2	1.9	6.9	4.6
Northern Allegheny Plateau	8.1.3	199	4	2.0	2.5	0.5	2.5	0.5
Idaho Batholith	6.2.15	188	9	4.8	5.9	1.1	9.6	4.8
Cascades	6.2.7	179	21	11.7	13.4	1.7	12.8	1.1
North Cascades	6.2.5	162	4	2.5	3.7	1.2	3.1	0.6
South Central Plains	8.3.7	153	8	5.2	19.0	13.7	19.6	14.4
Southern Coastal Plain	8.5.3	142	12	8.5	29.6	21.1	33.1	24.6
Southwestern Appalachians	8.4.9	117	0	0.0	0.0	0.0	0.9	0.9
Coast Range	7.1.8	115	0	0.0	0.0	0.0	0.0	0.0
Middle Atlantic Coastal Plain	8.5.1	105	3	2.9	5.7	2.9	5.7	2.9
Wasatch and Uinta Mountains	6.2.13	96	1	1.0	1.0	0.0	1.0	0.0
North Central Hardwood Forests	8.1.4	94	1	1.1	2.1	1.1	3.2	2.1
Columbia Mountains/Northern Rockies	6.2.3	86	4	4.7	5.8	1.2	5.8	1.2
Eastern Great Lakes Lowlands	8.1.1	83	1	1.2	2.4	1.2	2.4	1.2
Klamath Mountains	6.2.11	81	0	0.0	0.0	0.0	0.0	0.0
Interior Plateau	8.3.3	71	2	2.8	7.0	4.2	7.0	4.2
Blue Mountains	6.2.9	63	0	0.0	0.0	0.0	1.6	1.6
Ozark Highlands	8.4.5	56	1	1.8	3.6	1.8	3.6	1.8
Ouachita Mountains	8.4.8	42	0	0.0	2.4	2.4	2.4	2.4
Mississippi Valley Loess Plains	8.3.6	41	1	2.4	19.5	17.1	19.5	17.1
Strait of Georgia/Puget Lowland	7.1.7	38	1	2.6	2.6	0.0	5.3	2.6

Ecoregion (n=69)		Sulfur only - ANC = 50 ueq/L						
		Number of CLs			% Exceedances			
		Total Number	CL≤0		2018-2020		2014-2016	
Name	Code		No.	%	All	CL>0	All	CL>0
Western Allegheny Plateau	8.4.3	35	0	0.0	0.0	0.0	2.9	2.9
Southern Michigan/Northern Indiana Drift Plains	8.1.6	33	1	3.0	6.1	3.0	9.1	6.1
Arkansas Valley	8.4.7	31	0	0.0	6.5	6.5	3.2	3.2
Canadian Rockies	6.2.4	31	1	3.2	3.2	0.0	3.2	0.0
Eastern Cascades Slopes and Foothills	6.2.8	27	0	0.0	0.0	0.0	3.7	3.7
Cross Timbers	9.4.5	26	2	7.7	11.5	3.8	11.5	3.8
Western Corn Belt Plains	9.2.3	26	1	3.8	3.8	0.0	3.8	0.0
Arizona/New Mexico Mountains	13.1.1	25	0	0.0	0.0	0.0	0.0	0.0
Willamette Valley	7.1.9	24	0	0.0	0.0	0.0	0.0	0.0
Boston Mountains	8.4.6	23	0	0.0	0.0	0.0	0.0	0.0
Southern and Baja California Pine-Oak Mountains	11.1.3	22	1	4.5	4.5	0.0	9.1	4.5
Central Irregular Plains	9.2.4	21	1	4.8	9.5	4.8	14.3	9.5
California Coastal Sage, Chaparral, and Oak Woodlands	11.1.1	21	0	0.0	0.0	0.0	0.0	0.0
Northern Basin and Range	10.1.3	20	1	5.0	5.0	0.0	5.0	0.0
Mississippi Alluvial Plain	8.5.2	19	1	5.3	15.8	10.5	15.8	10.5
Interior River Valleys and Hills	8.3.2	18	0	0.0	11.1	11.1	11.1	11.1
Western Gulf Coastal Plain	9.5.1	16	0	0.0	0.0	0.0	0.0	0.0
Central Basin and Range	10.1.5	16	0	0.0	0.0	0.0	0.0	0.0
Driftless Area	8.1.5	15	0	0.0	0.0	0.0	0.0	0.0
Erie Drift Plain	8.1.10	14	0	0.0	7.1	7.1	7.1	7.1
Eastern Corn Belt Plains	8.2.4	14	0	0.0	14.3	14.3	14.3	14.3
East Central Texas Plains	8.3.8	10	1	10.0	10.0	0.0	10.0	0.0
Southeastern Wisconsin Till Plains	8.2.1	10	0	0.0	0.0	0.0	0.0	0.0
Flint Hills	9.4.4	7	2	28.6	28.6	0.0	28.6	0.0
Central Great Plains	9.4.2	5	0	0.0	0.0	0.0	0.0	0.0
Wyoming Basin	10.1.4	3	0	0.0	0.0	0.0	0.0	0.0
Texas Blackland Prairies	9.4.7	3	0	0.0	0.0	0.0	0.0	0.0
Northern Minnesota Wetlands	5.2.2	2	0	0.0	50.0	50.0	50.0	50.0
Snake River Plain	10.1.8	2	0	0.0	0.0	0.0	0.0	0.0
Central California Valley	11.1.2	2	0	0.0	0.0	0.0	0.0	0.0
Columbia Plateau	10.1.2	2	0	0.0	0.0	0.0	0.0	0.0
Central Corn Belt Plains	8.2.3	2	0	0.0	0.0	0.0	0.0	0.0
Northwestern Glaciated Plains	9.3.1	2	0	0.0	0.0	0.0	0.0	0.0
Colorado Plateaus	10.1.6	1	0	0.0	0.0	0.0	0.0	0.0

Table 5A-22. Percent exceedances of CLs for S only and ANC threshold of 50 µeq/L for deposition years of 2010-12, 2006-08 and 2001-03 in 69 ecoregions.

Ecoregion (n=69)		Sulfur only - ANC = 50 ueq/L								
		Number of CLs			% Exceedances					
		Total Number	CL≤0		2010-2012		2006-2008		2001-2003	
Name	Code		No.	%	All	CL>0	All	CL>0	All	CL>0
Northern Appalachian and Atlantic Maritime Highlands	5.3.1	2851	153	5	15.0	9.7	28.0	22.6	32.0	26.6
Blue Ridge	8.4.4	1972	103	5	26.6	21.4	55.5	50.3	63.1	57.9
Ridge and Valley	8.4.1	1292	28	2	17.3	15.2	40.3	38.2	48.5	46.3
Northern Lakes and Forests	5.2.1	839	11	1	13.6	12.3	20.0	18.7	26.7	25.4
Northeastern Coastal Zone	8.1.7	565	9	2	5.3	3.7	15.0	13.5	16.5	14.9
Piedmont	8.3.4	508	6	1	6.9	5.7	20.9	19.7	25.0	23.8
Middle Rockies	6.2.10	496	16	3	5.6	2.4	6.3	3.0	5.8	2.6
Acadian Plains and Hills	8.1.8	494	29	6	10.9	5.1	18.6	12.8	17.6	11.7
Southeastern Plains	8.3.5	390	21	5	16.2	10.8	25.4	20.0	29.5	24.1
Central Appalachians	8.4.2	372	22	6	33.6	27.7	55.1	49.2	63.2	57.3
Southern Rockies	6.2.14	372	30	8	12.1	4.0	12.9	4.8	12.9	4.8
Sierra Nevada	6.2.12	353	90	25	30.0	4.5	30.0	4.5	30.0	4.5
Atlantic Coastal Pine Barrens	8.5.4	234	28	12	33.8	21.8	53.0	41.0	61.1	49.1
Northern Piedmont	8.3.1	231	3	1	3.5	2.2	8.2	6.9	10.4	9.1
North Central Appalachians	5.3.3	216	5	2	14.4	12.0	34.3	31.9	43.1	40.7
Northern Allegheny Plateau	8.1.3	199	4	2	4.0	2.0	11.6	9.5	14.6	12.6
Idaho Batholith	6.2.15	188	9	5	10.1	5.3	9.6	4.8	7.4	2.7
Cascades	6.2.7	179	21	12	12.8	1.1	14.0	2.2	13.4	1.7
North Cascades	6.2.5	162	4	2	3.7	1.2	3.7	1.2	3.7	1.2
South Central Plains	8.3.7	153	8	5	19.6	14.4	24.2	19.0	24.8	19.6
Southern Coastal Plain	8.5.3	142	12	8	33.8	25.4	40.8	32.4	53.5	45.1
Southwestern Appalachians	8.4.9	117	0	0	2.6	2.6	39.3	39.3	53.8	53.8
Coast Range	7.1.8	115	0	0	0.0	0.0	0.0	0.0	0.0	0.0
Middle Atlantic Coastal Plain	8.5.1	105	3	3	9.5	6.7	26.7	23.8	29.5	26.7
Wasatch and Uinta Mountains	6.2.13	96	1	1	1.0	0.0	1.0	0.0	1.0	0.0
North Central Hardwood Forests	8.1.4	94	1	1	4.3	3.2	6.4	5.3	10.6	9.6
Columbia Mountains/Northern Rockies	6.2.3	86	4	5	5.8	1.2	5.8	1.2	5.8	1.2
Eastern Great Lakes Lowlands	8.1.1	83	1	1	2.4	1.2	6.0	4.8	6.0	4.8
Klamath Mountains	6.2.11	81	0	0	0.0	0.0	0.0	0.0	0.0	0.0
Interior Plateau	8.3.3	71	2	3	8.5	5.6	11.3	8.5	15.5	12.7
Blue Mountains	6.2.9	63	0	0	0.0	0.0	0.0	0.0	0.0	0.0
Ozark Highlands	8.4.5	56	1	2	5.4	3.6	5.4	3.6	5.4	3.6
Ouachita Mountains	8.4.8	42	0	0	7.1	7.1	11.9	11.9	11.9	11.9
Mississippi Valley Loess Plains	8.3.6	41	1	2	19.5	17.1	19.5	17.1	22.0	19.5
Strait of Georgia/Puget Lowland	7.1.7	38	1	3	2.6	0.0	2.6	0.0	5.3	2.6

Ecoregion (n=69)		Sulfur only - ANC = 50 ueq/L								
		Number of CLs			% Exceedances					
		Total Number	CL≤0		2010-2012		2006-2008		2001-2003	
Name	Code		No.	%	All	CL>0	All	CL>0	All	CL>0
Western Allegheny Plateau	8.4.3	35	0	0	11.4	11.4	25.7	25.7	37.1	37.1
Southern Michigan/Northern Indiana Drift Plains	8.1.6	33	1	3	15.2	12.1	27.3	24.2	27.3	24.2
Arkansas Valley	8.4.7	31	0	0	6.5	6.5	6.5	6.5	9.7	9.7
Canadian Rockies	6.2.4	31	1	3	3.2	0.0	3.2	0.0	3.2	0.0
Eastern Cascades Slopes and Foothills	6.2.8	27	0	0	0.0	0.0	0.0	0.0	0.0	0.0
Cross Timbers	9.4.5	26	2	8	11.5	3.8	15.4	7.7	19.2	11.5
Western Corn Belt Plains	9.2.3	26	1	4	3.8	0.0	15.4	11.5	15.4	11.5
Arizona/New Mexico Mountains	13.1.1	25	0	0	0.0	0.0	0.0	0.0	0.0	0.0
Willamette Valley	7.1.9	24	0	0	0.0	0.0	0.0	0.0	0.0	0.0
Boston Mountains	8.4.6	23	0	0	4.3	4.3	8.7	8.7	8.7	8.7
Southern and Baja California Pine-Oak Mountains	11.1.3	22	1	5	9.1	4.5	9.1	4.5	9.1	4.5
Central Irregular Plains	9.2.4	21	1	5	14.3	9.5	14.3	9.5	14.3	9.5
California Coastal Sage, Chaparral, and Oak Woodlands	11.1.1	21	0	0	0.0	0.0	0.0	0.0	0.0	0.0
Northern Basin and Range	10.1.3	20	1	5	5.0	0.0	5.0	0.0	5.0	0.0
Mississippi Alluvial Plain	8.5.2	19	1	5	15.8	10.5	26.3	21.1	26.3	21.1
Interior River Valleys and Hills	8.3.2	18	0	0	11.1	11.1	22.2	22.2	22.2	22.2
Western Gulf Coastal Plain	9.5.1	16	0	0	0.0	0.0	0.0	0.0	0.0	0.0
Central Basin and Range	10.1.5	16	0	0	0.0	0.0	0.0	0.0	0.0	0.0
Driftless Area	8.1.5	15	0	0	0.0	0.0	0.0	0.0	0.0	0.0
Erie Drift Plain	8.1.10	14	0	0	14.3	14.3	28.6	28.6	35.7	35.7
Eastern Corn Belt Plains	8.2.4	14	0	0	28.6	28.6	28.6	28.6	28.6	28.6
East Central Texas Plains	8.3.8	10	1	10	10.0	0.0	20.0	10.0	20.0	10.0
Southeastern Wisconsin Till Plains	8.2.1	10	0	0	0.0	0.0	0.0	0.0	0.0	0.0
Flint Hills	9.4.4	7	2	29	28.6	0.0	28.6	0.0	28.6	0.0
Central Great Plains	9.4.2	5	0	0	0.0	0.0	0.0	0.0	0.0	0.0
Wyoming Basin	10.1.4	3	0	0	0.0	0.0	0.0	0.0	0.0	0.0
Texas Blackland Prairies	9.4.7	3	0	0	0.0	0.0	0.0	0.0	0.0	0.0
Northern Minnesota Wetlands	5.2.2	2	0	0	50.0	50.0	50.0	50.0	50.0	50.0
Snake River Plain	10.1.8	2	0	0	0.0	0.0	0.0	0.0	0.0	0.0
Central California Valley	11.1.2	2	0	0	0.0	0.0	0.0	0.0	0.0	0.0
Columbia Plateau	10.1.2	2	0	0	0.0	0.0	0.0	0.0	0.0	0.0
Central Corn Belt Plains	8.2.3	2	0	0	0.0	0.0	0.0	0.0	0.0	0.0
Northwestern Glaciated Plains	9.3.1	2	0	0	0.0	0.0	0.0	0.0	0.0	0.0
Colorado Plateaus	10.1.6	1	0	0	0.0	0.0	0.0	0.0	0.0	0.0

Table 5A-23. Percent exceedances of aquatic CLs for S only and ANC threshold of 50/20 µeq/L for deposition years of 2018-20 and 2014-16 in 69 ecoregions.

Ecoregion (n=69)		Sulfur only - ANC = 50/20 ueq/L						
		Number of CLs			% Exceedances			
		Total Number	CL≤0		2018-2020		2014-2016	
Name	Code		n	%	All	CL>0	All	CL>0
Northern Appalachian and Atlantic Maritime Highlands	5.3.1	2851	153	5.4	9.7	4.3	11.9	6.6
Blue Ridge	8.4.4	1972	103	5.2	12.7	7.5	15.1	9.8
Ridge and Valley	8.4.1	1292	28	2.2	5.3	3.1	10.1	8.0
Northern Lakes and Forests	5.2.1	839	11	1.3	4.9	3.6	9.3	8.0
Northeastern Coastal Zone	8.1.7	565	9	1.6	2.8	1.2	3.4	1.8
Piedmont	8.3.4	508	6	1.2	3.5	2.4	4.5	3.3
Middle Rockies	6.2.10	496	0	0.0	0.2	0.2	0.6	0.6
Acadian Plains and Hills	8.1.8	494	29	5.9	7.9	2.0	8.9	3.0
Southeastern Plains	8.3.5	390	21	5.4	12.1	6.7	13.8	8.5
Central Appalachians	8.4.2	372	22	5.9	11.6	5.6	19.6	13.7
Southern Rockies	6.2.14	372	1	0.3	0.5	0.3	1.9	1.6
Sierra Nevada	6.2.12	353	11	3.1	4.2	1.1	5.9	2.8
Atlantic Coastal Pine Barrens	8.5.4	234	28	12.0	24.8	12.8	27.8	15.8
Northern Piedmont	8.3.1	231	3	1.3	1.7	0.4	2.6	1.3
North Central Appalachians	5.3.3	216	5	2.3	4.2	1.9	6.9	4.6
Northern Allegheny Plateau	8.1.3	199	4	2.0	2.5	0.5	2.5	0.5
Idaho Batholith	6.2.15	188	1	0.5	0.5	0.0	0.5	0.0
Cascades	6.2.7	179	4	2.2	3.9	1.7	5.0	2.8
North Cascades	6.2.5	162	1	0.6	0.6	0.0	0.6	0.0
South Central Plains	8.3.7	153	8	5.2	19.0	13.7	19.6	14.4
Southern Coastal Plain	8.5.3	142	12	8.5	29.6	21.1	33.1	24.6
Southwestern Appalachians	8.4.9	117	0	0.0	0.0	0.0	0.9	0.9
Coast Range	7.1.8	115	0	0.0	0.0	0.0	0.0	0.0
Middle Atlantic Coastal Plain	8.5.1	105	3	2.9	5.7	2.9	5.7	2.9
Wasatch and Uinta Mountains	6.2.13	96	0	0.0	0.0	0.0	0.0	0.0
North Central Hardwood Forests	8.1.4	94	1	1.1	2.1	1.1	3.2	2.1
Columbia Mountains/Northern Rockies	6.2.3	86	1	1.2	1.2	0.0	2.3	1.2
Eastern Great Lakes Lowlands	8.1.1	83	1	1.2	2.4	1.2	2.4	1.2
Klamath Mountains	6.2.11	81	0	0.0	0.0	0.0	0.0	0.0
Interior Plateau	8.3.3	71	2	2.8	7.0	4.2	7.0	4.2
Blue Mountains	6.2.9	63	0	0.0	0.0	0.0	0.0	0.0
Ozark Highlands	8.4.5	56	1	1.8	3.6	1.8	3.6	1.8
Ouachita Mountains	8.4.8	42	0	0.0	2.4	2.4	2.4	2.4
Mississippi Valley Loess Plains	8.3.6	41	1	2.4	19.5	17.1	19.5	17.1
Strait of Georgia/Puget Lowland	7.1.7	38	0	0.0	0.0	0.0	0.0	0.0
Western Allegheny Plateau	8.4.3	35	0	0.0	0.0	0.0	2.9	2.9

Ecoregion (n=69)		Sulfur only - ANC = 50/20 ueq/L						
		Number of CLs			% Exceedances			
		Total	CL≤0		2018-2020		2014-2016	
Name	Code	Number	n	%	All	CL>0	All	CL>0
Southern Michigan/Northern Indiana Drift Plains	8.1.6	33	1	3.0	6.1	3.0	9.1	6.1
Arkansas Valley	8.4.7	31	0	0.0	6.5	6.5	3.2	3.2
Canadian Rockies	6.2.4	31	0	0.0	0.0	0.0	0.0	0.0
Eastern Cascades Slopes and Foothills	6.2.8	27	0	0.0	0.0	0.0	0.0	0.0
Cross Timbers	9.4.5	26	2	7.7	11.5	3.8	11.5	3.8
Western Corn Belt Plains	9.2.3	26	1	3.8	3.8	0.0	3.8	0.0
Arizona/New Mexico Mountains	13.1.1	25	0	0.0	0.0	0.0	0.0	0.0
Willamette Valley	7.1.9	24	0	0.0	0.0	0.0	0.0	0.0
Boston Mountains	8.4.6	23	0	0.0	0.0	0.0	0.0	0.0
Southern and Baja California Pine-Oak Mountains	11.1.3	22	0	0.0	0.0	0.0	4.5	4.5
Central Irregular Plains	9.2.4	21	1	4.8	9.5	4.8	14.3	9.5
California Coastal Sage, Chaparral, and Oak Woodlands	11.1.1	21	0	0.0	0.0	0.0	0.0	0.0
Northern Basin and Range	10.1.3	20	0	0.0	0.0	0.0	0.0	0.0
Mississippi Alluvial Plain	8.5.2	19	1	5.3	15.8	10.5	15.8	10.5
Interior River Valleys and Hills	8.3.2	18	0	0.0	11.1	11.1	11.1	11.1
Western Gulf Coastal Plain	9.5.1	16	0	0.0	0.0	0.0	0.0	0.0
Central Basin and Range	10.1.5	16	0	0.0	0.0	0.0	0.0	0.0
Driftless Area	8.1.5	15	0	0.0	0.0	0.0	0.0	0.0
Erie Drift Plain	8.1.10	14	0	0.0	7.1	7.1	7.1	7.1
Eastern Corn Belt Plains	8.2.4	14	0	0.0	14.3	14.3	14.3	14.3
East Central Texas Plains	8.3.8	10	1	10.0	10.0	0.0	10.0	0.0
Southeastern Wisconsin Till Plains	8.2.1	10	0	0.0	0.0	0.0	0.0	0.0
Flint Hills	9.4.4	7	2	28.6	28.6	0.0	28.6	0.0
Central Great Plains	9.4.2	5	0	0.0	0.0	0.0	0.0	0.0
Wyoming Basin	10.1.4	3	0	0.0	0.0	0.0	0.0	0.0
Texas Blackland Prairies	9.4.7	3	0	0.0	0.0	0.0	0.0	0.0
Northern Minnesota Wetlands	5.2.2	2	0	0.0	50.0	50.0	50.0	50.0
Snake River Plain	10.1.8	2	0	0.0	0.0	0.0	0.0	0.0
Central California Valley	11.1.2	2	0	0.0	0.0	0.0	0.0	0.0
Columbia Plateau	10.1.2	2	0	0.0	0.0	0.0	0.0	0.0
Central Corn Belt Plains	8.2.3	2	0	0.0	0.0	0.0	0.0	0.0
Northwestern Glaciated Plains	9.3.1	2	0	0.0	0.0	0.0	0.0	0.0
Colorado Plateaus	10.1.6	1	0	0.0	0.0	0.0	0.0	0.0

Table 5A-24. Percent exceedances of CLs for S only and ANC threshold of 50/20 µeq/L for deposition years of 2010-12, 2006-08 and 2001-03 in 69 ecoregions.

Ecoregion (n=69)		Sulfur only - ANC = 50/20 ueq/L								
		Number of CLs			% Exceedances					
		Total Number	CL≤0		2010-2012		2006-2008		2001-2003	
Name	Code		No.	%	All	CL>0	All	CL>0	All	CL>0
Northern Appalachian and Atlantic Maritime Highlands	5.3.1	2851	153	5.4	15.0	9.7	28.0	22.6	32.0	26.6
Blue Ridge	8.4.4	1972	103	5.2	26.6	21.4	55.5	50.3	63.1	57.9
Ridge and Valley	8.4.1	1292	28	2.2	17.3	15.2	40.3	38.2	48.5	46.3
Northern Lakes and Forests	5.2.1	839	11	1.3	13.6	12.3	20.0	18.7	26.7	25.4
Northeastern Coastal Zone	8.1.7	565	9	1.6	5.3	3.7	15.0	13.5	16.5	14.9
Piedmont	8.3.4	508	6	1.2	6.9	5.7	20.9	19.7	25.0	23.8
Middle Rockies	6.2.10	496	0	0.0	0.8	0.8	0.8	0.8	0.8	0.8
Acadian Plains and Hills	8.1.8	494	29	5.9	10.9	5.1	18.6	12.8	17.6	11.7
Southeastern Plains	8.3.5	390	21	5.4	16.2	10.8	25.4	20.0	29.5	24.1
Central Appalachians	8.4.2	372	22	5.9	33.6	27.7	55.1	49.2	63.2	57.3
Southern Rockies	6.2.14	372	1	0.3	2.2	1.9	2.7	2.4	2.7	2.4
Sierra Nevada	6.2.12	353	11	3.1	5.1	2.0	5.1	2.0	6.2	3.1
Atlantic Coastal Pine Barrens	8.5.4	234	28	12.0	33.8	21.8	53.0	41.0	61.1	49.1
Northern Piedmont	8.3.1	231	3	1.3	3.5	2.2	8.2	6.9	10.4	9.1
North Central Appalachians	5.3.3	216	5	2.3	14.4	12.0	34.3	31.9	43.1	40.7
Northern Allegheny Plateau	8.1.3	199	4	2.0	4.0	2.0	11.6	9.5	14.6	12.6
Idaho Batholith	6.2.15	188	1	0.5	0.5	0.0	0.5	0.0	0.5	0.0
Cascades	6.2.7	179	4	2.2	3.9	1.7	3.9	1.7	3.9	1.7
North Cascades	6.2.5	162	1	0.6	0.6	0.0	0.6	0.0	0.6	0.0
South Central Plains	8.3.7	153	8	5.2	19.6	14.4	24.2	19.0	24.8	19.6
Southern Coastal Plain	8.5.3	142	12	8.5	33.8	25.4	40.8	32.4	53.5	45.1
Southwestern Appalachians	8.4.9	117	0	0.0	2.6	2.6	39.3	39.3	53.8	53.8
Coast Range	7.1.8	115	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Middle Atlantic Coastal Plain	8.5.1	105	3	2.9	9.5	6.7	26.7	23.8	29.5	26.7
Wasatch and Uinta Mountains	6.2.13	96	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
North Central Hardwood Forests	8.1.4	94	1	1.1	4.3	3.2	6.4	5.3	10.6	9.6
Columbia Mountains/Northern Rockies	6.2.3	86	1	1.2	3.5	2.3	3.5	2.3	3.5	2.3
Eastern Great Lakes Lowlands	8.1.1	83	1	1.2	2.4	1.2	6.0	4.8	6.0	4.8
Klamath Mountains	6.2.11	81	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Interior Plateau	8.3.3	71	2	2.8	8.5	5.6	11.3	8.5	15.5	12.7
Blue Mountains	6.2.9	63	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Ozark Highlands	8.4.5	56	1	1.8	5.4	3.6	5.4	3.6	5.4	3.6
Ouachita Mountains	8.4.8	42	0	0.0	7.1	7.1	11.9	11.9	11.9	11.9
Mississippi Valley Loess Plains	8.3.6	41	1	2.4	19.5	17.1	19.5	17.1	22.0	19.5
Strait of Georgia/Puget Lowland	7.1.7	38	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0

Ecoregion (n=69)		Sulfur only - ANC = 50/20 ueq/L								
		Number of CLs			% Exceedances					
		Total Number	CL≤0		2010-2012		2006-2008		2001-2003	
Name	Code		No.	%	All	CL>0	All	CL>0	All	CL>0
Western Allegheny Plateau	8.4.3	35	0	0.0	11.4	11.4	25.7	25.7	37.1	37.1
Southern Michigan/Northern Indiana Drift Plains	8.1.6	33	1	3.0	15.2	12.1	27.3	24.2	27.3	24.2
Arkansas Valley	8.4.7	31	0	0.0	6.5	6.5	6.5	6.5	9.7	9.7
Canadian Rockies	6.2.4	31	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Eastern Cascades Slopes and Foothills	6.2.8	27	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Cross Timbers	9.4.5	26	2	7.7	11.5	3.8	15.4	7.7	19.2	11.5
Western Corn Belt Plains	9.2.3	26	1	3.8	3.8	0.0	15.4	11.5	15.4	11.5
Arizona/New Mexico Mountains	13.1.1	25	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Willamette Valley	7.1.9	24	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Boston Mountains	8.4.6	23	0	0.0	4.3	4.3	8.7	8.7	8.7	8.7
Southern and Baja California Pine-Oak Mountains	11.1.3	22	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Central Irregular Plains	9.2.4	21	1	4.8	14.3	9.5	14.3	9.5	14.3	9.5
California Coastal Sage, Chaparral, and Oak Woodlands	11.1.1	21	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Northern Basin and Range	10.1.3	20	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Mississippi Alluvial Plain	8.5.2	19	1	5.3	15.8	10.5	26.3	21.1	26.3	21.1
Interior River Valleys and Hills	8.3.2	18	0	0.0	11.1	11.1	22.2	22.2	22.2	22.2
Western Gulf Coastal Plain	9.5.1	16	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Central Basin and Range	10.1.5	16	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Driftless Area	8.1.5	15	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Erie Drift Plain	8.1.10	14	0	0.0	14.3	14.3	28.6	28.6	35.7	35.7
Eastern Corn Belt Plains	8.2.4	14	0	0.0	28.6	28.6	28.6	28.6	28.6	28.6
East Central Texas Plains	8.3.8	10	1	10.0	10.0	0.0	20.0	10.0	20.0	10.0
Southeastern Wisconsin Till Plains	8.2.1	10	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Flint Hills	9.4.4	7	2	28.6	28.6	0.0	28.6	0.0	28.6	0.0
Central Great Plains	9.4.2	5	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Wyoming Basin	10.1.4	3	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Texas Blackland Prairies	9.4.7	3	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Northern Minnesota Wetlands	5.2.2	2	0	0.0	50.0	50.0	50.0	50.0	50.0	50.0
Snake River Plain	10.1.8	2	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Central California Valley	11.1.2	2	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Columbia Plateau	10.1.2	2	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Central Corn Belt Plains	8.2.3	2	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Northwestern Glaciated Plains	9.3.1	2	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Colorado Plateaus	10.1.6	1	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0

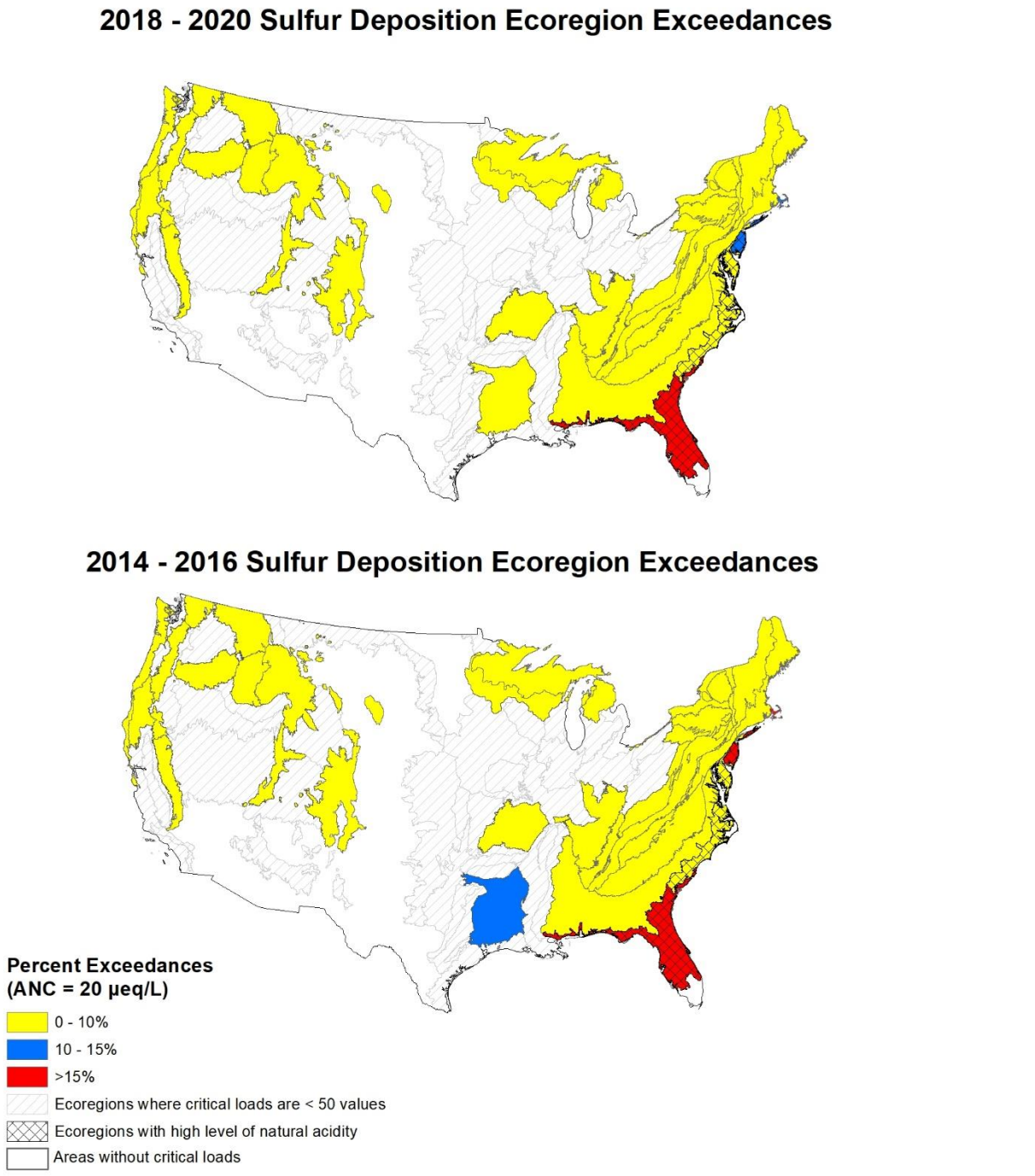

Figure 5A-32. Percent of CLs exceeded per ecoregion for S only deposition from 2018-20 (top) and 2014-16 (bottom) for an ANC threshold of 20 µeq/L. The Southern Coastal Plan (8.5.3) and Atlantic Coastal Pine Barrens (8.5.4) ecoregions are cross hatched to indicate natural high level of acidity.

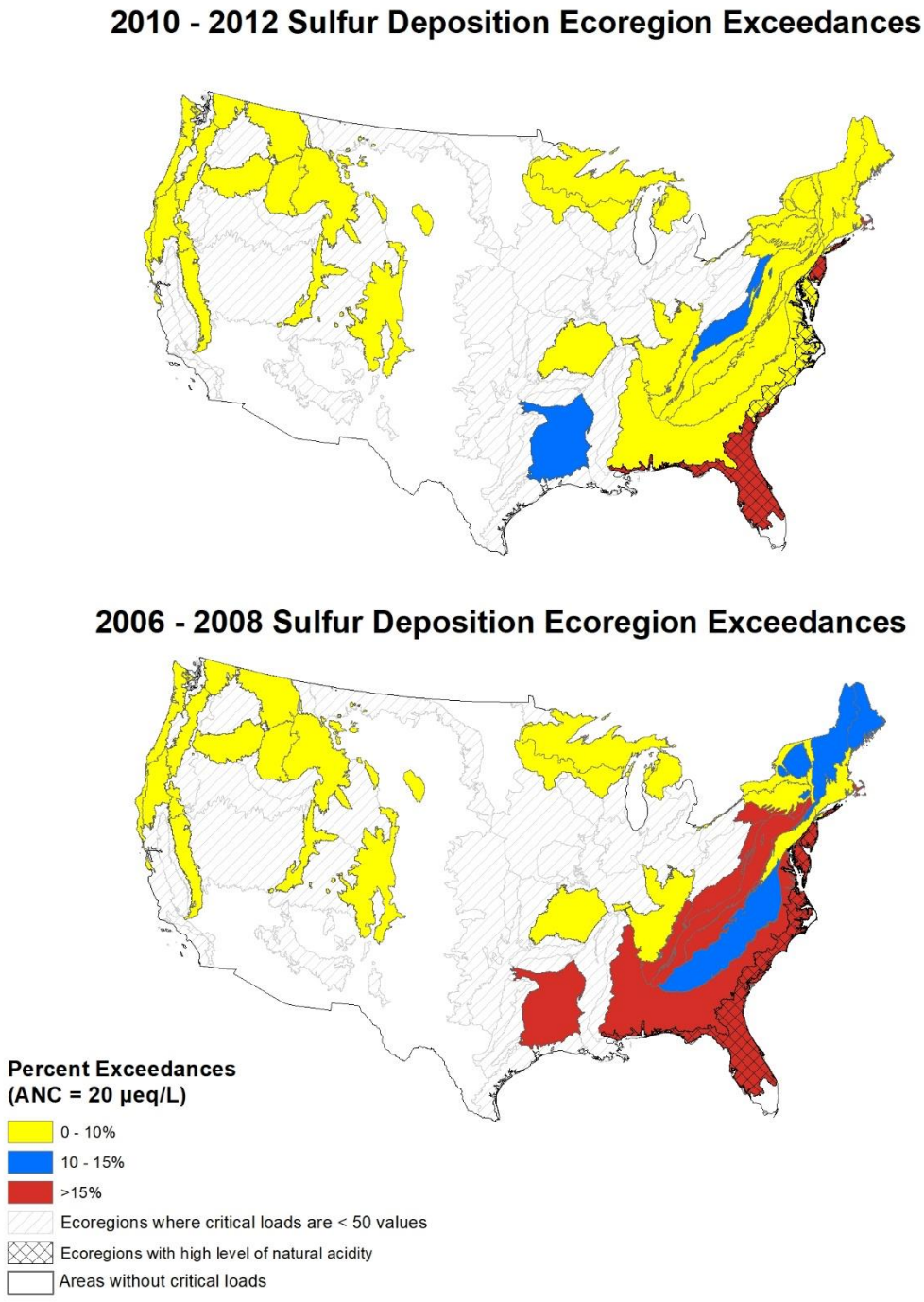

Figure 5A-33. Percent of CLs exceeded per ecoregion for S only deposition from 2010-12 (top) and 2006-08 (bottom) for an ANC threshold of 20 µeq/L. The Southern Coastal Plan (8.5.3) and Atlantic Coastal Pine Barrens (8.5.4) ecoregions are cross hatched to indicate natural high level of acidity.

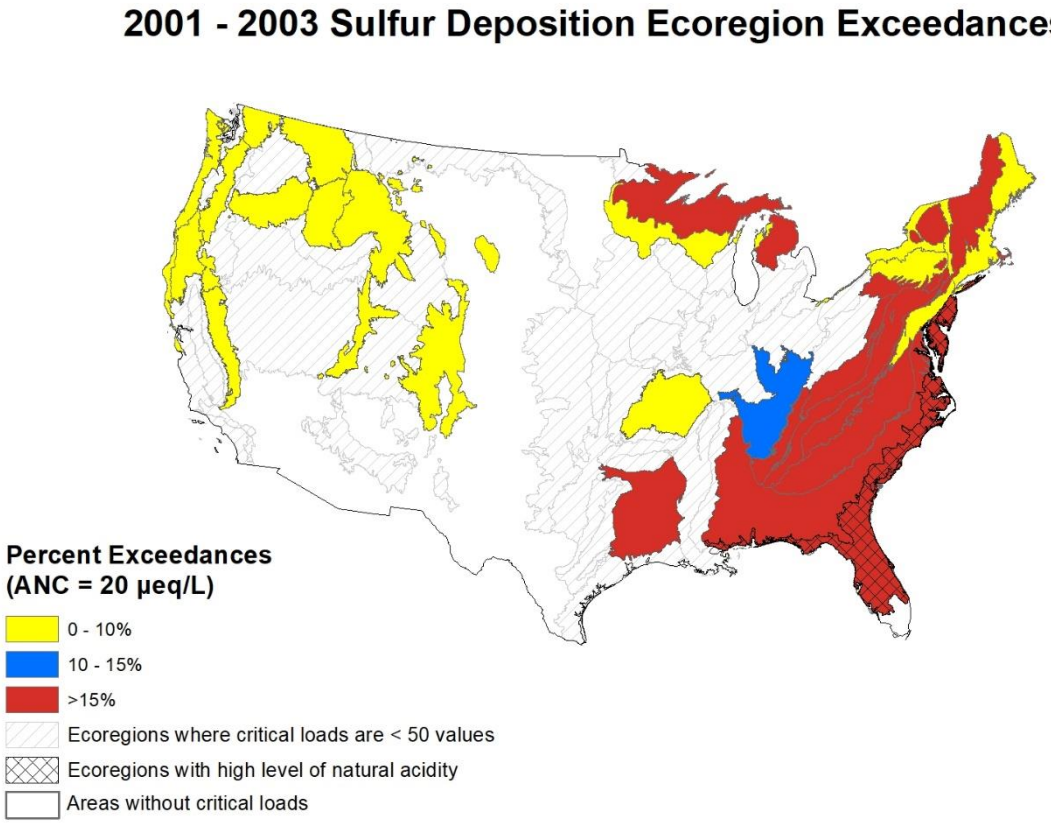

Figure 5A-34. Percent of CLs exceeded per ecoregion for S only deposition from 2001-02 for an ANC threshold of 20 µeq/L. The Southern Coastal Plan (8.5.3) and Atlantic Coastal Pine Barrens (8.5.4) ecoregions are cross hatched to indicate natural high level of acidity.

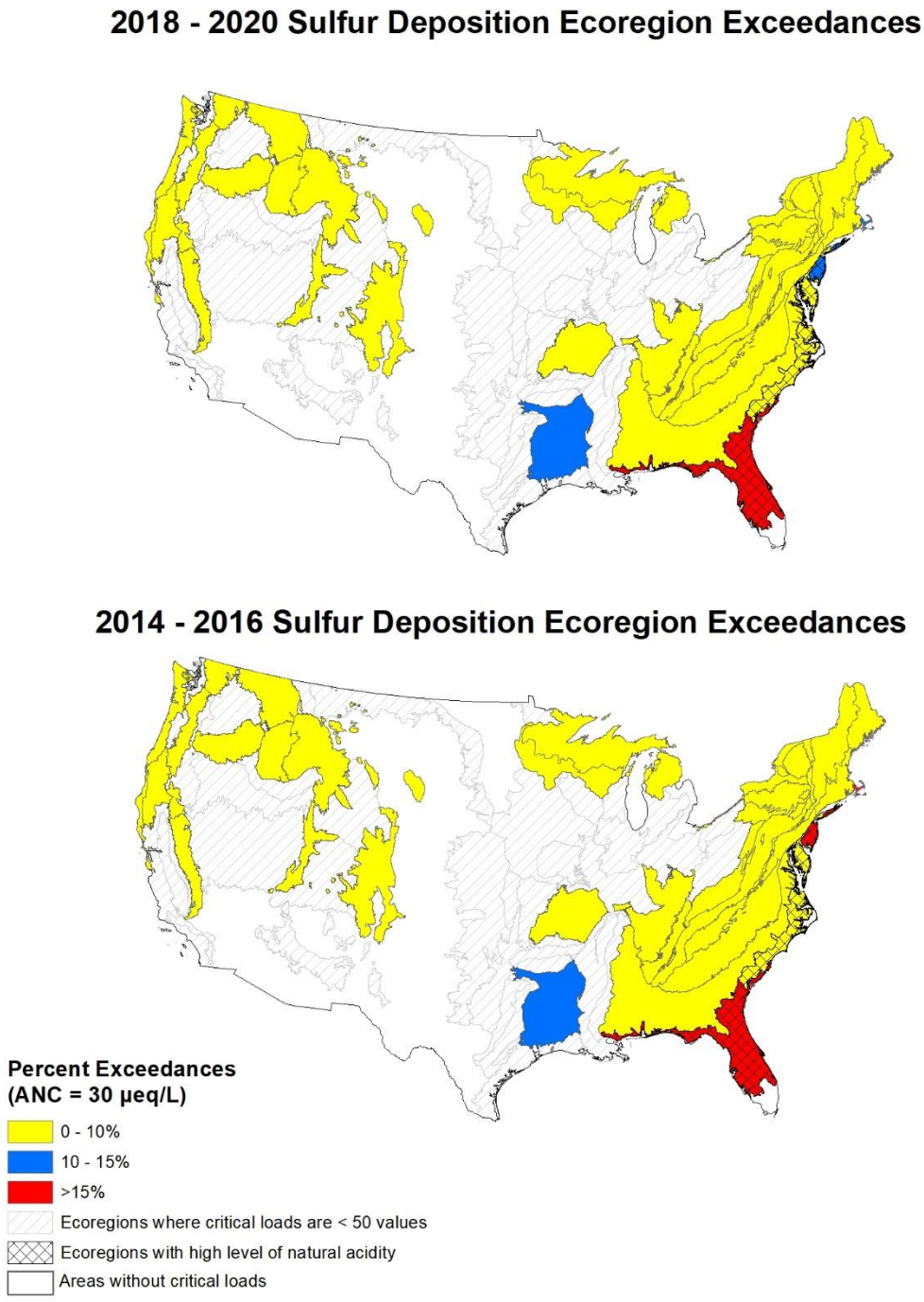

Figure 5A-35. Percent of CLs exceeded per ecoregion for S only deposition from 2018-20 (top) and 2014-16 (bottom) for an ANC threshold of 30 µeq/L. The Southern Coastal Plan (8.5.3) and Atlantic Coastal Pine Barrens (8.5.4) ecoregions are cross hatched to indicate natural high level of acidity.

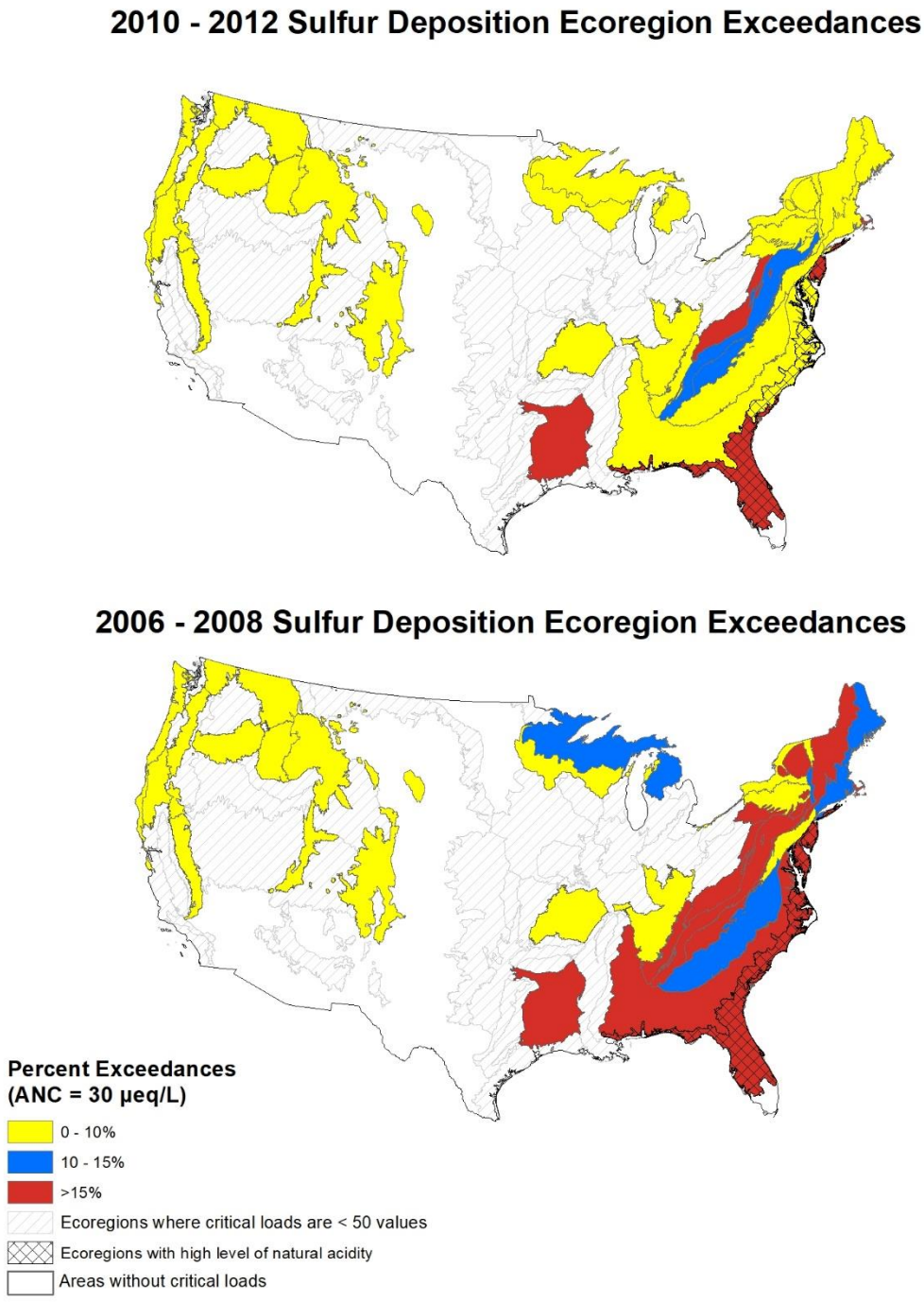

Figure 5A-36. Percent of CLs exceeded per ecoregion for S only deposition from 2010-12 (top) and 2006-08 (bottom) for an ANC threshold of 30 µeq/L. The Southern Coastal Plan (8.5.3) and Atlantic Coastal Pine Barrens (8.5.4) ecoregions are cross hatched to indicate natural high level of acidity.

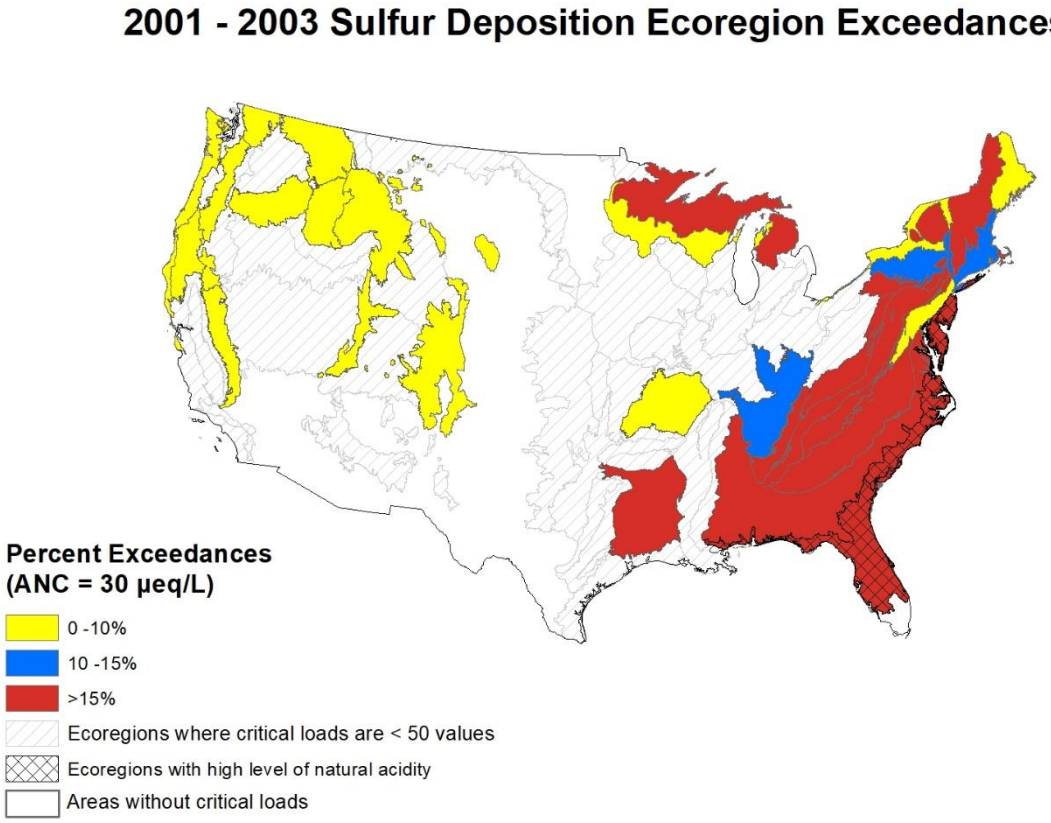

Figure 5A-37. Percent of CLs exceeded per ecoregion for S only deposition from 2001-03 for an ANC threshold of 30 µeq/L. The Southern Coastal Plan (8.5.3) and Atlantic Coastal Pine Barrens (8.5.4) ecoregions are cross hatched to indicate natural high level of acidity.

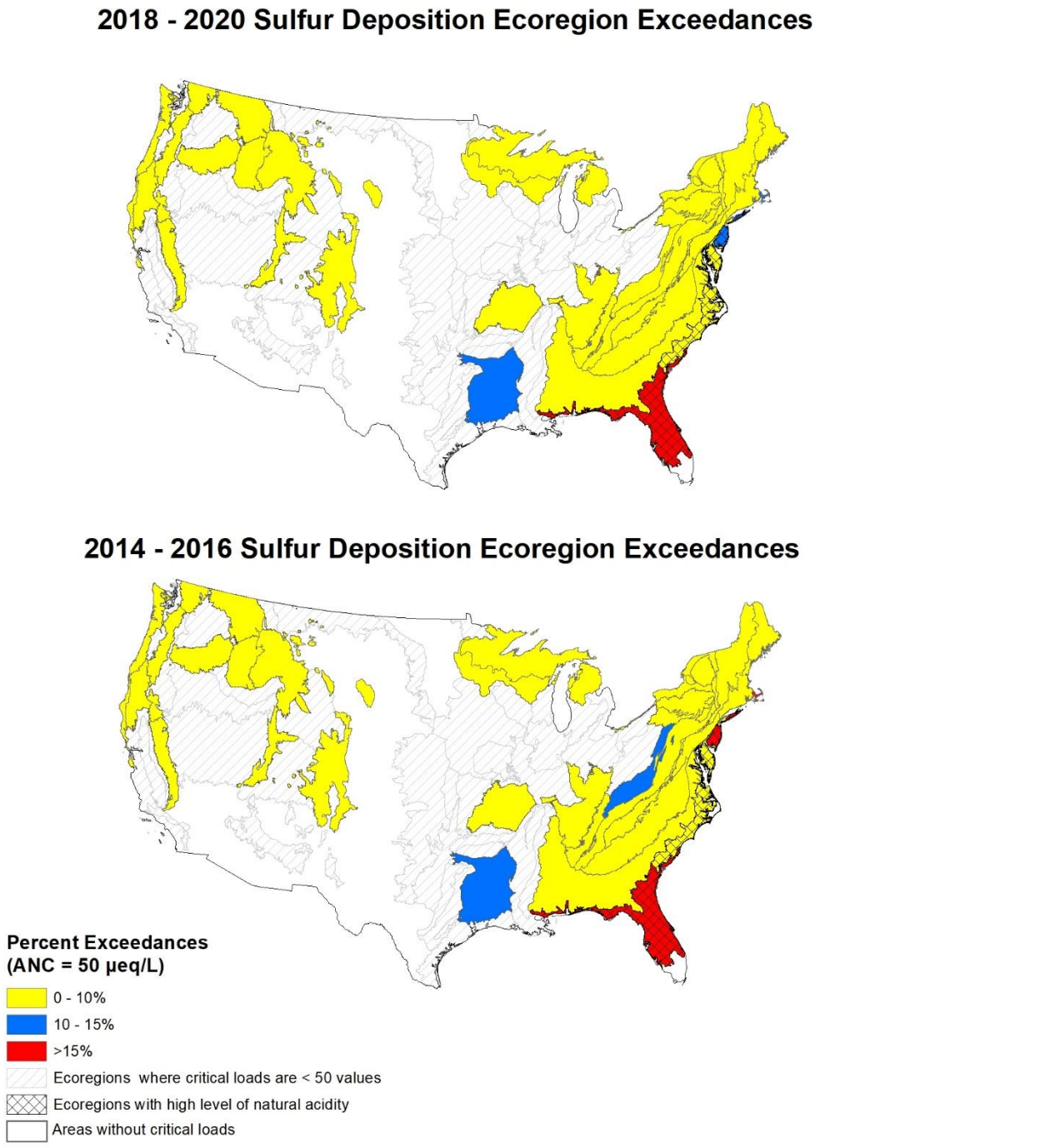

Figure 5A-38. Percent of CLs exceeded per ecoregion for S only deposition from 2018-20 (top) and 2014-16 (bottom) for an ANC threshold of 50 µeq/L. The Southern Coastal Plan (8.5.3) and Atlantic Coastal Pine Barrens (8.5.4) ecoregions are cross hatched to indicate natural high level of acidity.

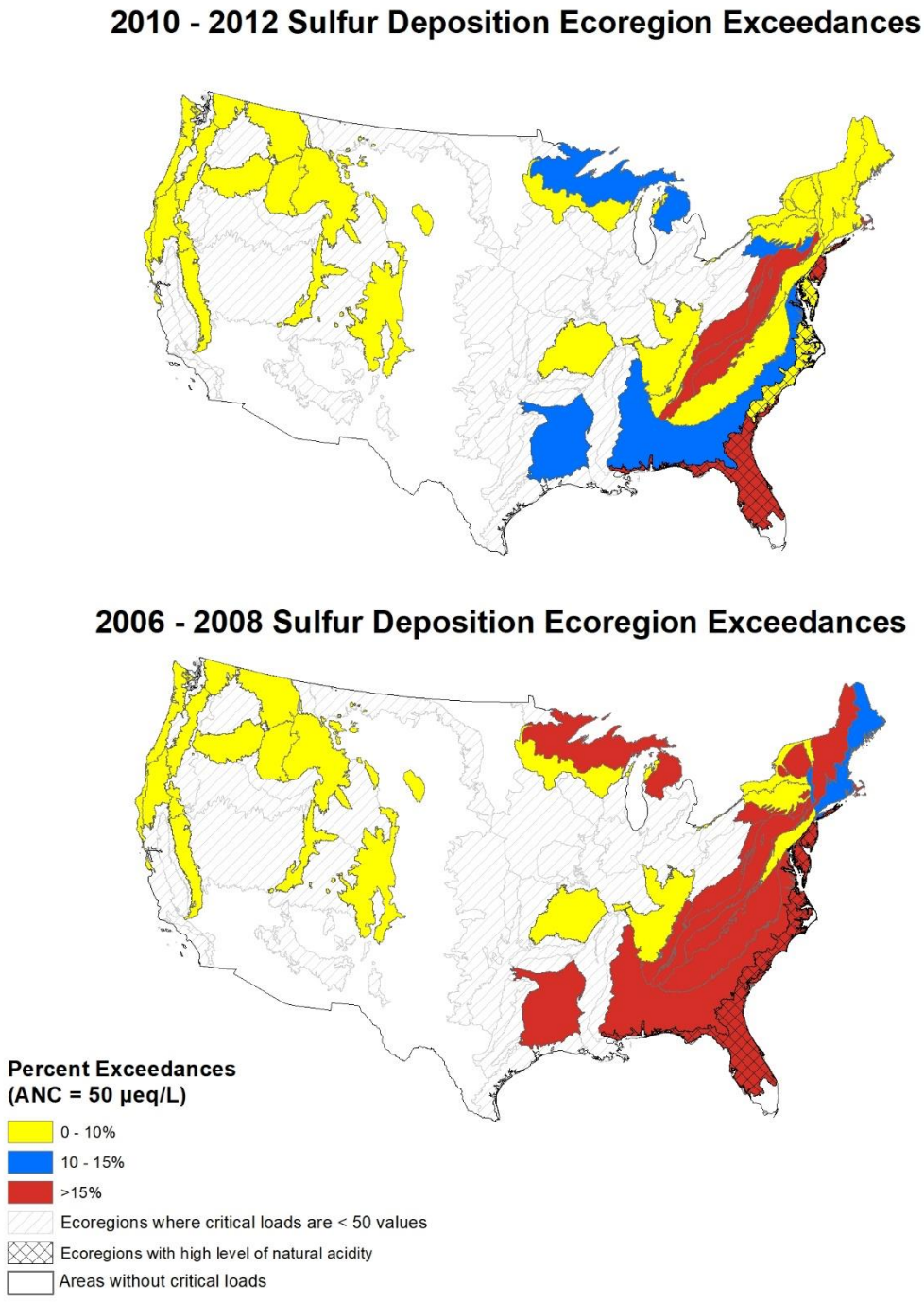

Figure 5A-39. Percent of CLs exceeded per ecoregion for S only deposition from 2010-12 (top) and 2006-08 (bottom) for an ANC threshold of 50 µeq/L. The Southern Coastal Plan (8.5.3) and Atlantic Coastal Pine Barrens (8.5.4) ecoregions are cross hatched to indicate natural high level of acidity.

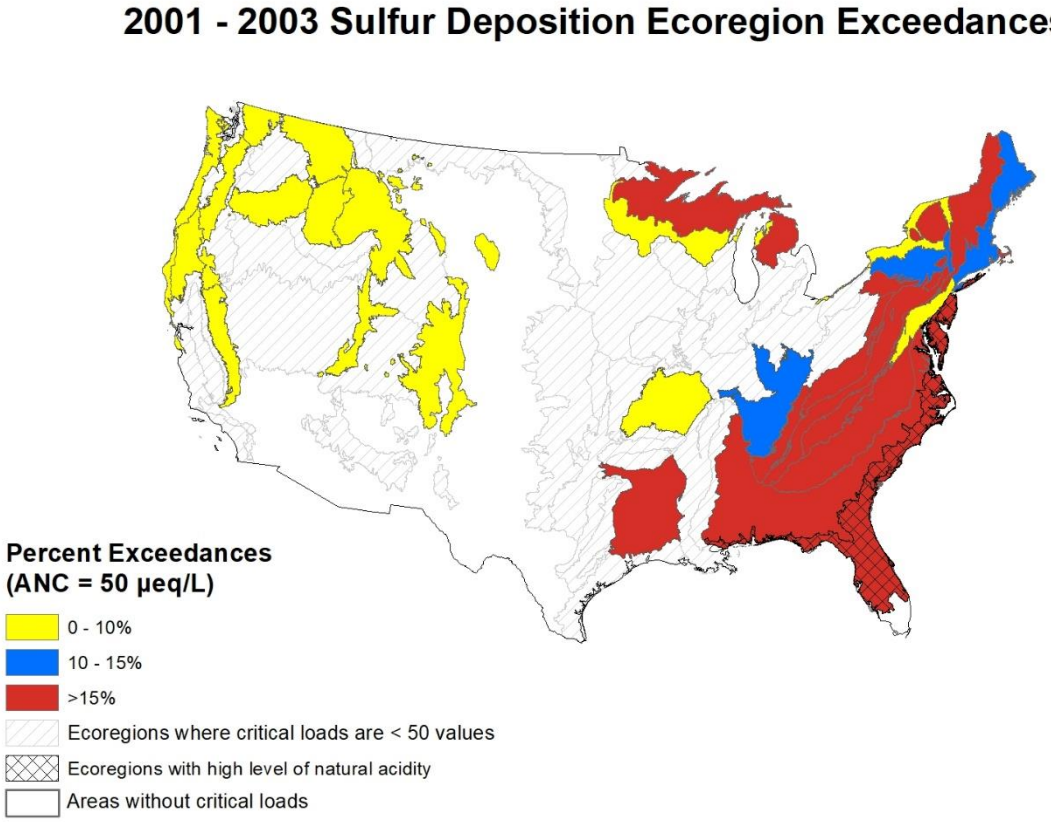

Figure 5A-40. Percent of CLs exceeded per ecoregion for S only deposition from 2001-03 for an ANC threshold of 50 µeq/L. The Southern Coastal Plan (8.5.3) and Atlantic Coastal Pine Barrens (8.5.4) ecoregions are cross hatched to indicate natural high level of acidity.

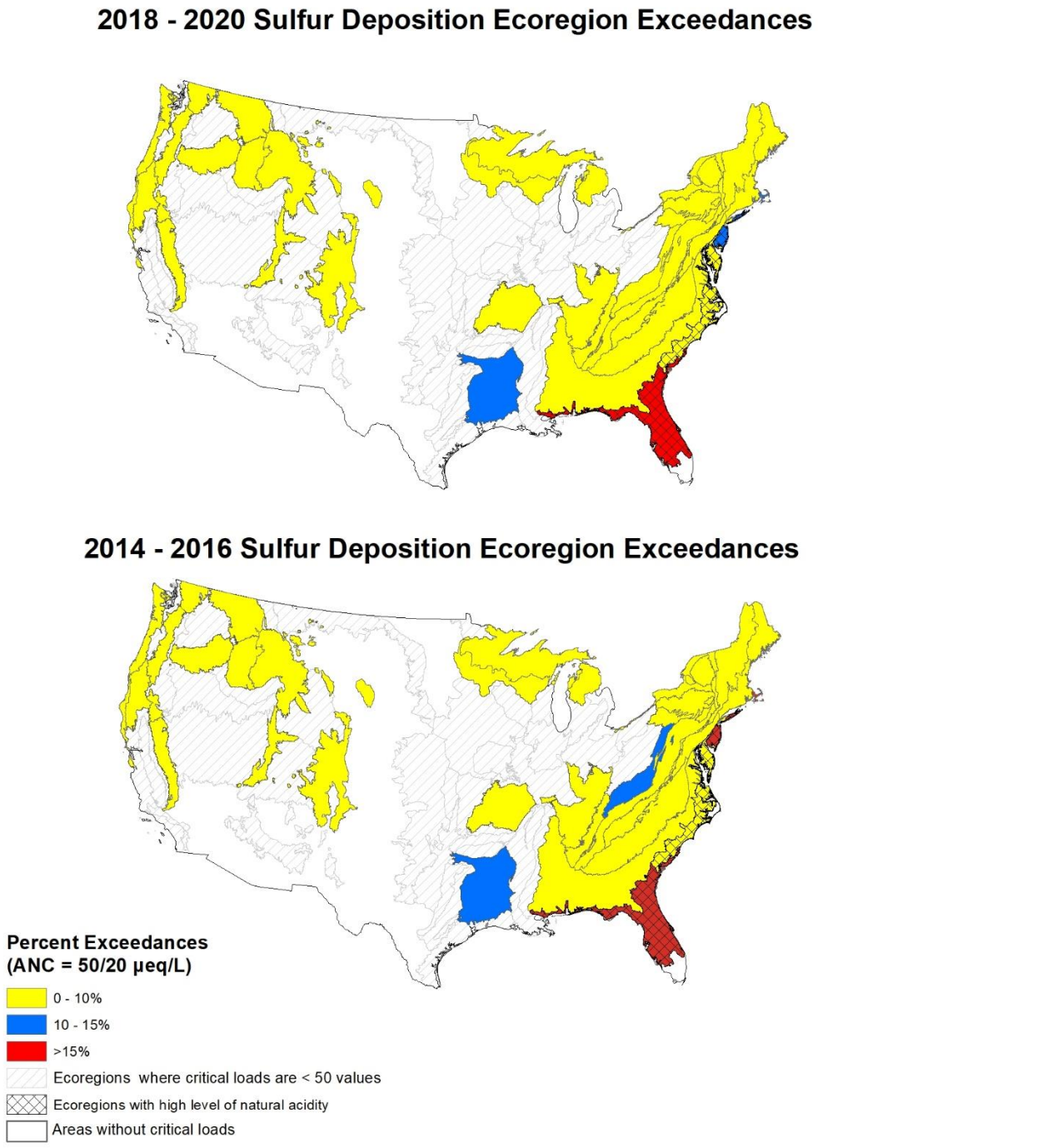

Figure 5A-41. Percent of CLs exceeded per ecoregion for S only deposition from 2018-20 (top) and 2014-16 (bottom) for an ANC threshold of 50 µeq/L for East and 20 µeq/L for the West. The Southern Coastal Plan (8.5.3) and Atlantic Coastal Pine Barrens (8.5.4) ecoregions are cross hatched to indicate natural high level of acidity.

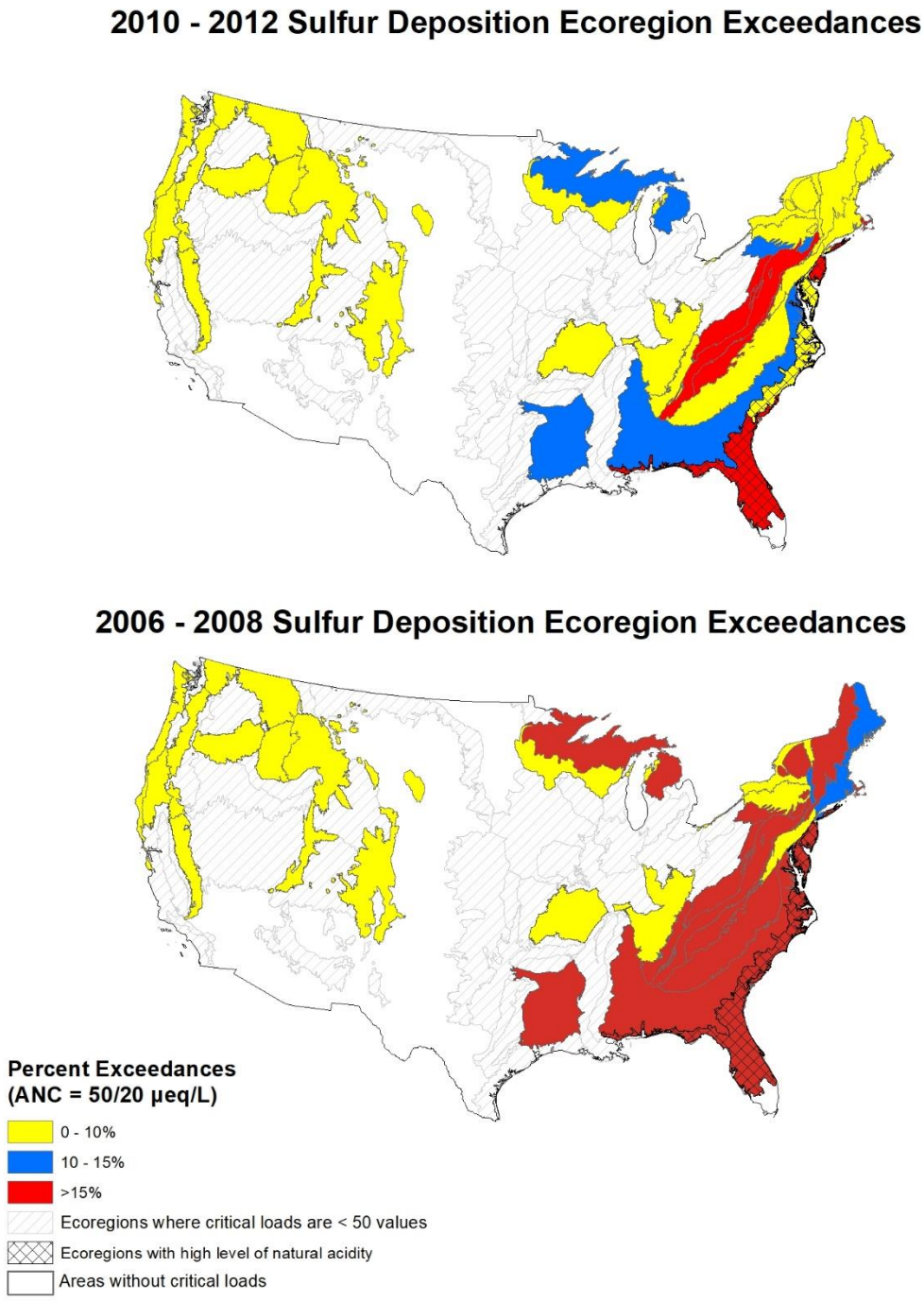

Figure 5A-42. Percent of CLs exceeded per ecoregion for S only deposition from 2010-12 (top) and 2006-08 (bottom) for an ANC threshold of 50 µeq/L for East and 20 µeq/L for the West. The Southern Coastal Plan (8.5.3) and Atlantic Coastal Pine Barrens (8.5.4) ecoregions are cross hatched to indicate natural high level of acidity.

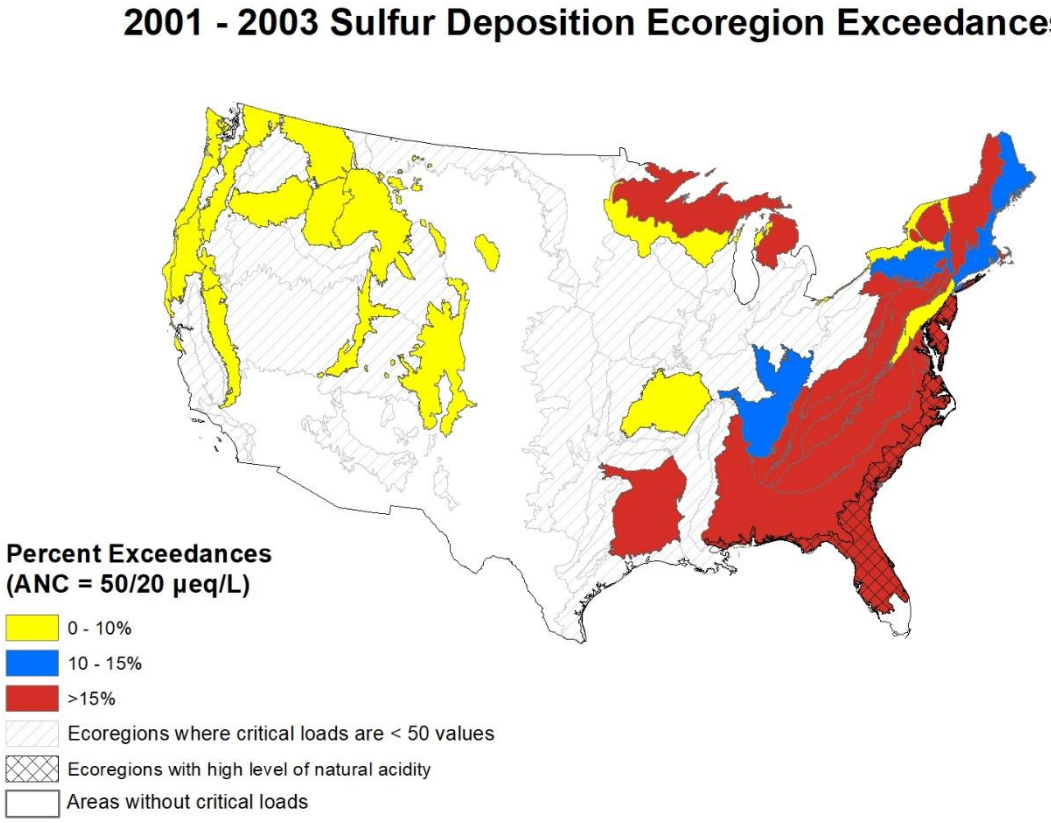

Figure 5A-43. Percent of CLs exceeded per ecoregion for S only deposition from 2001-03 for an ANC threshold of 50 µeq/L for East and 20 µeq/L for the West. The Southern Coastal Plan (8.5.3) and Atlantic Coastal Pine Barrens (8.5.4) ecoregions are cross hatched to indicate natural high level of acidity.

5A.2.2.2 Ecoregion Summary – Percent Exceedances as a Function of Total S deposition

In this section, the results for the deposition estimates across the five deposition periods (2001-03, 2006-08, 2010-12, 2014-06, 2018-20) are summarized by the number of ecoregions with over 10, 15, 20, 25, and 30% of their CLs estimated to be exceeded. Ecoregions included in this analysis are those for which there are at least 50 waterbodies with CLs and that (1) are not one of the three ecoregions identified as naturally acidic (see 5A.2.2.1 above), and (2) had waterbodies with a CL greater than zero (for ANC of 50 µeq/L in the East and 20 µeq/L in the West) that was exceeded during any of the five time periods. These criteria yield a total of 25 ecoregions across the CONUS with 18 and 7 ecoregions in the eastern and western U.S., respectively.

In the discussions below, ecoregion S deposition for each time period is represented by the median across waterbodies with CLs in the ecoregion. Table 5A-25 provides the minimum, maximum, and median of these ecoregion medians. Deposition levels were summarized for the five deposition periods and three ANC thresholds (20, 30, and 50 μeq/L) for the eastern and western U.S. separately and together. Deposition for ecoregions in the eastern U.S. ranged from a median value (across waterbodies with CLs) of 11.08 kg S/ha-yr in 2001-03 to one of 2.04 kg S/ha-yr in 2018-20. Total S deposition for ecoregions in the western U.S. was lower, ranging from a median of 1.40 kg S/ha-yr in 2001-03 to 0.87 kg S/ha-yr in 2018-20.

Table 5A-25. Minimum, maximum, and median S deposition for 25 ecoregions in analysis. Ecoregion deposition values are medians of deposition at sites with CLs in the ecoregion.

	Median Sulfur Deposition, kg S/ha-yr				
	2001-03	**2006-08**	**2010-12**	**2014-16**	**2018-20**
	All 18 Eastern Ecoregions				
Minimum	4.01	3.10	2.34	1.88	1.31
Maximum	17.27	14.44	7.25	4.58	3.88
Median	11.08	9.36	4.76	2.97	2.04
	All 7 Western Ecoregions				
Minimum	1.18	1.22	1.02	1.08	0.62
Maximum	1.94	1.83	1.47	1.56	1.19
Median	1.40	1.52	1.29	1.17	0.87
	All 25 Ecoregions in Analysis				
Minimum	1.18	1.22	1.02	1.08	0.62
Maximum	17.27	14.44	7.25	4.58	3.88
Median	7.77	6.50	3.71	2.32	1.73

The summaries below are intended to look at the percent exceedances per ecoregion as a function of annual average total S deposition. For example, for estimated deposition at or below 2 kg S/ha-yr across all ecoregions and deposition periods, there are no ecoregions that have more than 10% of sites exceeding their CLs for an ANC threshold of 50 μeq/L (Table 5A-26). Among the ecoregion-time period combinations with S deposition at or below 3 kg S/ha-yr, there is only one such ecoregion. However, for deposition at or below 10 kg S/ha-yr, there are 22 ecoregion-time periods with >10% EX and 1 with >30% EX. At or below 6, 10, and 15 kg S/ha-yr, there were 13, 22, and 33 ecoregion-time periods, respectively, with >10% of sites exceeding CLs, and 2, 6, and 14 ecoregion-time periods with >20% of sites exceeding CLs. These summaries were done for ANC thresholds of 20, 30, and 50 μeq/L for the eastern U.S., 20 μeq/L western U.S., and combined 50/20, 30/20, and 20 μeq/L for both eastern and western U.S. Results are summarized in Tables 5A-26, 5A-28, 5A-30, 5A-32, 5A-34, 5A-36, and 5A-38.

The cumulative percentages of ecoregion-time periods achieving the various ANC thresholds were also determined and graphed as a function of deposition bin. For example, for ANC of 50 μeq/L and for the eastern U.S, 100% of ecoregions-time period combinations with S deposition at/below 2 kg/ha-yr have less than 10% of sites exceeding their CLs while 60% of ecoregion-time period combinations with S deposition at/below 18 kg S/ha-yr have less than 10% of sites exceeding their CLs, i.e., 40% have > 10% EX (Table 5A-27, Figure 5A-44). Results for the other ANC thresholds are summarized in Tables 5A-29, 5A-31, 5A-33, 5A-35, 5A-37, 5A-39. These cumulative results are graphed in Figures 5A-44 to 5A-49.

Table 5A-26. Number of ecoregion-time period combinations with more than 10, 15, 20, 25 and 30% of waterbodies exceeding their CLs for ANC target of 50 μeq/L. Includes 18 ecoregions in the eastern U.S.

Total Sulfur Deposition (kg S/ha-yr)	No. Eastern Ecoregion-Time Period Combinations	Number of Ecoregion-Time Period Combinations with Specified Percent of Waterbodies Exceeding their CLs				
		10%	15%	20%	25%	30%
≤2	10	0	0	0	0	0
≤3	29	1	0	0	0	0
≤4	41	3	1	0	0	0
≤5	51	9	3	2	1	0
≤6	59	13	4	2	1	0
≤7	63	14	5	3	1	0
≤8	67	18	9	5	3	0
≤9	69	19	9	5	3	0
≤10	73	22	11	6	4	1
≤11	76	24	13	7	4	1
≤12	79	27	15	9	6	3
≤13	81	28	16	10	6	3
≤14	84	31	18	12	8	5
≤15	86	33	20	14	10	7
≤16	88	34	21	15	11	8
≤17	88	34	21	15	11	8
≤18*	90	36	23	17	13	10

* Highest ecoregion median (across sites with CLs) S deposition estimate across the five time periods is 17.27 kg/ha-yr.

Table 5A-27. Cumulative percentage of ecoregion-time period combinations with less than 10, 15, 20, 25, and 30% of waterbodies per ecoregion exceeding their CLs for the ANC target of 50 µeq/L as a function of total S deposition. 100% indicates there were no ecoregion-time period combinations that had percent exceedances above specified value. For the 18 eastern U.S. ecoregions and five deposition periods (2001-03, 2006-08, 2010-12, 2014-06, 2018-20) (See Table 5A-26 for data).

Total Sulfur Deposition (kg S/ha-yr)	Percent of Exceedances Across the 5 deposition Periods				
	10%	15%	20%	25%	30%
≤2	100%	100%	100%	100%	100%
≤3	97%	100%	100%	100%	100%
≤4	93%	98%	100%	100%	100%
≤5	82%	94%	96%	98%	100%
≤6	78%	93%	97%	98%	100%
≤7	78%	92%	95%	98%	100%
≤8	73%	87%	93%	96%	100%
≤9	72%	87%	93%	96%	100%
≤10	70%	85%	92%	95%	99%
≤11	68%	83%	91%	95%	99%
≤12	66%	81%	89%	92%	96%
≤13	65%	80%	88%	93%	96%
≤14	63%	79%	86%	90%	94%
≤15	62%	77%	84%	88%	92%
≤16	61%	76%	83%	88%	91%
≤17	61%	76%	83%	88%	91%
≤18	60%	74%	81%	86%	89%

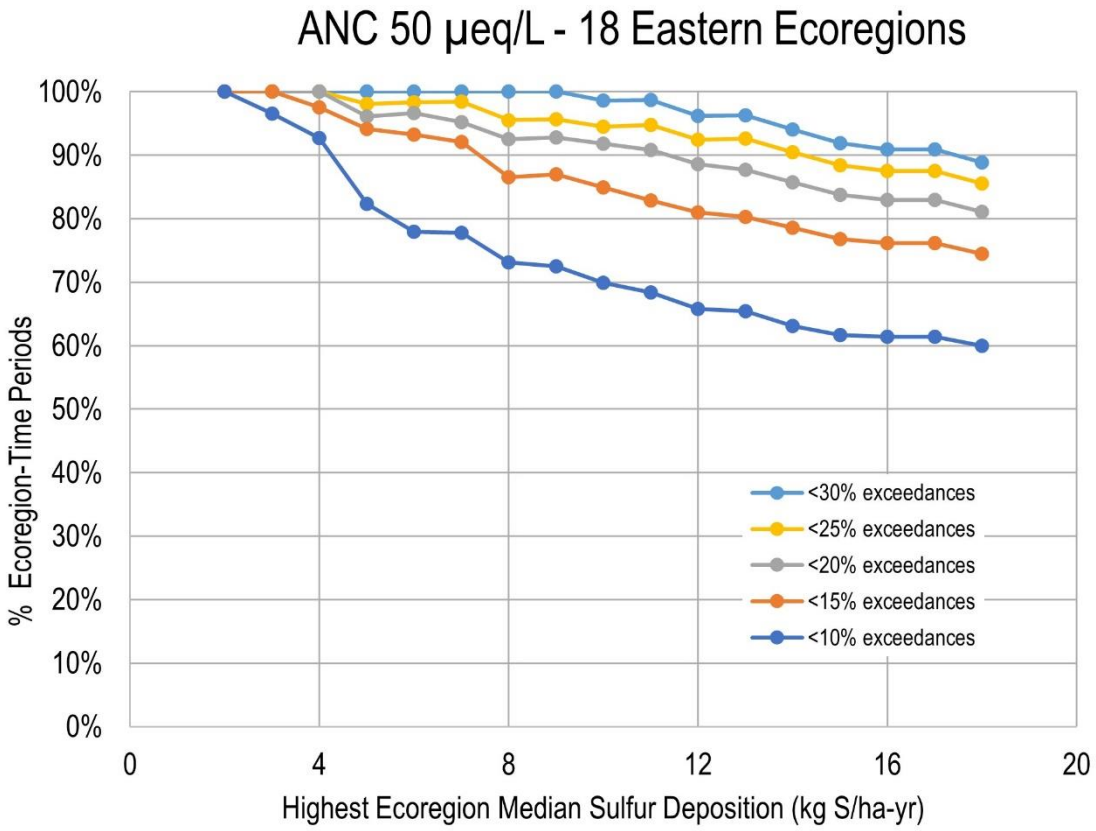

Figure 5A-44. Cumulative percentage of ecoregion-time period combinations with CL exceedances below 10, 15, 20, 25, or 30%. 100% indicates there was no ecoregion that had a percent exceedance above 10, 15, 20, 25, or 30% for that deposition level bin. Critical load exceedances based on ANC target of 50 µeq/L for the 18 eastern U.S. ecoregions and five deposition periods (2001-03, 2006-08, 2010-12, 2014-06, 2018-20) (See Table 5A-27 for values).

Table 5A-28. Number of ecoregion-time period combinations with >10, >15, >20, >25, >30% of waterbodies exceeding their CLs for ANC target of 30 µeq/L as a function of total S deposition across all 5 deposition periods (2001-03, 2006-08, 2010-12, 2014-06, 2018-20). Includes 18 ecoregions in the eastern U.S.

Total Sulfur Deposition (kg S/ha-yr)	Number of Ecoregion-Time Period Combinations with Specified Percent of Waterbodies Exceeding their CLs				
	10%	15%	20%	25%	30%
≤2	0	0	0	0	0
≤3	0	0	0	0	0
≤4	2	0	0	0	0
≤5	4	1	0	0	0
≤6	7	1	0	0	0
≤7	8	2	0	0	0
≤8	12	6	1	0	0
≤9	13	6	1	0	0
≤10	16	8	2	1	1
≤11	18	9	3	1	1
≤12	21	11	5	3	3
≤13	22	12	5	3	3
≤14	25	14	7	5	4
≤15	27	16	9	7	6
≤16	28	17	10	8	7
≤17	28	17	10	8	7
≤18	30	19	12	10	9

* Highest ecoregion median (across sites with CLs) S deposition estimate across the five time periods is 17.27 kg/ha-yr.

Table 5A-29. Cumulative percent of ecoregion-time period combinations with less than 10, 15, 20, 25 and 30% of waterbodies per ecoregion exceeding their CLs for the ANC target of 30 μeq/L as a function of total S deposition. 100% indicates there were no ecoregion-time period combinations that had percent exceedances above the specified values. Critical load exceedances for 18 eastern U.S. ecoregions and five deposition periods (2001-03, 2006-08, 2010-12, 2014-06, 2018-20) (See Table 5A-28 for data).

Total Sulfur Deposition (kg S/ha-yr)	Number of Ecoregion with Percent of Exceedances Across the 5 deposition Periods				
	10%	15%	20%	25%	30%
≤2	100%	100%	100%	100%	100%
≤3	100%	100%	100%	100%	100%
≤4	95%	100%	100%	100%	100%
≤5	92%	98%	100%	100%	100%
≤6	88%	98%	100%	100%	100%
≤7	87%	97%	100%	100%	100%
≤8	82%	91%	99%	100%	100%
≤9	81%	91%	99%	100%	100%
≤10	78%	89%	97%	99%	99%
≤11	76%	88%	96%	99%	99%
≤12	73%	86%	94%	96%	96%
≤13	73%	85%	94%	96%	96%
≤14	70%	83%	92%	94%	95%
≤15	69%	81%	90%	92%	93%
≤16	68%	81%	89%	91%	92%
≤17	68%	81%	89%	91%	92%
≤18	67%	79%	87%	89%	90%

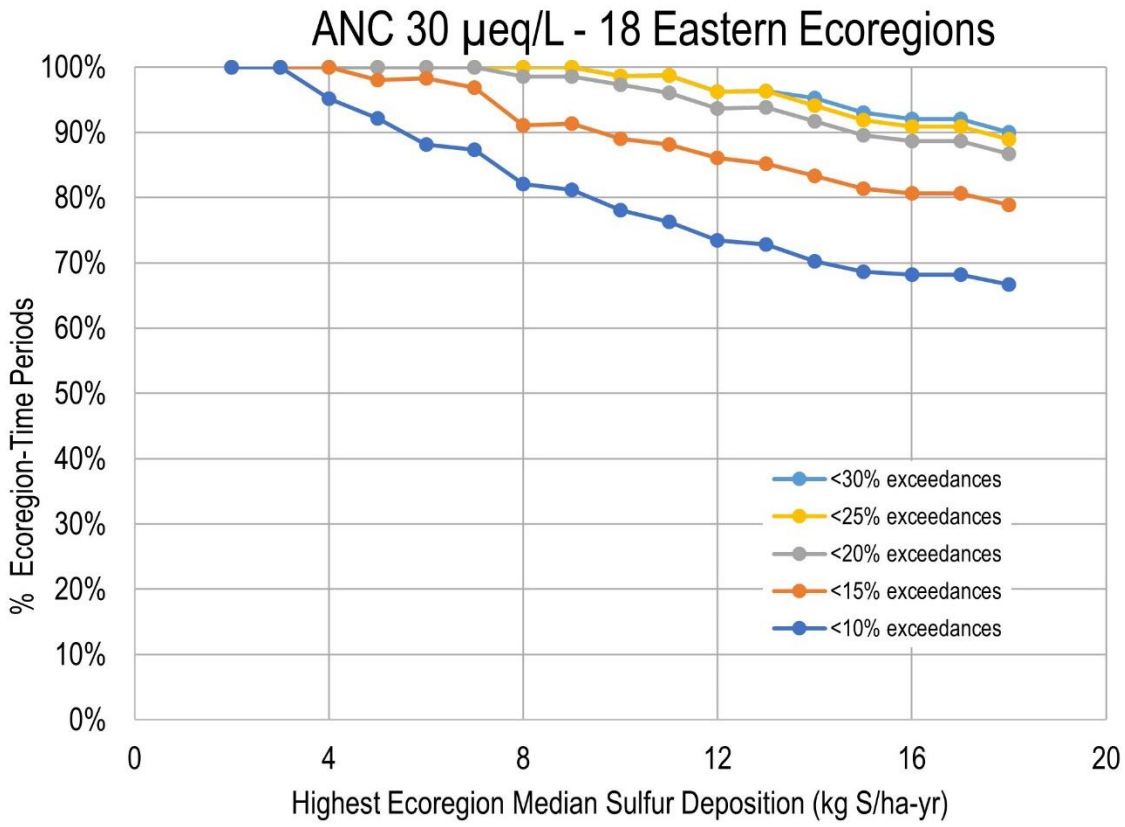

Figure 5A-45. Cumulative percent of ecoregion-time period combinations with CL exceedances below 10, 15, 20, 25, 30%. 100% indicates there was no ecoregion that had a percent exceedance above 10, 15, 20, 25, or 30% for a given deposition level bin. Critical load exceedances based on ANC target of 30 µeq/L for the 18 eastern U.S. ecoregions five deposition periods (2001-03, 2006-08, 2010-12, 2014-06, 2018-20) (See Table 5A-29 for values).

Table 5A-30. Number of ecoregion-time period combinations with >10, >15, >20, >25, >30% of waterbodies exceeding their CLs for ANC target of 20 µeq/L as a function of total S deposition across all 5 deposition periods (2001-03, 2006-08, 2010-12, 2014-06, 2018-20). Includes 18 ecoregions in the eastern U.S.

Total Sulfur Deposition (kg S/ha-yr)	Number of Ecoregion-Time Period Combinations with Specified Percent of Waterbodies Exceeding their CLs				
	10%	15%	20%	25%	30%
≤2	0	0	0	0	0
≤3	0	0	0	0	0
≤4	0	0	0	0	0
≤5	2	1	0	0	0
≤6	4	1	0	0	0
≤7	5	1	0	0	0
≤8	9	4	0	0	0
≤9	9	4	0	0	0
≤10	11	6	1	1	1
≤11	13	7	2	1	1
≤12	15	9	4	3	2
≤13	16	10	4	3	2
≤14	19	12	6	4	3
≤15	21	14	8	6	4
≤16	22	15	9	7	5
≤17	22	15	9	7	5
≤18	24	17	11	9	7

* Highest ecoregion median (across sites with CLs) S deposition estimate across the five time periods is 17.27 kg/ha-yr.

Table 5A-31. Cumulative percent of ecoregion-time period combinations with less than 10, 15, 20, 25 and 30% of waterbodies per ecoregion exceeding their CLs for the ANC target of 20 µeq/L as a function of total S deposition. 100% indicates there were no ecoregion-time period combinations that had percent exceedances above the specified values. Critical load exceedances for 18 eastern ecoregions and five deposition periods (2001-03, 2006-08, 2010-12, 2014-06, 2018-20) (See Table 5A-30 for data).

Total Sulfur Deposition (kg S/ha-yr)	Number of Ecoregion with Percent of Exceedances Across the 5 deposition Periods				
	10%	15%	20%	25%	30%
≤2	100%	100%	100%	100%	100%
≤3	100%	100%	100%	100%	100%
≤4	100%	100%	100%	100%	100%
≤5	96%	98%	100%	100%	100%
≤6	93%	98%	100%	100%	100%
≤7	92%	98%	100%	100%	100%
≤8	87%	94%	100%	100%	100%
≤9	87%	94%	100%	100%	100%
≤10	85%	92%	99%	99%	99%
≤11	83%	91%	97%	99%	99%
≤12	81%	89%	95%	96%	97%
≤13	80%	88%	95%	96%	98%
≤14	77%	86%	93%	95%	96%
≤15	76%	84%	91%	93%	95%
≤16	75%	83%	90%	92%	94%
≤17	75%	83%	90%	92%	94%
≤18	73%	81%	88%	90%	92%

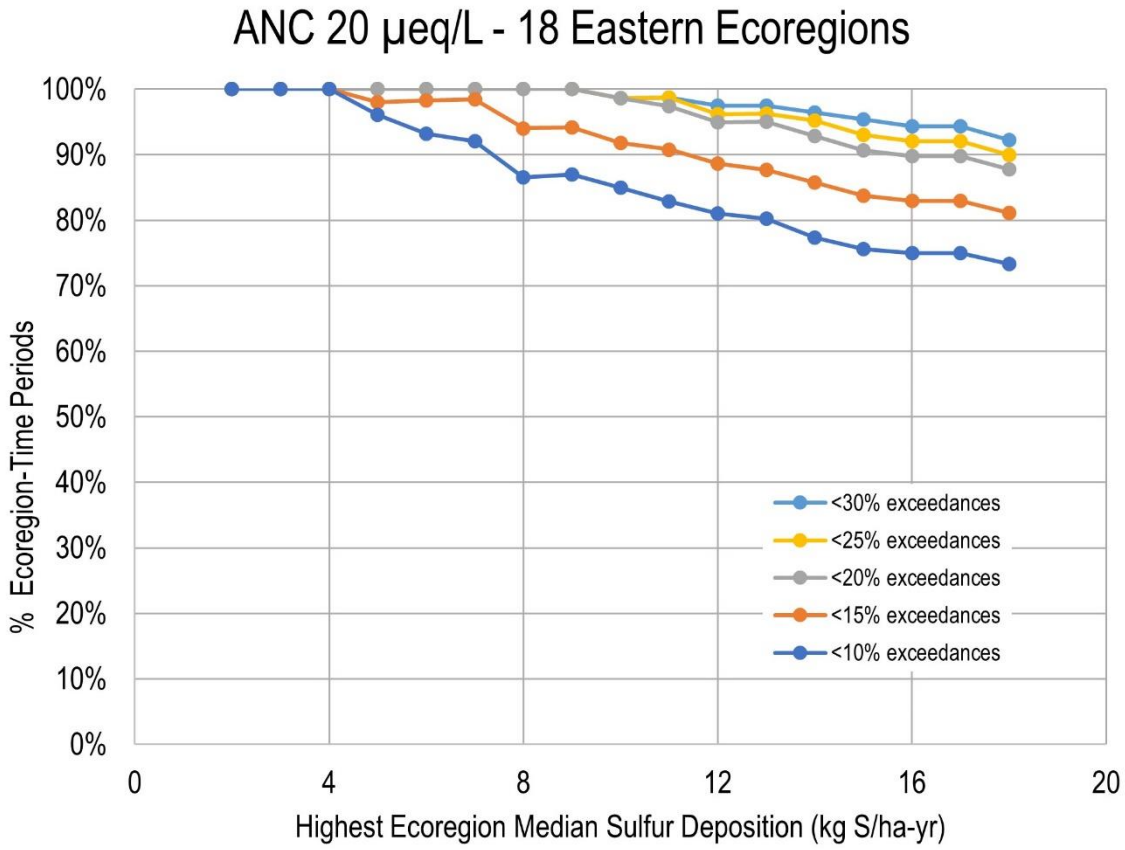

Figure 5A-46. Cumulative percent of ecoregion-time period combinations with CL exceedances below 10, 15, 20, 25, or 30%. 100% indicates there was no ecoregion that had a percent exceedance above 10, 15, 20, 25, or30%. Critical load exceedances based on ANC target of 20 µeq/L for the 18 eastern U.S. ecoregions across all 5 deposition periods (2001-03, 2006-08, 2010-12, 2014-06, 2018-20) (See Table 5A-31 for values).

Table 5A-32. Number of ecoregion-time period combinations with >10, >15, >20, >25, >30% of waterbodies exceeding their CLs for ANC target of 20 µeq/L as a function of total S deposition across all 5 deposition periods (2001-03, 2006-08, 2010-12, 2014-06, 2018-20) for 7 ecoregions in the western U.S.

Total Sulfur Deposition (kg S/ha-yr)	Number of Ecoregion-Time Period Combinations with Specified Percent of Waterbodies Exceeding their CLs				
	10%	15%	20%	25%	30%
≤2*	0	0	0	0	0
* Highest ecoregion median (across sites with CLs) S deposition estimate for 7 western ecoregions across the five time periods is 1.94 kg/ha-yr.					

Table 5A-33. Cumulative percent of waterbodies in ecoregions meeting the target ANC values as a function of total S deposition across all 5 deposition periods (2001-03, 2006-08, 2010-12, 2014-06, 2018-20). 100% indicates there were no ecoregions that had percent exceedances above >10, >15, >20, >25, >30% for a given deposition level. Critical load exceedances based on ANC target of 20 µeq/L for the western U.S. (See Table 5A-32 for data).

Total Sulfur Deposition (kg S/ha-yr)	Number of Ecoregion with Percent of Exceedances Across the 5 deposition Periods				
	10%	15%	20%	25%	30%
≤2	100%	100%	100%	100%	100%

Table 5A-34. Number of ecoregion-time period combinations with >10, >15, >20, >25, >30% of waterbodies exceeding their CLs for ANC target of 50 µeq/L for the east and 20 µeq/L for the west as a function of total S deposition across all 5 deposition periods (2001-03, 2006-08, 2010-12, 2014-06, 2018-20). Includes 25 ecoregions across the U.S.

Total Sulfur Deposition (kg S/ha-yr)	Number of Ecoregion-Time Period Combinations with Specified Percent of Waterbodies Exceeding their CLs				
	10%	15%	20%	25%	30%
≤2	0	0	0	0	0
≤3	1	0	0	0	0
≤4	3	1	0	0	0
≤5	9	3	2	1	0
≤6	13	4	2	1	0
≤7	14	5	3	1	0
≤8	18	9	5	3	0
≤9	19	9	5	3	0
≤10	22	11	6	4	1
≤11	24	13	7	4	1
≤12	27	15	9	6	3
≤13	28	16	10	6	3
≤14	31	18	12	8	5
≤15	33	20	14	10	7
≤16	34	21	15	11	8
≤17	34	21	15	11	8
≤18	36	23	17	13	10

* Highest ecoregion median (across sites with CLs) S deposition estimate across the five time periods is 17.27 kg/ha-yr.

Table 5A-35. Cumulative percent of ecoregion-time period combinations with less than 10, 15, 20, 25 and 30% of waterbodies per ecoregion exceeding their CLs for the ANC target of 50 µeq/L for the east and 20 µeq/L for the west as a function of total S deposition. 100% indicates there were no ecoregion-time period combinations that had percent exceedances above the specified values. Critical load exceedances for 18 eastern and 7 western ecoregions and five deposition periods (2001-03, 2006-08, 2010-12, 2014-06, 2018-20) (See Table 5A-34 for data).

Total Sulfur Deposition (kg S/ha-yr)	Number of Ecoregion with Percent of Exceedances Across the 5 deposition Periods				
	10%	15%	20%	25%	30%
≤2	100%	100%	100%	100%	100%
≤3	98%	100%	100%	100%	100%
≤4	96%	99%	100%	100%	100%
≤5	90%	97%	98%	99%	100%
≤6	86%	96%	98%	99%	100%
≤7	86%	95%	97%	99%	100%
≤8	82%	91%	95%	97%	100%
≤9	82%	91%	95%	97%	100%
≤10	80%	90%	94%	96%	99%
≤11	78%	88%	94%	96%	99%
≤12	76%	87%	92%	95%	97%
≤13	76%	86%	91%	95%	97%
≤14	74%	85%	90%	93%	96%
≤15	73%	83%	88%	92%	94%
≤16	72%	83%	88%	91%	93%
≤17	72%	83%	88%	91%	93%
≤18	71%	82%	86%	90%	92%

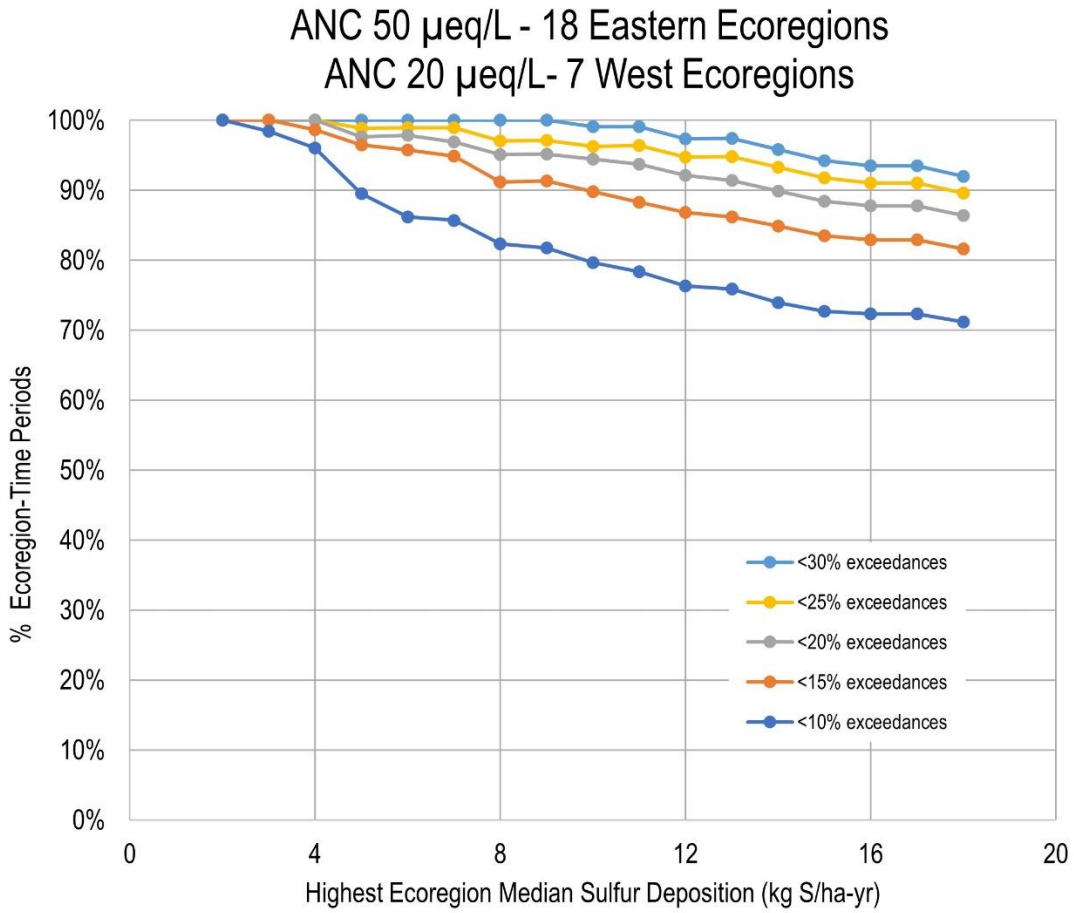

Figure 5A-47. Cumulative percent of ecoregion-time period combinations with CL exceedances below 10, 15, 20, 25, or 30%. 100% indicates there was no ecoregion that had a percent exceedance above 10, 15, 20, 25, or 30% for a given deposition level bin. Critical load exceedances based on ANC target of 50 µeq/L for the 18 east ecoregions and 20 µeq/L for the 7 west ecoregions and five deposition periods (2001-03, 2006-08, 2010-12, 2014-06, 2018-20) (See Table 5A-35 for values).

Table 5A-36. Number of ecoregion-time period combinations with >10, >15, >20, >25, >30% of waterbodies exceeding their CLs for ANC target of 30 µeq/L for the east and 20 µeq/L for the west as a function of total S deposition across all 5 deposition periods (2001-03, 2006-08, 2010-12, 2014-06, 2018-20). Includes 25 ecoregions across the U.S.

Total Sulfur Deposition (kg S/ha-yr)	Number of Ecoregion-Time Period Combinations with Specified Percent of Waterbodies Exceeding their CLs				
	10%	15%	20%	25%	30%
≤2	0	0	0	0	0
≤3	0	0	0	0	0
≤4	2	0	0	0	0
≤5	4	1	0	0	0
≤6	7	1	0	0	0
≤7	8	2	0	0	0
≤8	12	6	1	0	0
≤9	13	6	1	0	0
≤10	16	8	2	1	1
≤11	18	9	3	1	1
≤12	21	11	5	3	3
≤13	22	12	5	3	3
≤14	25	14	7	5	4
≤15	27	16	9	7	6
≤16	28	17	10	8	7
≤17	28	17	10	8	7
≤18	30	19	12	10	9

* Highest ecoregion median (across sites with CLs) S deposition estimate across the five time periods is 17.27 kg/ha-yr.

Table 5A-37. Cumulative percent of ecoregion-time period combinations with less than 10, 15, 20, 25 and 30% of waterbodies per ecoregion exceeding their CLs for the ANC target of 30 µeq/L for the east and 20 µeq/L for the west as a function of total S deposition. 100% indicates there were no ecoregion-time period combinations that had percent exceedances above the specified values. Critical load exceedances for the 18 eastern and 7 western ecoregions and five deposition periods (2001-03, 2006-08, 2010-12, 2014-06, 2018-20) (See Table 5A-36 for data).

Total Sulfur Deposition (kg S/ha-yr)	Number of Ecoregion with Percent of Exceedances Across the 5 deposition Periods				
	10%	15%	20%	25%	30%
≤2	100%	100%	100%	100%	100%
≤3	100%	100%	100%	100%	100%
≤4	97%	100%	100%	100%	100%
≤5	95%	99%	100%	100%	100%
≤6	93%	99%	100%	100%	100%
≤7	92%	98%	100%	100%	100%
≤8	88%	94%	99%	100%	100%
≤9	88%	94%	99%	100%	100%
≤10	85%	93%	98%	99%	99%
≤11	84%	92%	97%	99%	99%
≤12	82%	90%	96%	97%	97%
≤13	81%	90%	96%	97%	97%
≤14	79%	88%	94%	96%	97%
≤15	78%	87%	93%	94%	95%
≤16	77%	86%	92%	93%	94%
≤17	77%	86%	92%	93%	94%
≤18	76%	85%	90%	92%	93%

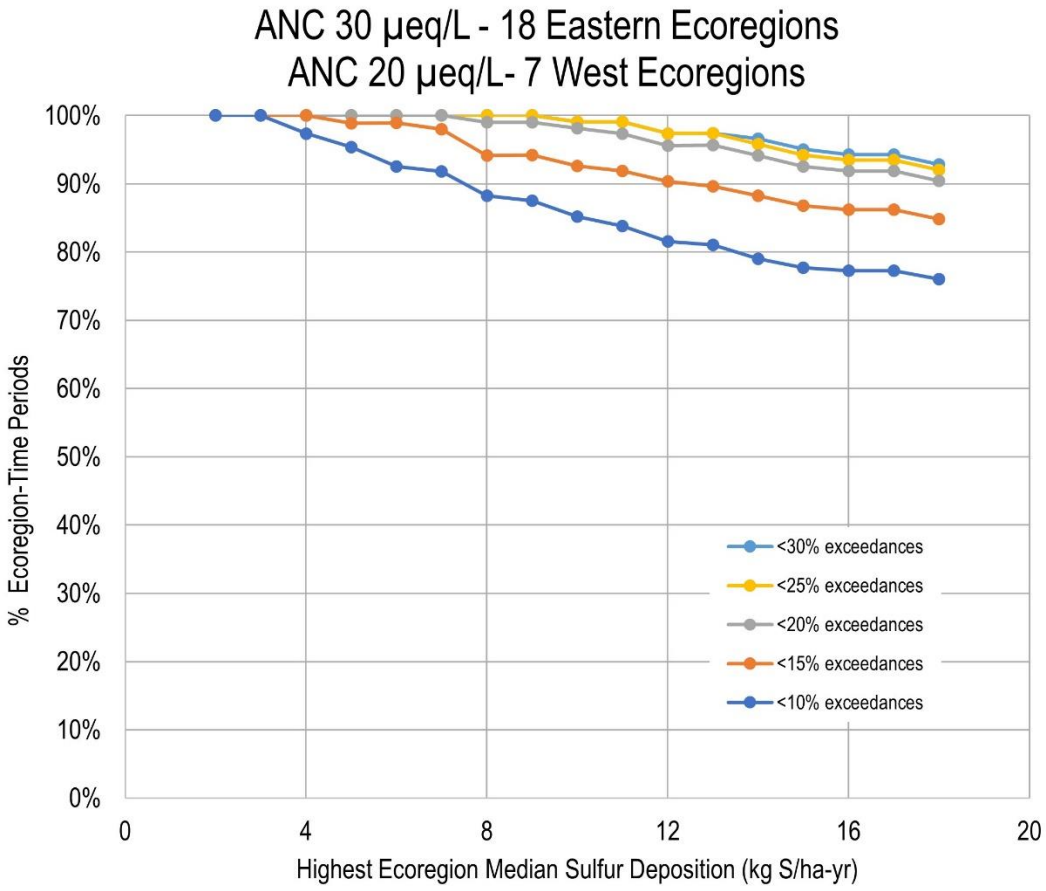

Figure 5A-48. Cumulative percent of ecoregion-time period combinations with CL exceedances below10, 15, 20, 25, or 30%. 100% indicates there was no ecoregion that had a percent exceedance above 10, 15, 20, 25, or 30% for a given deposition level bin. Critical load exceedances based on ANC target of 30 µeq/L for the 18 east ecoregions and 20 µeq/L for the 7 west ecoregions and five deposition periods (2001-03, 2006-08, 2010-12, 2014-06, 2018-20) (See Table 5A-37 for values).

Table 5A-38. Number of ecoregion-time period combinations with >10, >15, >20, >25, >30% of waterbodies exceeding their CLs for ANC target of 20 µeq/L for both the east and west as a function of total S deposition across all 5 deposition periods (2001-03, 2006-08, 2010-12, 2014-06, 2018-20). Includes 25 ecoregions across the U.S.

Total Sulfur Deposition (kg S/ha-yr)	Number of Ecoregion-Time Period Combinations with Specified Percent of Waterbodies Exceeding their CLs				
	10%	15%	20%	25%	30%
≤2	0	0	0	0	0
≤3	0	0	0	0	0
≤4	0	0	0	0	0
≤5	2	1	0	0	0
≤6	4	1	0	0	0
≤7	5	1	0	0	0
≤8	9	4	0	0	0
≤9	9	4	0	0	0
≤10	11	6	1	1	1
≤11	13	7	2	1	1
≤12	15	9	4	3	2
≤13	16	10	4	3	2
≤14	19	12	6	4	3
≤15	21	14	8	6	4
≤16	22	15	9	7	5
≤17	22	15	9	7	5
≤18	24	17	11	9	7

* Highest ecoregion median (across sites with CLs) S deposition estimate across the five time periods is 17.27 kg/ha-yr.

Table 5A-39. Cumulative percent of ecoregion-time period combinations with less than 10, 15, 20, 25 and 30% of waterbodies per ecoregion exceeding their CLs for the ANC target of 20 μeq/L as a function of total S deposition. 100% indicates there were no ecoregion-time period combinations that had percent exceedances above the specified values. Critical load exceedances for 18 eastern and 7 western ecoregions and five deposition periods (2001-03, 2006-08, 2010-12, 2014-06, 2018-20) (See Table 5A-38 for data).

Total Sulfur Deposition (kg S/ha-yr)	Number of Ecoregion with Percent of Exceedances Across the 5 deposition Periods				
	10%	15%	20%	25%	30%
≤2	100%	100%	100%	100%	100%
≤3	100%	100%	100%	100%	100%
≤4	100%	100%	100%	100%	100%
≤5	98%	99%	100%	100%	100%
≤6	96%	99%	100%	100%	100%
≤7	95%	99%	100%	100%	100%
≤8	91%	96%	100%	100%	100%
≤9	91%	96%	100%	100%	100%
≤10	90%	94%	99%	99%	99%
≤11	88%	94%	98%	99%	99%
≤12	87%	92%	96%	97%	98%
≤13	86%	91%	97%	97%	98%
≤14	84%	90%	95%	97%	97%
≤15	83%	88%	93%	95%	97%
≤16	82%	88%	93%	94%	96%
≤17	82%	88%	93%	94%	96%
≤18	81%	86%	91%	93%	94%

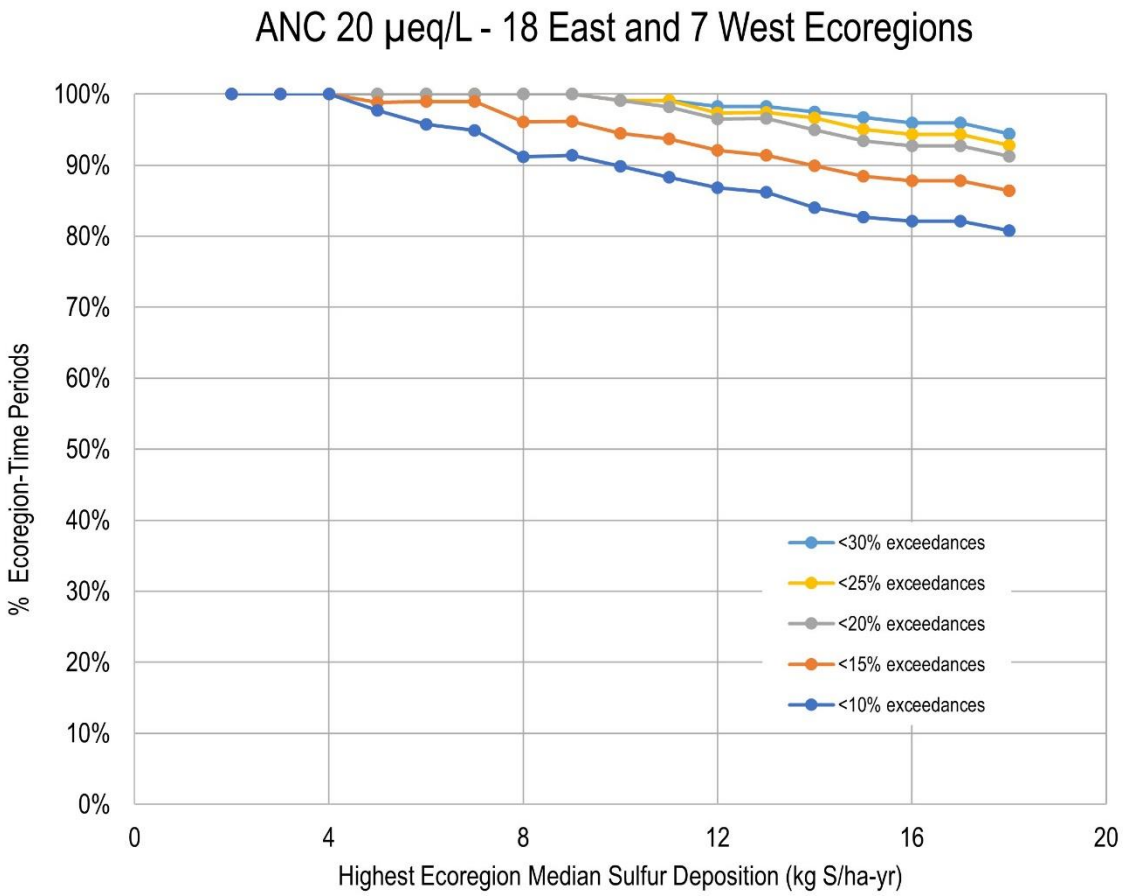

Figure 5A-49. Cumulative percent of ecoregion-time period combinations with CL exceedances below 10, 15, 20, 25, or 30%. 100% indicates there was no ecoregion that had a percent exceedance above 10, 15, 20, 25, or 30% for a given deposition level bin. Critical load exceedances based on ANC target of 20 µeq/L for the 18 east ecoregions and 7 west ecoregions and five deposition periods (2001-03, 2006-08, 2010-12, 2014-06, 2018-20) (See Table 5A-39 for values).

Figure 5A-50 summarizes the percentage of waterbodies per each of the 25 ecoregions that were estimated to achieve ANC values of 20, (E&W), 30 (E only) and 50 (E only) µeq/L, based on CLs greater than zero and annual average S deposition for 2018-2020 and 2014-2016. These percentages per ecoregion are graphed versus that ecoregion's median deposition (across sites with CLs). For 2014-16 deposition estimates, more than 10% of waterbodies in two ecoregions exceed their CLs for ANC of 50 µeq/L and just one ecoregion for ANC of 20 µeq/L. In terms of percentage of waterbodies estimated to achieve the ANC targets, this means that more than 80% of waterbodies in each of the 25 ecoregions were estimated to achieve an ANC at or above 50 µeq/L. For the 2018-20 deposition estimates at or above 90% of waterbodies in each

of the 25 ecoregions were estimated to achieve an ANC at or above 20 µeq/L and at or above 90% in all but one ecoregion were estimated to achieve an ANC level at or above 20 µeq/L.

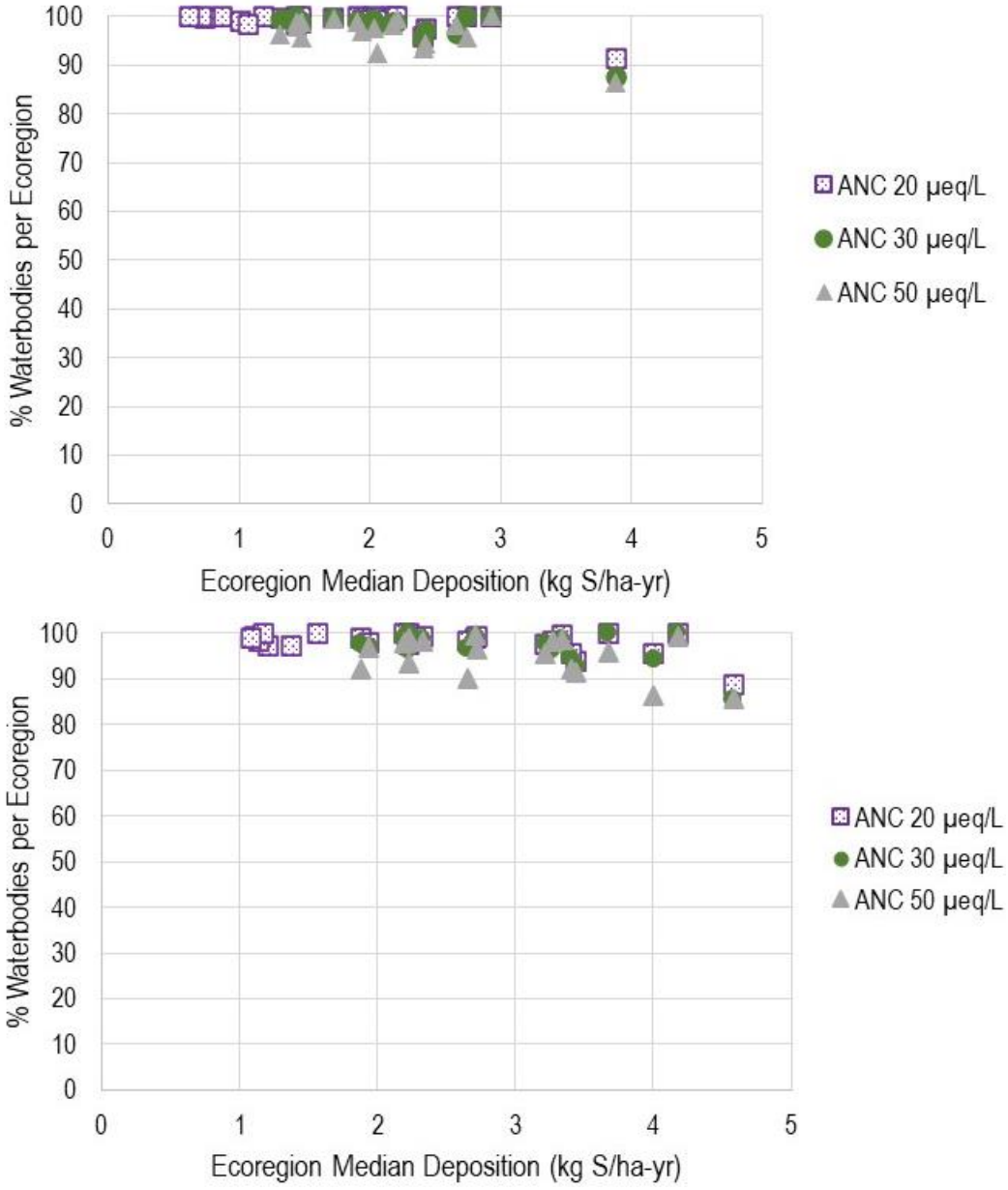

Figure 5A-50. Percentage of waterbodies per each of the 25 ecoregions that were estimated to achieve ANC values of 20 (E&W), 30 (E only) and 50 (E only) µeq/L based on CLs greater than zero and annual average S deposition for 2018-2020 (upper) and 2014-2016 (lower) by ecoregion median (across sites with CLs).

Figure 5A-51 presents percentage of waterbodies achieving ANC of 20 µeq/L, 30 µeq/L, 50 µeq/L and 50/20 µeq/L (50 in eastern ecoregions and 20 in western ecoregions) based on deposition estimates in each of the five time periods.

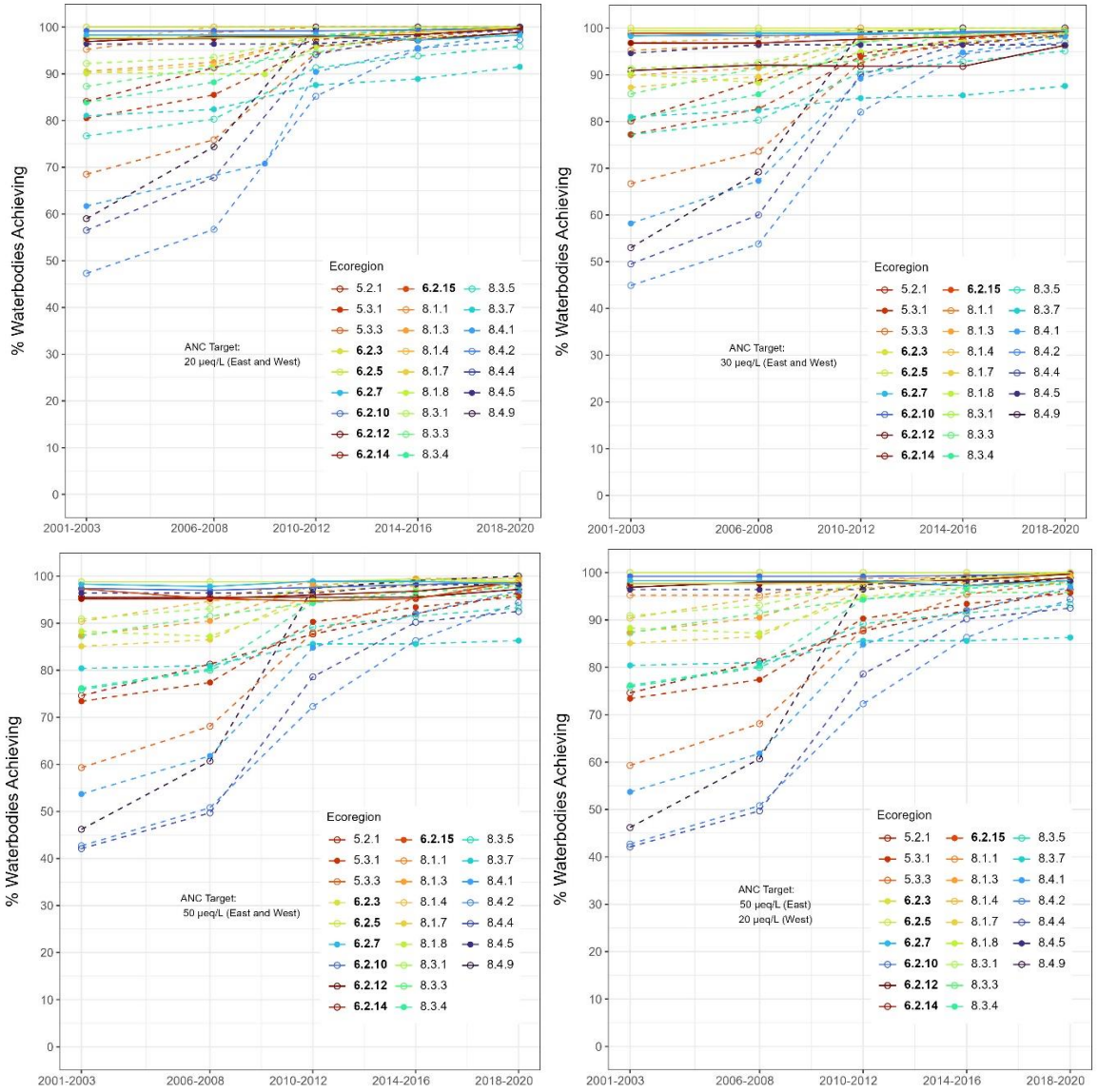

Figure 5A-51. Percent of waterbodies per ecoregion estimated to achieve ANC of 20 µeq/L (top left), 30 µeq/L (top right), 50 µeq/L (bottom left) and 50/20 µeq/L in E/W (2001-2020). Bold text, solid lines indicate western ecoregions.

5A.2.3 Case Study Analysis of Acidification Risk

The areas included in the case study analysis represent geographic diverse acid sensitive areas across the CONUS that have sufficient data to complete a quantitative analysis (Figure 5A-53). This includes the necessary air quality information to assess varying levels of deposition, including monitoring and deposition information. In addition, recent deposition levels across this set of case studies generally reflect variation also observed across the CONUS (Table 5A-40). Five case study areas were identified that meet the criteria (Figure 5A-52). Three of the areas, Northern Minnesota (NOMN), Shenandoah Valley (SHVA) and White Mountain National Forest (WHMT), are in the eastern U.S. and two areas are in the western U.S. (Rocky Mountain National Park [ROMO] and Sierra Nevada Mountains [SINE]). Two of the five areas – WHMT and ROMO - are made up completely of one park or forest. Two other areas, NOMN and SINE, are made up of several contiguous parks, forests, and wilderness areas. The fifth area, SHVA, includes multiple parks or forest areas that are non-contiguous, so the case study boundary was defined by a rectangle incorporating the areas of interest.

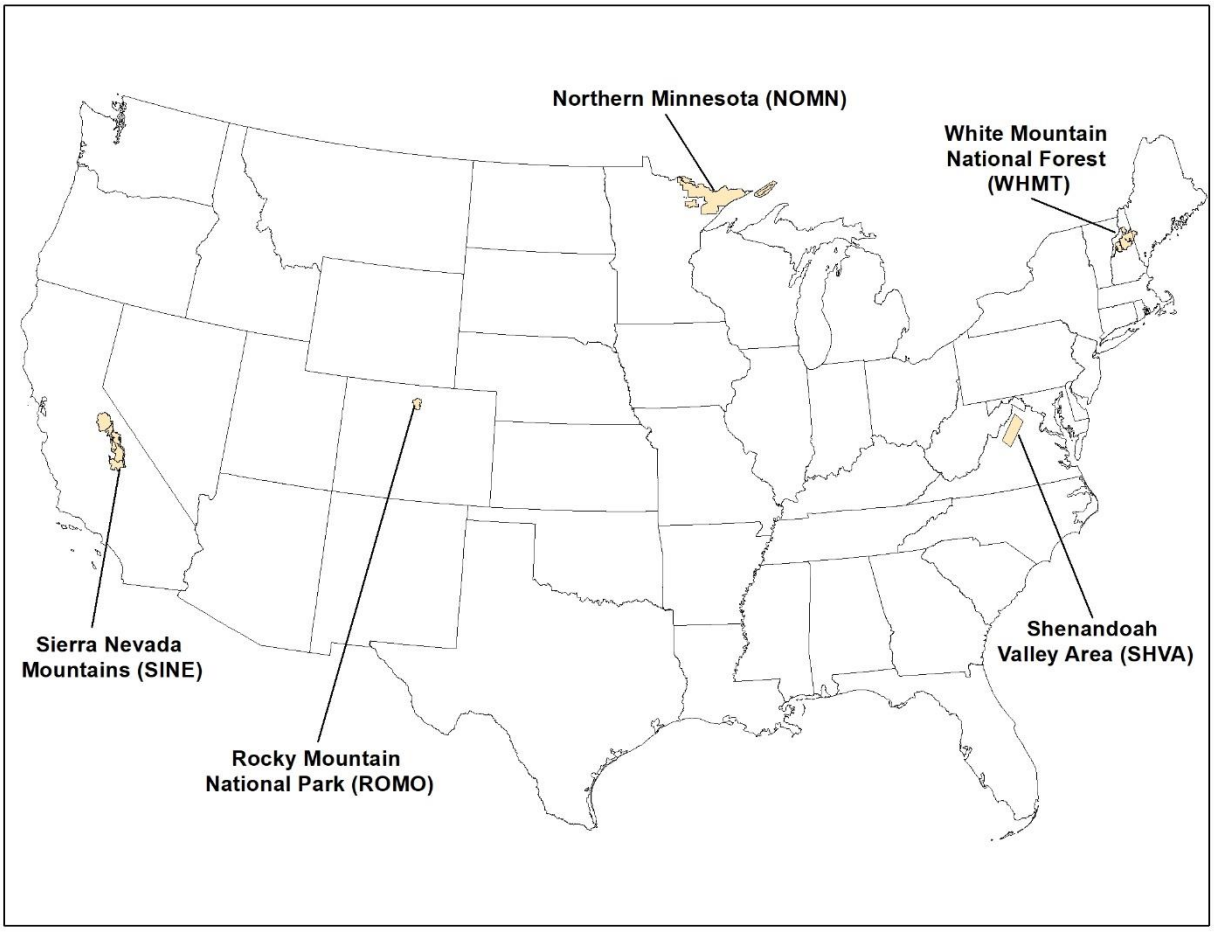

Figure 5A-52. Location of the case study areas.

Table 5A-40. Estimated annual deposition in five case study areas (2018-2020 average).

	Northern Minnesota	Shenandoah Valley	White Mountain National Forest	Rocky Mountain National Park	Sierra Nevada Mountain
S Deposition, kg S/ha-yr mean (range)	1.0 (0.8 – 1.5)	1.6 (1.1 – 2.0)	1.3 (1.0 – 1.6)	0.7 (0.4 -0.9)	1.0 (0.5 – 1.9)
N Deposition, kg S/ha-yr mean (range)	5.7 (3.7 – 6.9)	9.4 (6.6 – 13.4)	4.9 (3.7 – 6.1)	4.5 (3.2 – 6.1)	6.7 (2.3 - 31.8)
NH_x, as fraction of N mean (range)	0.6 (0.6 - 0.7).	0.7 (.5 - 0.8)	0.5 (0.5-0.5)	0.6 (0.5 - 0.6)	0.5 (0.3 - 0.8)
NH_3, as fraction of N mean (range)	0.3 (0.03 - 0.5).	0.5 (0.2 - 0.8)	0.2 (0.2-0.3)	0.3 (0.2 - 0.3).	0.2 (0.05 - 0.6).

NOTE: All estimates from TDEP. The fraction of dry NH_3 is not available from TDEP and was calculated as Dry NH_3 deposition/Total N Deposition.

5A.2.3.1 Descriptive Information for Case Study Areas

A broad sampling of ecoregions in the U.S. (as specified by the level I classification, Omernik 1987; Omernik and Griffith, 2014) are represented by the five case studies, as are a wide array of land uses (based on the 2016 National Land Cover Database [NLCD]).[14] While some of the case study areas fall within a single ecoregion, parts of some other areas are in a second ecoregion. Some land cover types are much more widespread than others (e.g., evergreen forests) but less common land cover types are represented as well. All of the natural land cover types were represented at some level by the case study areas. This excludes the four developed land cover types and two agricultural types, although the latter two were represented to some degree in SHVA. As noted above some land cover types, particularly forests are very high percentages of the total area which is what is seen in the case study areas. But less common land cover types like wetlands are also represented within the case study areas as well, including perennial ice and snow areas. This indicates that overall, the case study areas provide a relative broad coverage of the land cover types. The ecoregions and land uses of the five case studies are summarized briefly below, beginning with the three eastern areas and followed by the two in the West. Of the three eastern case study areas, the White Mountain National Forest (WHMT case study) occurs in two different ecoregions: Northeastern Coastal Zone (8.1.7) and Northern Appalachian and Atlantic Maritime Highlands[15] (5.3.1) (Figure 5A-53). Within the WHMT are

[14] We used the 2016 National Land Cover Database (NLCD), developed by the U.S. Geological Survey, in partnership with several other federal agencies, to assess the general landcover represented in each of the case study areas. The National Land Cover Database (NLCD) provides nationwide data on land cover and land cover change at a 30m resolution with a 16-class legend based on a modified Anderson Level II classification system (Anderson et al., 1976; Jin et al., 2019; https://www.mrlc.gov/data/nlcd-2016-land-cover-conus).

[15] The U.S. portion of the ecoregion is referred to as Northeastern Highlands as shown in Figure 5A-52.

six wilderness areas and two are Class I areas (Great Gulf and Presidential Range-Dry River Wilderness Areas, established in 1964 and 1975, respectively). The WHMT case study location is dominated by forested areas, with a mixture of evergreen, deciduous and mixed forest cover (Figure 5A-54; Table 5A-41).

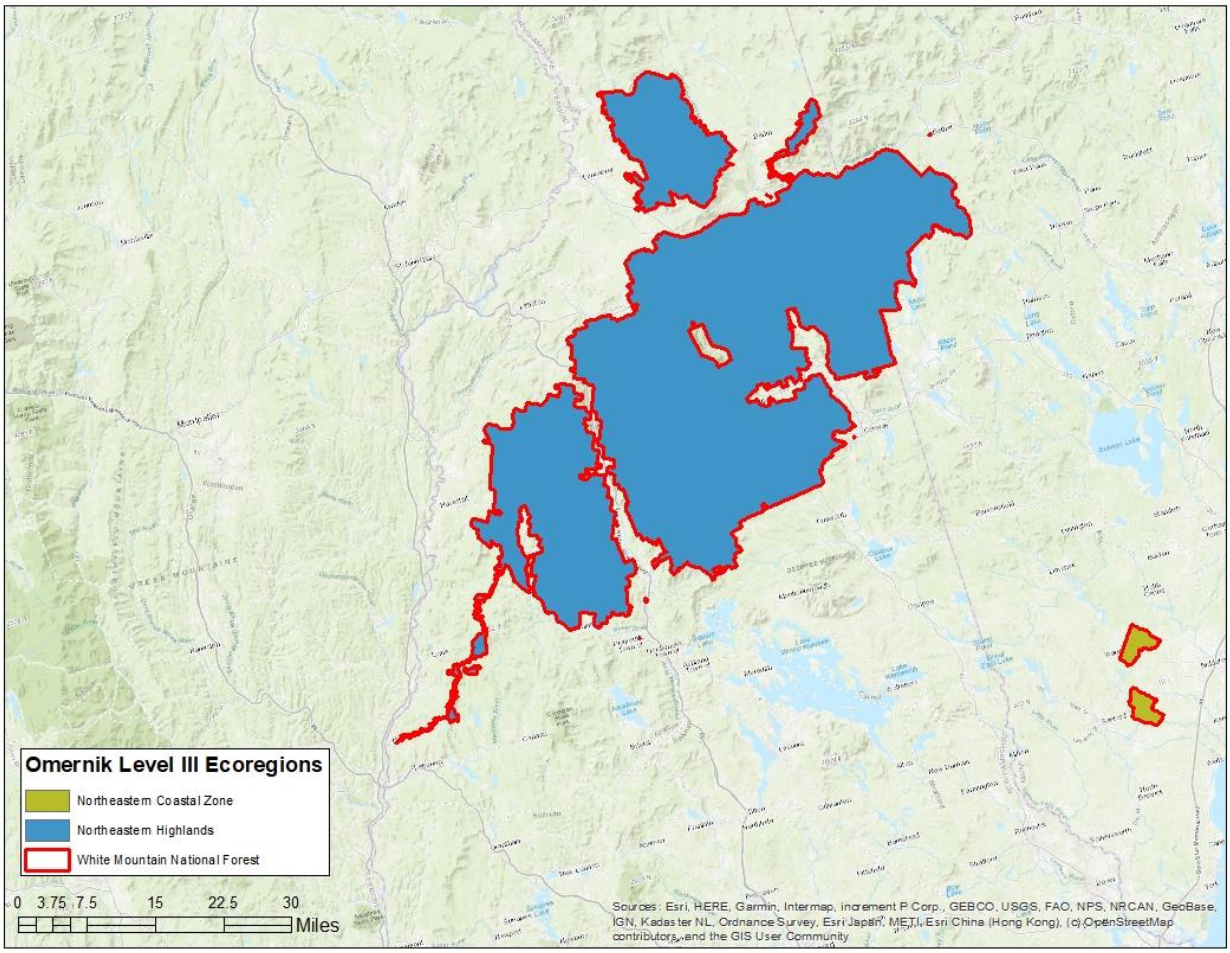

Figure 5A-53. Level III ecoregions in which WHMT occurs.

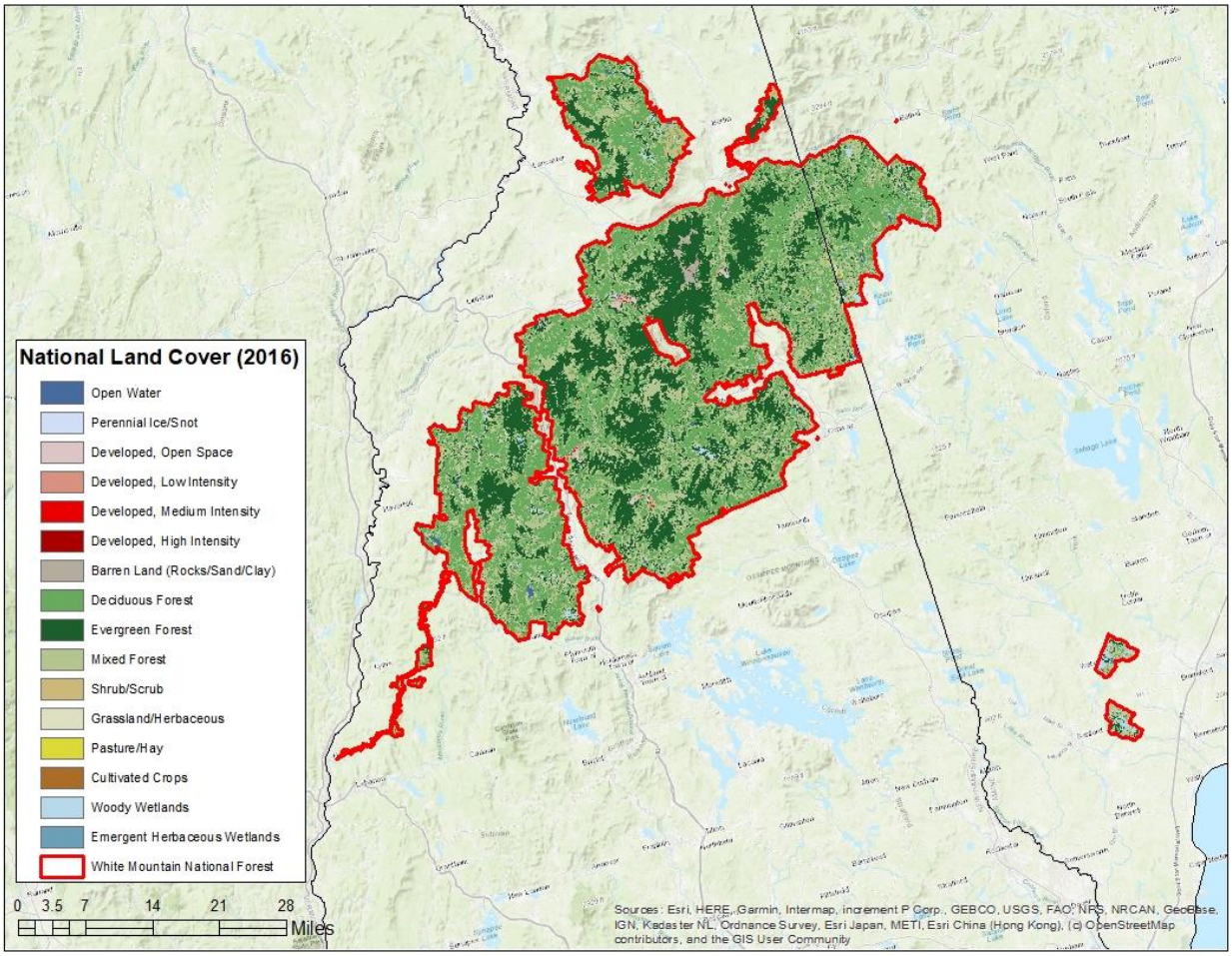

Figure 5A-54. Types of land cover in WHMT based on 2016 NLCD.

Table 5A-41. Distribution of land cover types in WHMT based on the 2016 NLCD.

Land Cover	Great Gulf Wilderness	Presidential Range Wilderness	Total
Open Water	0%	<0.1%	0.3%
Perennial Ice/Snow	0%	0%	0%
Developed, Open Space	0.9%	0%	1.4%
Developed, Low Intensity	0.8%	0%	0.3%
Developed, Medium Intensity	0.3%	0%	0.1%
Developed, High Intensity	<0.1%	0%	<0.1%
Barren Land (Rock/Sand/Clay)	13.0%	1.0%	0.4%
Deciduous Forest	4.7%	20.3%	34.9 %
Evergreen Forest	67.6%	58.0%	28.9%
Mixed Forest	10.8%	19.5%	29.9%
Shrub/Scrub	0.5%	0.8%	1.5%
Grassland/Herbaceous	1.4%	0.4%	0.4%
Pasture/Hay	0%	<0.1%	0.2%
Cultivated Crops	0%	0%	<0.1%
Woody Wetlands	0%	<0.1%	1.5%
Emergent Herbaceous Wetlands	0%	0%	0.1%

The SHVA case study, which includes the city of Harrisonburg and smaller towns, occurs in three different ecoregions: Blue Ridge (8.4.4), Northern Piedmont (8.3.1) and Ridge and Valley (8.4.1) (Figure 5A-55). This case study area is dominated by deciduous forest (~44%) and mixed forests (~14%) but also includes nearly 29% of the land cover in pasture, hay, and cultivated crops (Figure 5A-56, Table 5A-42). This case study area includes a National Park, a National Forest and two Virginia State Parks (Shenandoah River and Seven Bends State Parks). In Shenandoah National Park, nearly 84% of the area is deciduous forests. In George Washington National Forest nearly 86% of the area is deciduous or mixed forest. This area also includes portions of the Blue Ridge Parkway and Appalachian National Scenic Trail, managed by the National Park Service.

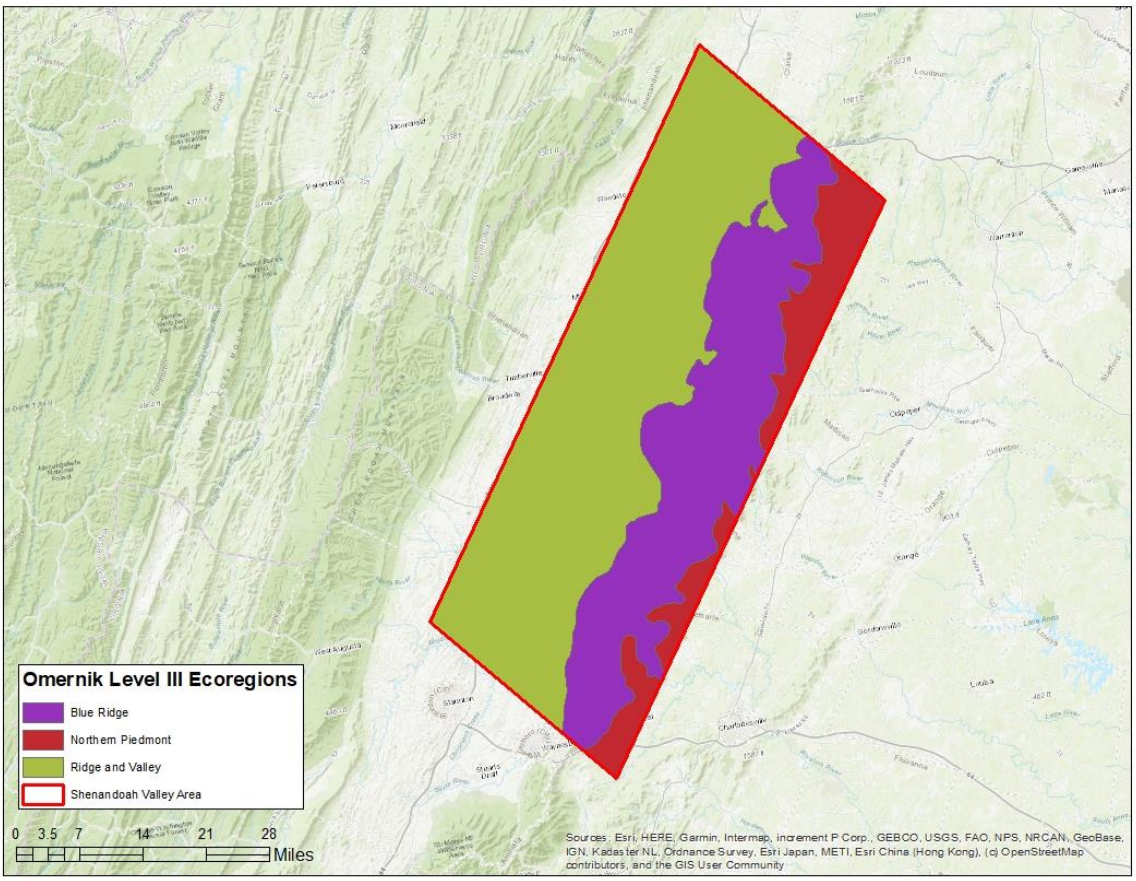

Figure 5A-55. Level III ecoregions in which SHVA occurs.

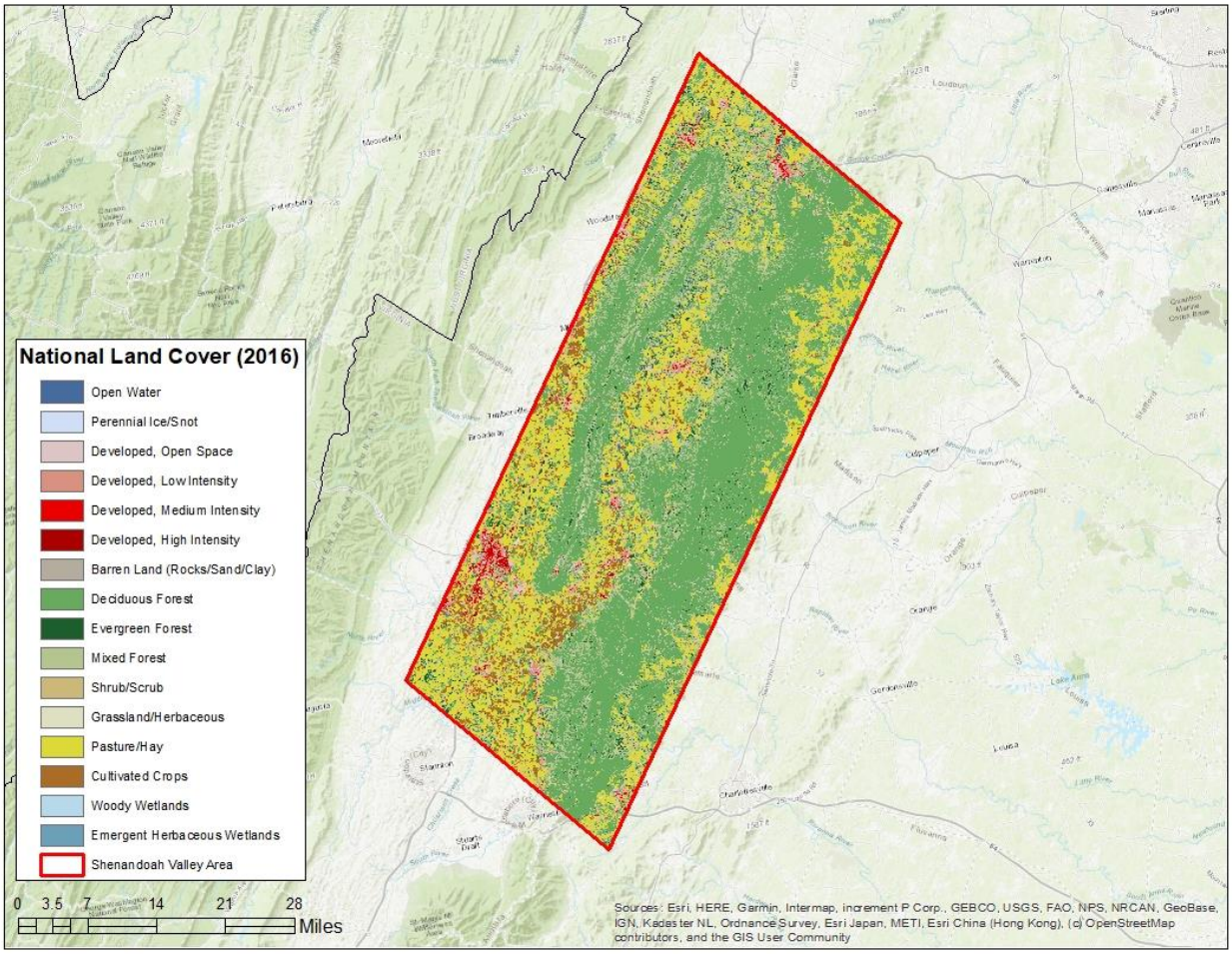

Figure 5A-56. Types of land cover in SHVA based on the 2016 NLCD.

Table 5A-42. Distribution of land cover types in SHVA based on the 2016 NLCD.

Land Cover	Shenandoah National Park	Washington National Forest	Virginia State Parks	Total
Open Water	0%	0.5%	1.1%	0.6%
Perennial Ice/Snow	0%	0%	0%	0%
Developed, Open Space	1.2%	2.9%	3.2%	4.9%
Developed, Low Intensity	0.1%	0.2%	0.1%	2.5%
Developed, Medium Intensity	<0.1%	<0.1%	<0.1%	0.7%
Developed, High Intensity	0%	<0.1%	0%	0.3%
Barren Land (Rock/Sand/Clay)	<0.1%	<0.1%	<0.1%	0.1%
Deciduous Forest	84.0%	60.9%	50.2%	44.5%
Evergreen Forest	1.6%	2.6%	0.7%	2.7%
Mixed Forest	12.7%	25.6%	32.2%	14.4%
Shrub/Scrub	0.1%	0.2%	0.6%	0.2%
Grassland/Herbaceous	0.3%	0.7%	0.2%	0.5%
Pasture/Hay	<0.1%	6.0%	9.7%	24.5%
Cultivated Crops	0%	0.2%	2.0%	4.1%
Woody Wetlands	<0.1%	<0.1%	0%	<0.1%
Emergent Herbaceous Wetlands	<0.1%	0%	0%	<0.1%

The NOMN case study, which is composed of Voyageurs National Park and Superior National Forest, and borders Minnesota and Canada, occurs in two different ecoregions: Northern Lakes and Forest (5.2.1) and Northern Minnesota Wetlands (5.2.2) (Figure 5A-57). The Northern Minnesota area has approximately 10% open water and 50% evergreen and mixed forest areas (Figure 5A-58 and Table 5A-43). The area also has a significant amount of woody wetlands (~29%). Voyageurs National Park and Boundary Waters-Canoe Wilderness have more open water than the entire area, but generally similar patterns of land cover.

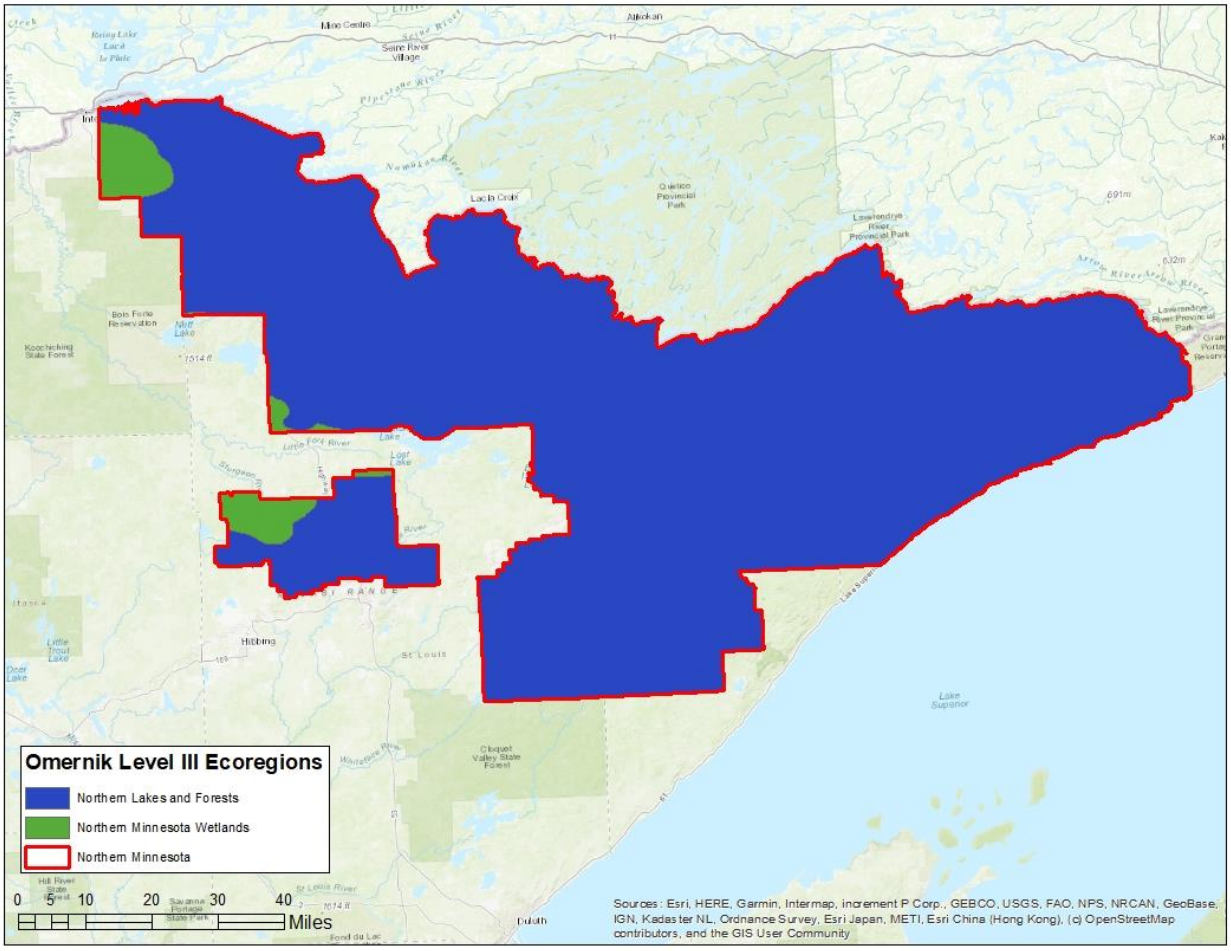

Figure 5A-57. Level III ecoregions in which NOMN occurs.

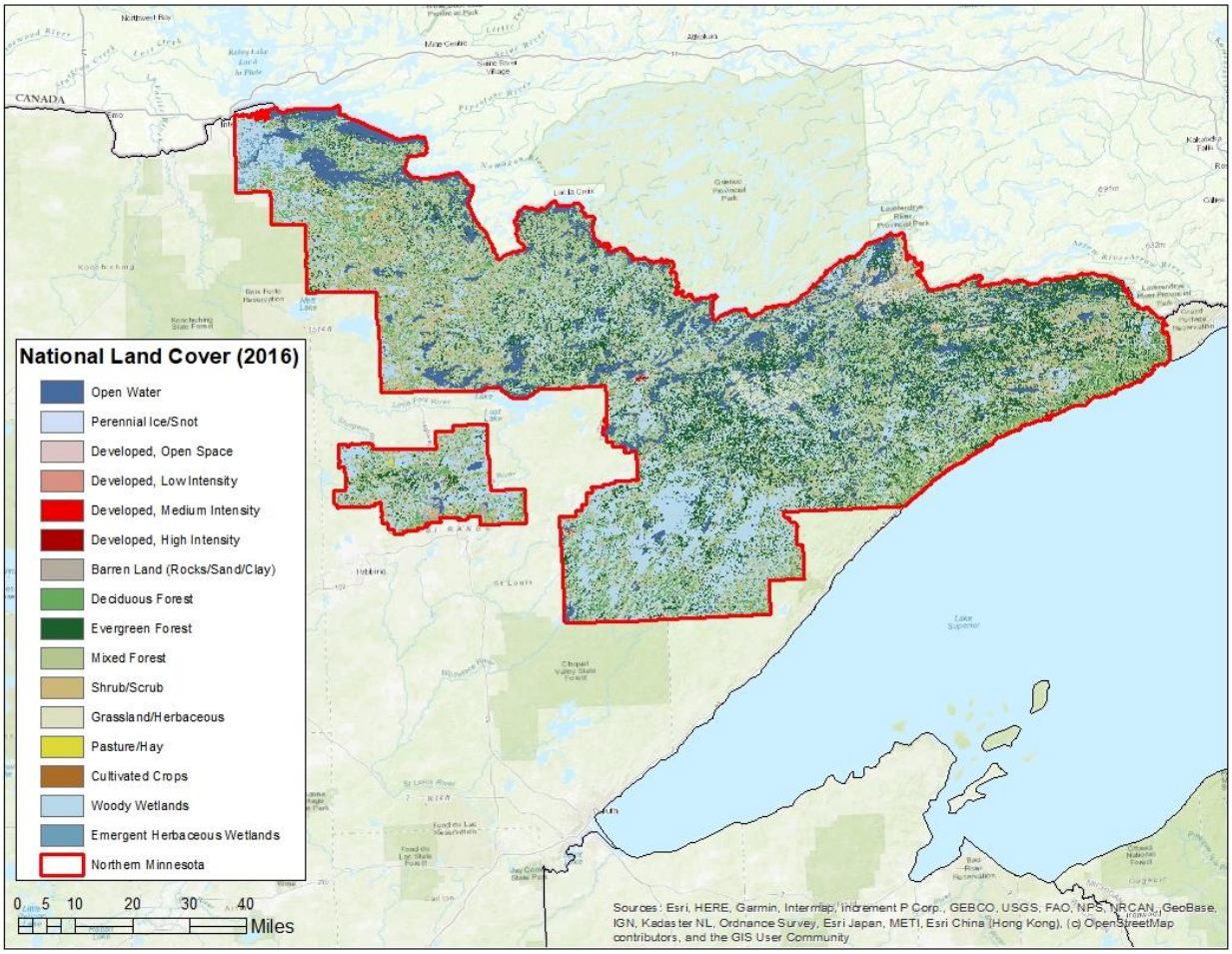

Figure 5A-58. Types of land cover in NOMN based on the 2016 NLCD.

Table 5A-43. Distribution of land cover types in NOMN based on the 2016 NLCD.

Land Cover	Voyageurs National Park	Superior National Forest	Boundary Waters-Canoe Wilderness	Total
Open Water	36.6%	9.5%	18.2%	10.8%
Perennial Ice/Snow	0%	0%	0%	0%
Developed, Open Space	<0.1%	1.1%	0.1%	1.1%
Developed, Low Intensity	<0.1%	0.2%	0%	<0.1%
Developed, Medium Intensity	<0.1%	0.1%	0%	0.1%
Developed, High Intensity	0%	<0.1%	0%	<0.1%
Barren Land (Rock/Sand/Clay)	0%	0.2%	<0.1%	0.2%
Deciduous Forest	7.7%	7.0%	3.2%	7.0%
Evergreen Forest	8.6%	16.3%	20.6%	15.9%
Mixed Forest	28.1%	26.2%	28.9%	26.3%
Shrub/Scrub	0.9%	5.2%	2.4%	5.0%
Grassland/Herbaceous	0.7%	2.6%	4.6%	2.5%
Pasture/Hay	<0.1%	0.1%	0%	0.1%
Cultivated Crops	0%	<0.1%	0%	0%
Woody Wetlands	12.5%	30.0%	20.6%	29.2%
Emergent Herbaceous Wetlands	4.9%	1.7%	1.5%	1.8%

In the western U.S. the northwestern forested mountain ecoregions are relatively well represented as is the Sierra. The Great Plains and intermountain west are not well represented by our case study areas. The ROMO case study occurs in only one ecoregion, the Northwestern Forested Mountains, Southern Rockies ecoregion (6.2.14). This case study is composed of Rocky Mountain National Park, which was designated one of the first World Biosphere Reserves by the United Nations Educational, Scientific and Cultural Organization in 1977, is a Class I area under the Clean Air Act.[16] The dominant land cover in Rocky Mountain National Park is evergreen forest (~54%), but there is also substantial shrub (~22%) and herbaceous/grasslands (~13%) (Figure 5A-59 and Table 5A-44). The area also has high elevation barren areas (5%) and some areas with perennial snow.

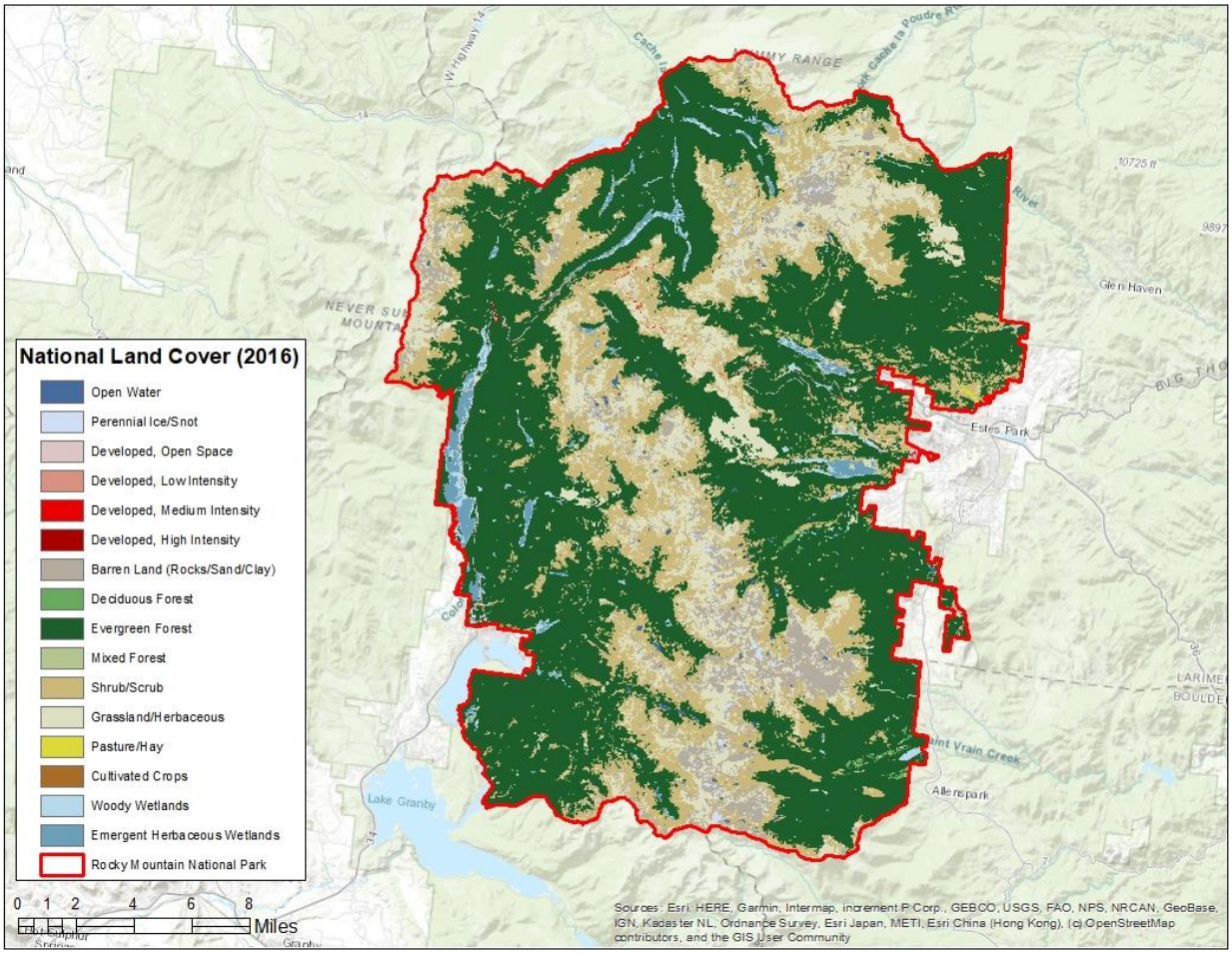

Figure 5A-59. Types of land cover in ROMO based on the 2016 NLCD.

[16] https://www.nps.gov/romo/learn/management/statistics.htm

Table 5A-44. Distribution of land cover types in ROMO based on the 2016 NLCD.

Land Cover	Total
Open Water	0.3%
Perennial Ice/Snow	0.6%
Developed, Open Space	0.3%
Developed, Low Intensity	0.1%
Developed, Medium Intensity	<0.1%
Developed, High Intensity	<0.1%
Barren Land (Rock/Sand/Clay)	5.2%
Deciduous Forest	0.3%
Evergreen Forest	54.7%
Mixed Forest	<0.1%
Shrub/Scrub	22.2%
Grassland/Herbaceous	13.3%
Pasture/Hay	<0.1%
Cultivated Crops	0%
Woody Wetlands	1.4%
Emergent Herbaceous Wetlands	1.6%

The SINE case study area incorporates a large area within the Sierra Nevada Mountains of eastern Calif, such that it occurs in four different ecoregions: Central Basin and Range (10.1.5), Mojave Basin and Range (10.2.1), Sierra Nevada (6.2.12) and Southern and Central California Chapparal and Oak Woodland (11.1.1) (Figure 5A-60). The Sierra Nevada area is mostly dominated by evergreen forests (~39%) and shrub/scrub (~38%) (Figure 5A-61, Table 5A-45). The case study area extends from Yosemite national Park in the north to Sequoia National Park in the south and includes Kings Canyon National Park (contiguous with Sequoia National Park)[17] and two wilderness areas that connect these parks (Ansel Adams and John Muir Wilderness Areas, both of which are Class I areas). The John Muir and Pacific Crest Trails pass through these areas. Across the five parks and two wilderness areas the dominant land cover is similar, with primarily differences in the percent cover of barren lands.

[17] The area comprised of these two parks was designated a UNESCO Biosphere Reserve in 1976 (https://www.nps.gov/seki/learn/news/quick-fact-sheet.htm), and Yosemite National Park was designated a World Heritage site in 1984 (https://www.nps.gov/articles/nps-geodiversity-atlas-yosemite-national-park.htm).

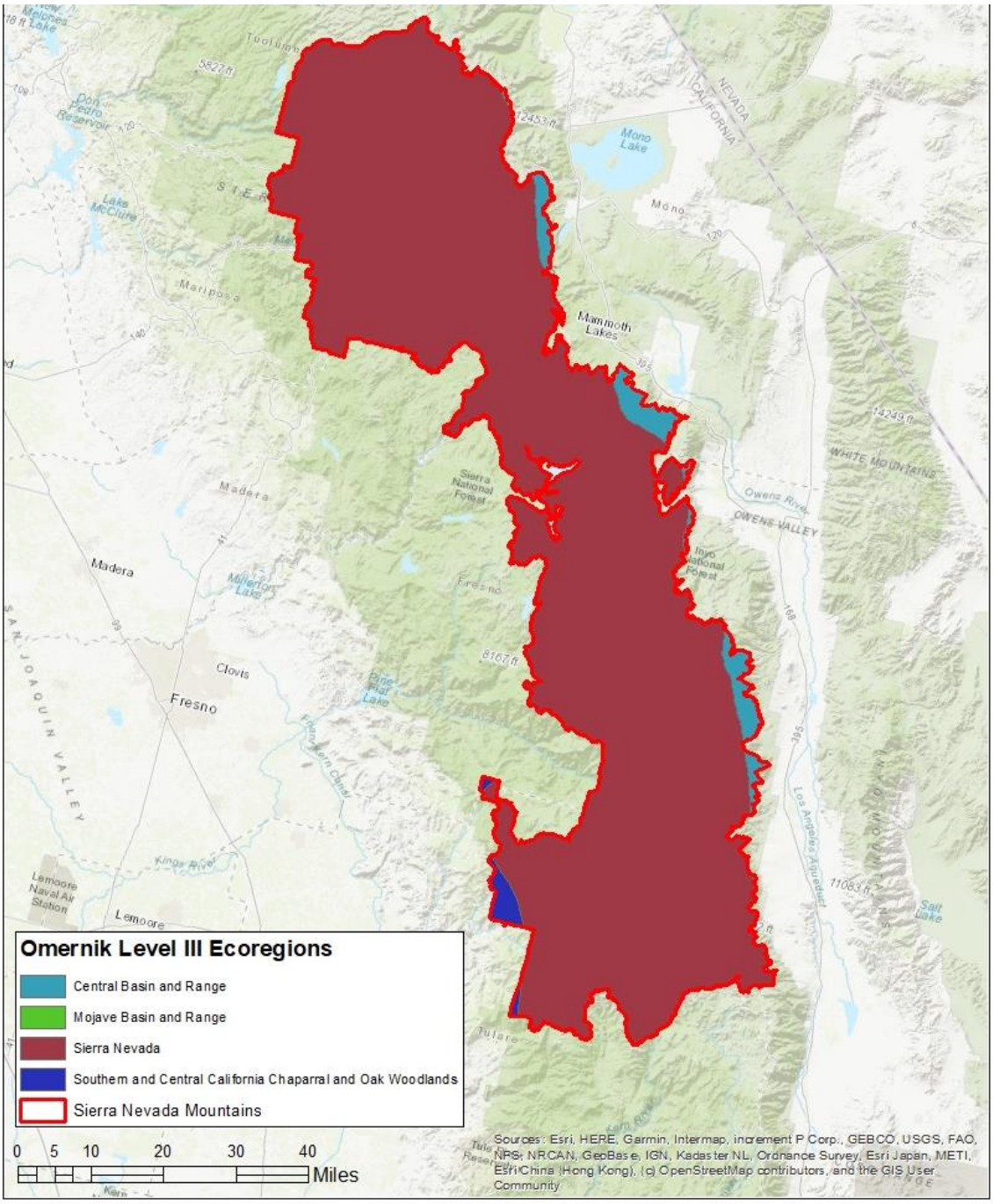

Figure 5A-60. Level III ecoregions in which SINE occurs.

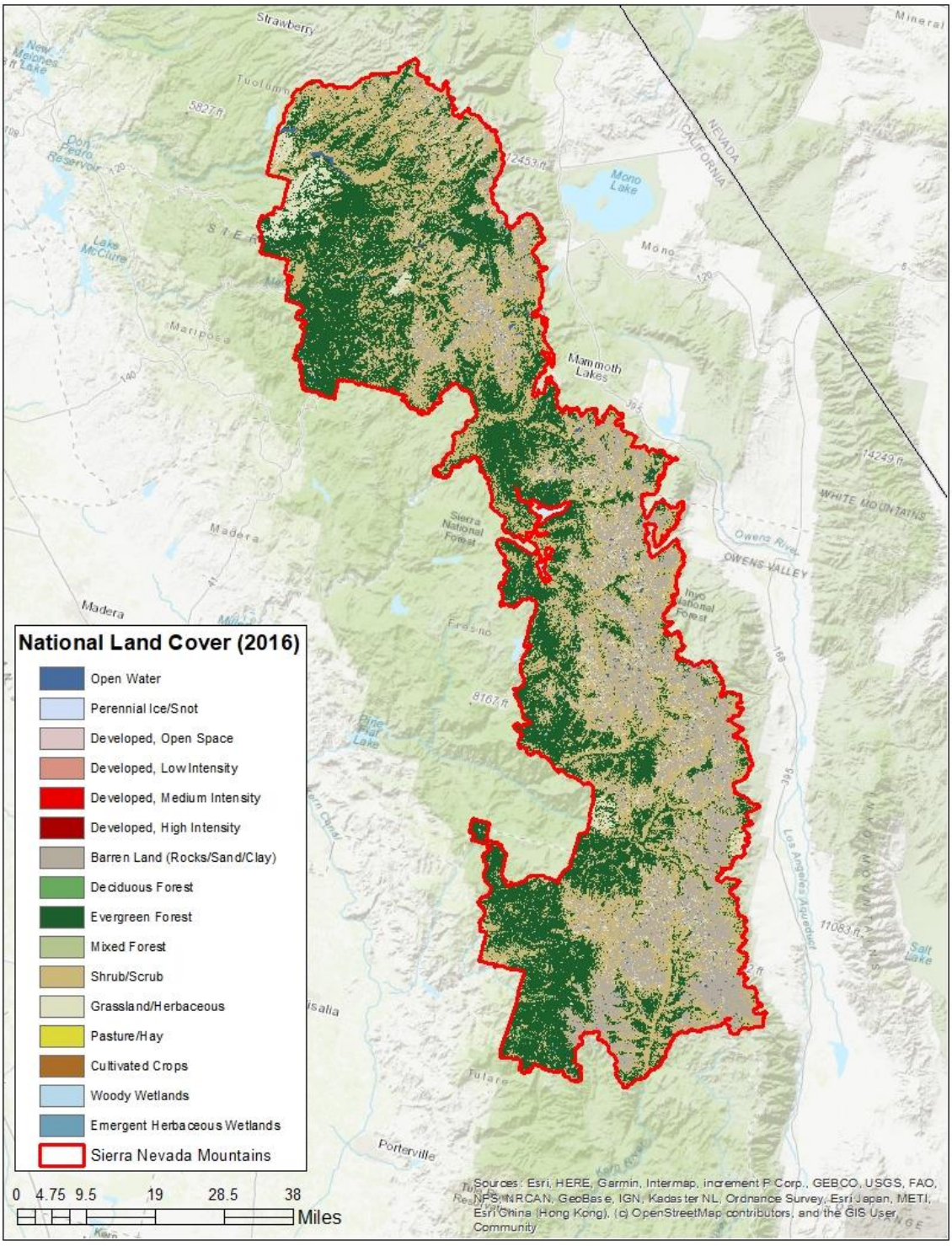

Figure 5A-61. Types of land cover in SINE based on the 2016 NLCD.

Table 5A-45. Distribution of land cover types in SINE based on the 2016 NLCD.

Land Cover	Yosemite National Park	Sequoia National Park	Kings Canyon National Park	Ansel Adams Wilderness	John Muir Wilderness	Total
Open Water	1.0%	0.5%	1.2%	0.9%	1.1%	1.0%
Perennial Ice/Snow	0.1%	0.1%	0.2%	0.3%	0.3%	0.2%
Developed, Open Space	0.3%	0.2%	0.1%	<0.1%	<0.1%	0.1%
Developed, Low Intensity	<0.1%	<0.1%	<0.1%	0%	0%	<0.1%
Developed, Medium Intensity	0%	0%	0%	0%	0%	0%
Developed, High Intensity	0%	0%	0%	0%	0%	0%
Barren Land (Rock/Sand/Clay)	5.0%	21.2%	22.5%	8.3%	21.5%	15.3%
Deciduous Forest	<0.1%	0.6%	0.1%	0.1%	0.1%	0.1%
Evergreen Forest	47.4%	43.1%	28.6%	43.3%	31.1%	38.8%
Mixed Forest	0.1%	0.8%	0.1%	0.14%	0.1%	0.2%
Shrub/Scrub	37.6%	30.5%	41.4%	44.8%	40.1%	38.4%
Grassland/Herbaceous	7.8%	2.5%	5.6%	1.9%	5.2%	5.3%
Pasture/Hay	0%	0%	0%	0%	0%	0%
Cultivated Crops	0%	0%	0%	0%	0%	0%
Woody Wetlands	0.3%	0.2%	0.1%	0.1%	0.2%	0.2%
Emergent Herbaceous Wetlands	0.5%	0.4%	0.1%	0.2%	0.4%	0.3%

5A.2.3.2 Case Study Air Quality

To relate air quality to deposition, we identified a set of monitors within an area of influence (maximum radius of 500 km), described in section 5A.2.3.2.2 below.[18] The monitors identified included monitors collecting and chemically speciating particulate matter with mass median diameter of 2.5 microns ($PM_{2.5}$) and Federal Reference Method (FRM) $PM_{2.5}$ monitors (used to inform compliance with the NAAQS) for which there are data in the EPA's Air Quality System (AQS) database, and Clean Air Status and Trends Network (CASTNET) monitoring sites.[19] Some monitors are sited specifically for tracking local sources and are not representative of regional conditions. Monitors that are designated in AQS with a measurement scale listed as "microscale" or "middle scale", which indicates that the monitor is strongly influenced by local emission sources, are also excluded from this analysis. Only a few monitors were removed from consideration in this step. Among the FRM $PM_{2.5}$ monitors, the monitor with the maximum annual average $PM_{2.5}$ design value is selected. The air quality monitors used in each case study area are listed in Table 5A-46 and shown in Figures 5A-62 to 5A-66. These included National Atmospheric Deposition Program (NADP) wet deposition monitors that are part of the National Trends Network (NTN), described further in Chapter 2, section 2.3.4, as well as CASTNET total S and total NO_3^- monitors, and $PM_{2.5}$ chemical composition monitors.

[18] This approach for relating monitor locations and concentrations to deposition estimates (described further in section 5A.2.3.2.2) differs from the approaches for analyses presented in Chapter 6.

[19] These different monitor types and their networks are described in Chapter 2, sections 2.3.3 and 2.3.4.

Table 5A-46. Air quality and wet deposition monitors used to assess the relationship between air concentration and deposition, and trends.

Case Study Areas	Wet Deposition (NADP)	Total S and Total N (CASTNET)	$PM_{2.5}$ Mass (FRM)	$PM_{2.5}$ Composition
Northern Minnesota	NTN MN32: Voyageurs Nat Pk, MN NTN MN18: Fernberg, MN NTN MN08: Hovland, MN NTN MN99: Wolfland, MN NTN MN16: Marcell Exp Forest NTN MI97: Isle Royale, MI	VOY413: Voyageurs National Park, MN RED004: Red Lake Band of Chippewa Indians	27-053-0963: Minneapolis, MN	27-137-0034: Voyageurs National Park, MN 27-075-0005: Boundary Waters, MN 26-083-9000: Isle Royale National Park, MI 27-053-0963: Minneapolis, MN
Rocky Mountain National Park (RMNP)	NTN CO98: RMNP-Loch Vale NTN CO19: RMNP-Beaver Meadows	ROM406: RMNP	08-031-0002: Denver, CO	08-069-0007: RMNP 08-123-0008: Platteville, CO 08-001-0006: Commerce City, CO
Shenandoah Valley Area	NTN VA28: Shenandoah Nat Park NTN MD08: Frostburg, MD NTN WV18: Parsons, WV NTN VA00: Charlottesville, VA NTN MD99: Beltsville, MD	SHN418: Shenandoah National Park PAR207: Parsons, WV LRL117: Laurel Hill, PA ARE128: Arendtsville, PA BEL116: Beltsville, MD	42-003-0008: Pittsburgh, PA	51-113-0003: Shenandoah National Park 54-093-9000: Tucker County, WV 24-023-0002: Grantsville, MD 42-129-0008: Greensburg, PA 42-003-0008: Pittsburgh, PA 42-125-5001: Washington County, PA 24-033-0030: Beltsville, MD 39-099-0014: Youngstown, OH
Sierra Nevada Mountains	NTN CA99: Yosemite National Park NTN CA75: Sequoia National Park	YOS404: Yosemite Nat Pk SEK430: Sequoia National Park - Ash Mountain SEK402: Sequoia National Park - Lookout Pt	06-107-2002: Visalia, CA	06-107-1001: Sequoia National Pk-Ash Mtn 06-043-0003: Yosemite National Park 06-107-2002: Visalia, CA 06-099-0005: Modesto, CA 06-029-0014: Bakersfield, CA
White Mountain National Forest	NTN NH02: Hubbard Brook, NH	WST109: Woodstock, NH HBR183: Hubbard Brook, NH	25-025-0042: Boston, MA	33-007-4002: Coos County, NH 33-011-5001: Peterborough, NH 50-007-0012: Burlington, VT 25-015-4002: Ware, MA 25-025-0042: Boston, MA

Figure 5A-62. Monitoring sites used for NOMN to analyze relationships and trends.

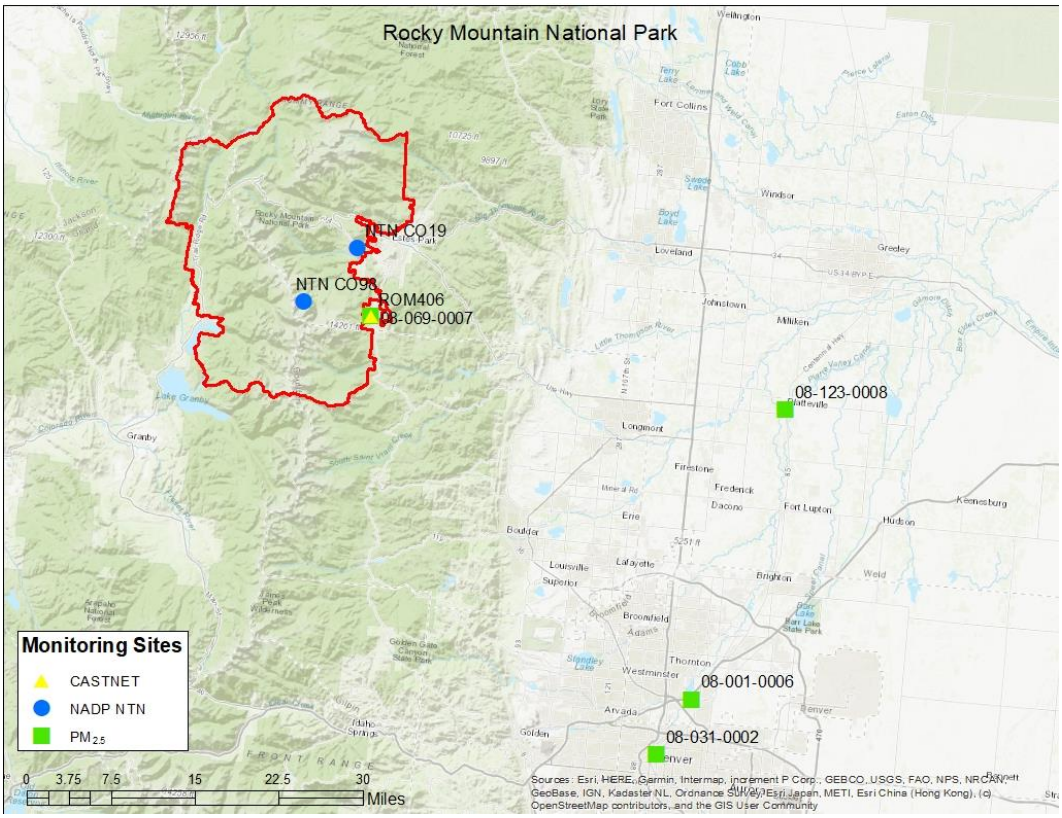

Figure 5A-63. Monitoring sites used for ROMO to analyze relationships and trends.

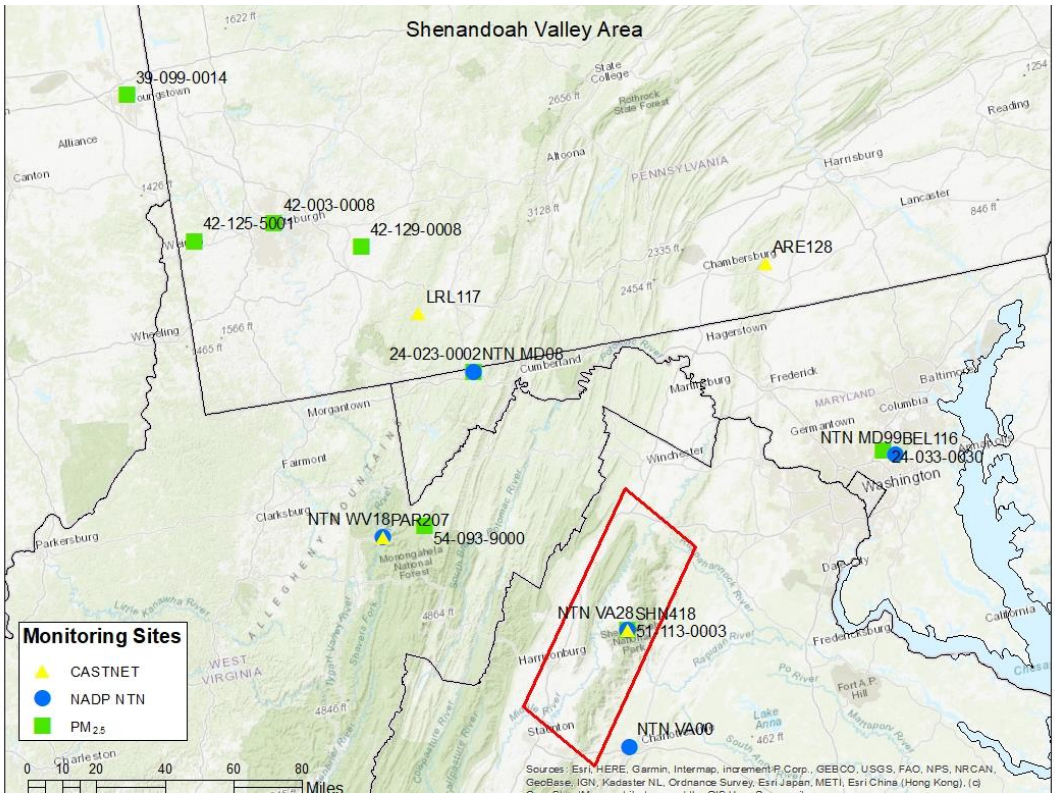

Figure 5A-64. Monitoring sites used for SHVA to analyze relationships and trends.

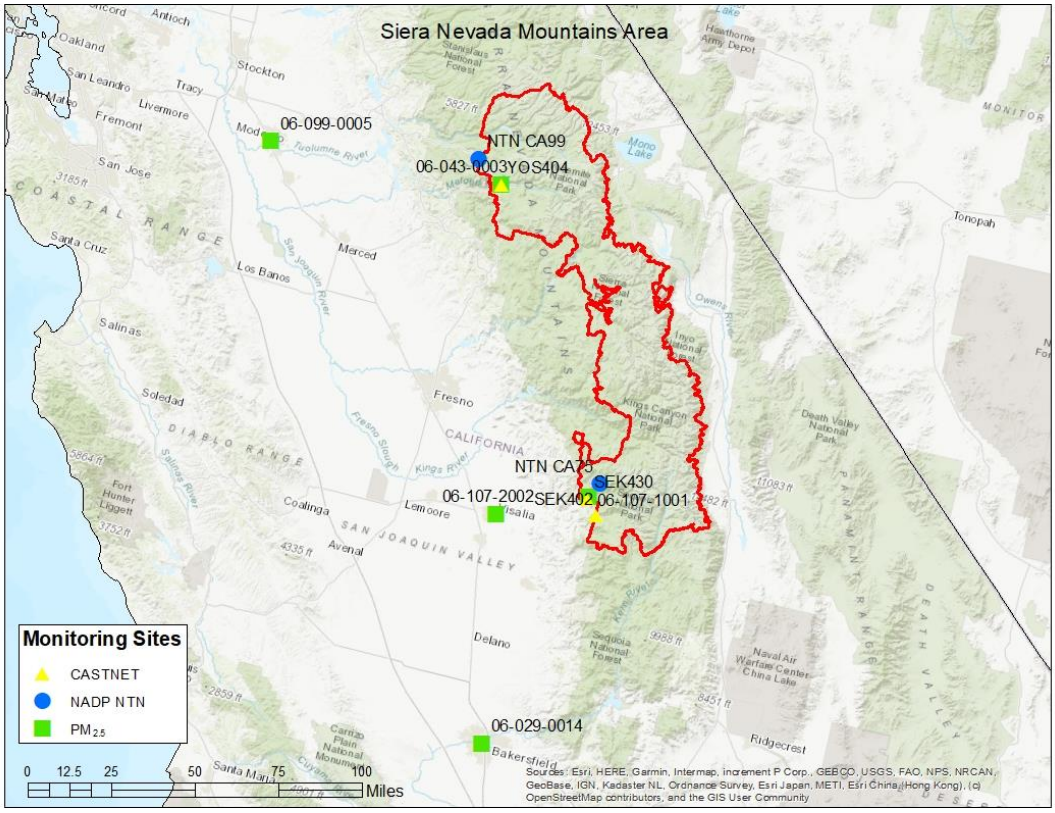

Figure 5A-65. Monitoring sites used for SINE to analyze relationships and trends.

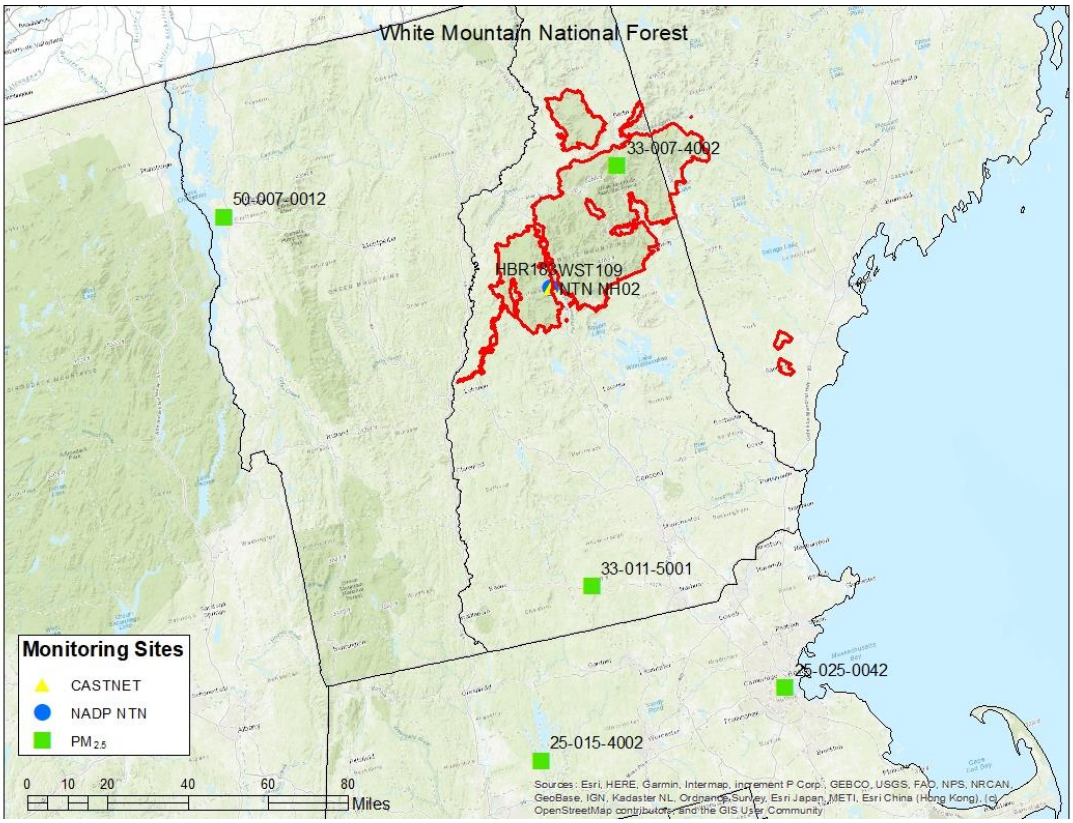

Figure 5A-66. Monitoring sites used for WHMT to analyze relationships and trends.

5A.2.3.2.1 Correlation of Deposition and Air Quality

In this subsection, we consider the extent to which the maximum annual average $PM_{2.5}$ concentration in the area of influence is linked to the annual deposition in the case study area by analyzing the correlation between annual average $PM_{2.5}$ (at the FRM monitor with highest concentration) and annual total S and N deposition, as estimated by TDEP methods, averaged spatially across the case study area.

Table 5A-47 shows the correlation between $PM_{2.5}$ at the maximum monitor and the S and N total deposition estimated using the TDEP method. The highest correlations, for both S and N deposition are seen for the two farthest east areas. A high correlation is also seen for S deposition in the northern Minnesota case study area, which has a less strong correlation for N deposition. Correlations are lower for both S and N in the two western locations; in the Sierra Nevada case study area, there is no discernable correlation between $PM_{2.5}$ and S deposition and the correlation coefficient for N deposition is below 0.5 (Table 5A-47).

Table 5A-47. Correlation coefficients for TDEP total deposition estimates with annual average concentrations at the $PM_{2.5}$ over the period 2000-2019.

Case Study Areas	Correlation between total S deposition and $PM_{2.5}$ mass	Correlation between total N deposition and $PM_{2.5}$ mass
Northern Minnesota	0.96	0.70
Rocky Mountain National Park	0.64	0.68
Shenandoah Valley Area	0.97	0.93
Sierra Nevada Mountains	-0.02	0.35
White Mountain National Forest	0.97	0.89

The relationship between air concentration and deposition depends on several factors, including the chemical form of sulfur and nitrogen, the vertical distribution in the atmosphere, and the frequency of precipitation (See Chapter 2 and Chapter 6 of main document). Each of these vary across the different case study areas. In the eastern U.S., where SO_2 and N oxides emissions have declined the most, measurements of $PM_{2.5}$ and wet deposition show a strong correlation. In the western U.S., where dry deposition and ammonia play a larger role, in some cases there is no correlation between measured wet deposition and surface $PM_{2.5}$ mass concentrations (see Chapter 6 of main document).

5A.2.3.2.2 Air Quality Scenarios

In this case study analysis, critical load exceedances were calculated for several air quality scenarios that reflected an area meeting the most controlling[20] current secondary NAAQS for that area (of the standards for SO_2, NO_2 and PM), which in all cases was that for $PM_{2.5}$. For each case study area, historic air quality was examined to find a time when the monitors within or near the area influencing the case study area[21] had design values that were within 10% of the current standard level (i.e., 15 $\mu g/m^3$). To examine how changing air quality and corresponding deposition could affect these estimated exceedances, additional scenarios for air quality at these locations in other years were also analyzed with the aim of having similar maximum $PM_{2.5}$ annual design values across the case studies. For these additional scenarios, time periods were selected where the highest monitor in the area of influence was within 10% of 12 $\mu g/m^3$ and 10 $\mu g/m^3$. For some locations, it was not possible to select a 3-year historical period as $PM_{2.5}$ concentrations, currently and in the past, have not been as high as the threshold for that scenario.

[20] The scenarios selected had air quality for which the $PM_{2.5}$ design value for the highest monitor was just equal to the current annual secondary standard.

[21] The premise for the area of influence definition is a region where a change in emissions could be expected to lead to a change in deposition at the case study area. A recent study of Class I areas found that the area of influence for nitrogen deposition can vary, and the radius was estimated to range between 500 – 1200 km (Lee et al., 2016). In identifying locations for emissions and concentrations in the area of influence that could be expected to be relevant, this analysis uses a maximum radius of 500 km.

For each of the selected air quality periods, the TDEP data were extracted for S and N. The air quality periods analyzed, and associated deposition levels are shown in Tables 5A-48 and 5A-49.

For one case study area, the Sierra Nevada, there is no historical period that is at or near the target $PM_{2.5}$ concentration (the annual average exceeds 15 ug/m^3 by more than 10% throughout the historical period). So it is not possible to use a historical dataset of deposition. As an alternative, the air quality and TDEP data (from the 2014-16 time period) were adjusted downwards to reflect each air quality scenario based on a regression-based analysis. An approximation of the change in deposition due to a change in $PM_{2.5}$ concentration at the maximum monitor is used based on a regression based on CMAQ modeling. A linear model was fit using air concentration and total (wet plus dry) deposition from a 21-year CMAQ model simulation. First, the air concentration and deposition values were normalized by their mean value. A linear model was fit to predict total deposition from air concentration. The slope was an estimate of the change in deposition due to a change in $PM_{2.5}$ concentration. The linear model was used to calculate the percent change in deposition (from the 2014-16 TDEP estimate) when the $PM_{2.5}$ concentration at the highest monitor was reduced from the 3-yr annual average concentration (in 2014-16) to 10 µg m^{-3}, 12 µg m^{-3}, and 15 µg m^{-3}. The prediction interval at each of these concentration levels was 40%, which indicates that there are a range of deposition levels that are consistent with these air concentration targets. The predicted deposition change for nitrogen and sulfur were different by a small amount, reflecting differences in the relationship between $PM_{2.5}$ and deposition. To clarify, this is not a prediction, but used as a plausible deposition scenario associated with maximum $PM_{2.5}$ concentrations for each target level.

Table 5A-48. The 3-year historical periods used for each case study area.

Case Study Area	TDEP years for 15 µg m-3	TDEP years for 12 µg m-3	TDEP years for 10 µg m-3
Northern Minnesota	$PM_{2.5}$ concentrations have not been this high	2000—2002	2007-2009
Rocky Mountain National Park	$PM_{2.5}$ concentrations have not been this high	$PM_{2.5}$ concentrations have not been this high	2000-2002
Shenandoah Valley	2005—2007	2009—2011	2014-2016
Sierra Nevada	S deposition: 2014-16 (multiplied by 0.70)* N deposition: 2014-16 (multiplied by 0.72)*	S deposition: 2014-16 (multiplied by 0.56)* N deposition: 2014-16 (multiplied by 0.57)*	S deposition: 2014-16 (multiplied by 0.46)* N deposition: 2014-16 (multiplied by 0.48)*
White Mountain National Forest	2000—2002	2005-2007	2009-2011

*The air quality and associated deposition estimates for Sierra Nevada case study are based on a linear regression-based "roll down" approach. The S and N deposition estimate assigned to each scenario (15, 12 and 10 µg/m^3) was derived by multiplying the factor shown here by the 2014-2016 TDEP estimates. The factors shown here were derived by multiplying the unit S or N deposition per unit $PM_{2.5}$ concentration (from a regression based on 21-year CMAQ simulation) by a factor equal to the air quality scenario $PM_{2.5}$ concentration (15, 12 and 10 µg/m^3) by the 2014-16 $PM_{2.5}$ concentration at the highest monitor.

Table 5A-49. For each 3-year period described in Table 5A-48, this is the estimated 3-year average annual average deposition, based on spatial averaging of TDEP dataset estimates across the case study area, for N and S deposition.

Case study	Mean N deposition, kg N ha^{-1} $year^{-1}$ (min-max)			Mean S deposition, kg S ha^{-1} $year^{-1}$ (min-max)		
	15 µg/m^3	12 µg/m^3	10 µg/m^3	15 µg/m^3	12 µg/m^3	10 µg/m^3
Northern Minnesota	NA	6.8 (4.1 - 8.7)	6.0 (3.7 - 8.1)	NA	3.4 (2.5 – 5.4)	3.0 (2.0 – 4.4)
Rocky Mountain National Park	NA	NA	6.6 (4.4 – 9.5)	NA	NA	2.3 (1.4 – 4.6)
Shenandoah National Park	11 (7.8 - 16)	8.7 (6.4 - 13)	8.3 (6.6 - 10)	10 (8.0 – 13.4)	5.0 (3.4 – 6.3)	3.1 (2.4 – 3.8)
Sierra Nevada*	4.9* (2.2 – 9.9)	3.9* (1.8 – 7.8)	3.3* (1.5 – 6.6)	0.80* (0.40 – 1.5)	0.64* (0.32 – 1.2)	0.53* (0.27 – 1.0)
White Mountain National Forest (New Hampshire)	7.6 (5.6 – 10)	6.7 (5.2 – 8.9)	5.2 (3.9 – 7.0)	7.2 (4.9 - 11)	7.1 (5.2 – 9.9)	3.8 (2.8 – 5.5)

*Increased uncertainty is recognized for this case study due the approach used to assign deposition estimates to the three air quality scenarios, which is described in the text and table above.

5A.2.3.3 Critical Loads Analysis

This section describes the findings of the case study analyses. Using the methodology described in section 5A.1.2 above, CL exceedances were estimated for waterbodies in the five case study areas based on the deposition estimates for three air quality scenarios. Aquatic CLs and exceedances are summarized in the subsections below using the following steps:

(1) CLs were extracted from the NCLD for each of the case study areas for the following ANC thresholds: 20, 30, and 50 µeq/L.

(2) CLs were summarized for each area in terms of the average, 70th and 90th percentile.

(3) Exceedances were calculated for each of the air quality scenarios for all three ANC thresholds for S only and N+S.

(4) The exceedances were summarized in terms of counts and percent of all CL sites in each study area.

5A.2.3.3.1 Case Study Waterbody Critical Loads

A total of 523 CLs were found in the 5 case study areas, excluding SHVA which had complete coverage (4977 CLs in total, with 704 CLs in sensitive sites). The ROMO, SINE, NOMN, and WHMT areas had 119, 139, 190, and 75 CLs respectively (Figure 5A-67). Despite the relatively high number of aquatic CLs for these five case studies, they do not represent a complete coverage of water resources. This summary of the CLs and exceedances only represents the waterbodies that have been modelled. Table 5A-50 provides average, 10th and

30th percentile CLs for S only for each case study areas in units of kg S/ha-yr. Table 5A-51 provides the same information for S only CLs in units of meq/m^2-yr, and also provide this information for S plus N CLs. Critical loads for S only were found to be similar for the waterbodies modelled among the case study areas with higher CL values for the lower ANC thresholds. Average S only CL values for an ANC threshold of 50 µeq/L range from 6.6 to 9.8 kg S/ha-yr or 41.3 to 61.3 meq/m^2-yr. For an ANC threshold of 20 µeq/L, the 10th percentile CLs for S only 1.8 to 7.1 kg S/ha-yr (3.6 to 7.1, excluding SINE).

Table 5A-50. Average, 10th and 30th percentile of CLs for kg S in each case study area.

	ANC of 20 µeq/L			ANC of 30 µeq/L			ANC of 50 µeq/L		
	Ave.	30th	10th	Ave.	30th	10th	Ave.	30th	10th
	Sulfur (S) only CLs (kg S/ha-yr)								
ROMO	9.5	5.4	3.6	8.5	4.5	2.6	6.6	2.7	0.5
SINE	12.0	4.1	1.8	11.0	2.8	0.5	9.3	0.6	0.1
NOMN	10.8	5.5	4.2	10.4	5.3	3.9	9.8	4.7	3.2
WHMT	10.6	6.9	4.4	9.6	6.1	3.3	7.4	4.1	0.7
SHVA	12.4	9.4	7.1	11.4	8.4	6.3	9.4	6.3	4.1
Note: CL units are kg S/ha-yr, and									

Table 5A-51. Average, 10th and 30th percentile of CLs for meq S in each case study area.

	ANC of 20 µeq/L			ANC of 30 µeq/L			ANC of 50 µeq/L		
	Ave.	30th	10th	Ave.	30th	10th	Ave.	30th	10th
	Sulfur (S) only CLs (meq/m^2-yr)								
ROMO	59.1	34.0	22.6	5.30	28.4	16.1	41.2	16.7	3.4
SINE	75.0	25.4	11.0	68.7	17.3	2.9	58.4	3.5	0.1
NOMN	67.4	34.5	26.0	65.3	32.4	29.1	61.0	29.3	20.1
WHMT	66.3	43.4	27.8	59.7	38.3	20.8	46.3	25.6	4.4
SHVA	77.4	58.9	44.6	71.3	52.4	39.1	59	39.5	25.8
10th and 30th percentiles refer to the 10th and 30th percentile lowest CL values.									

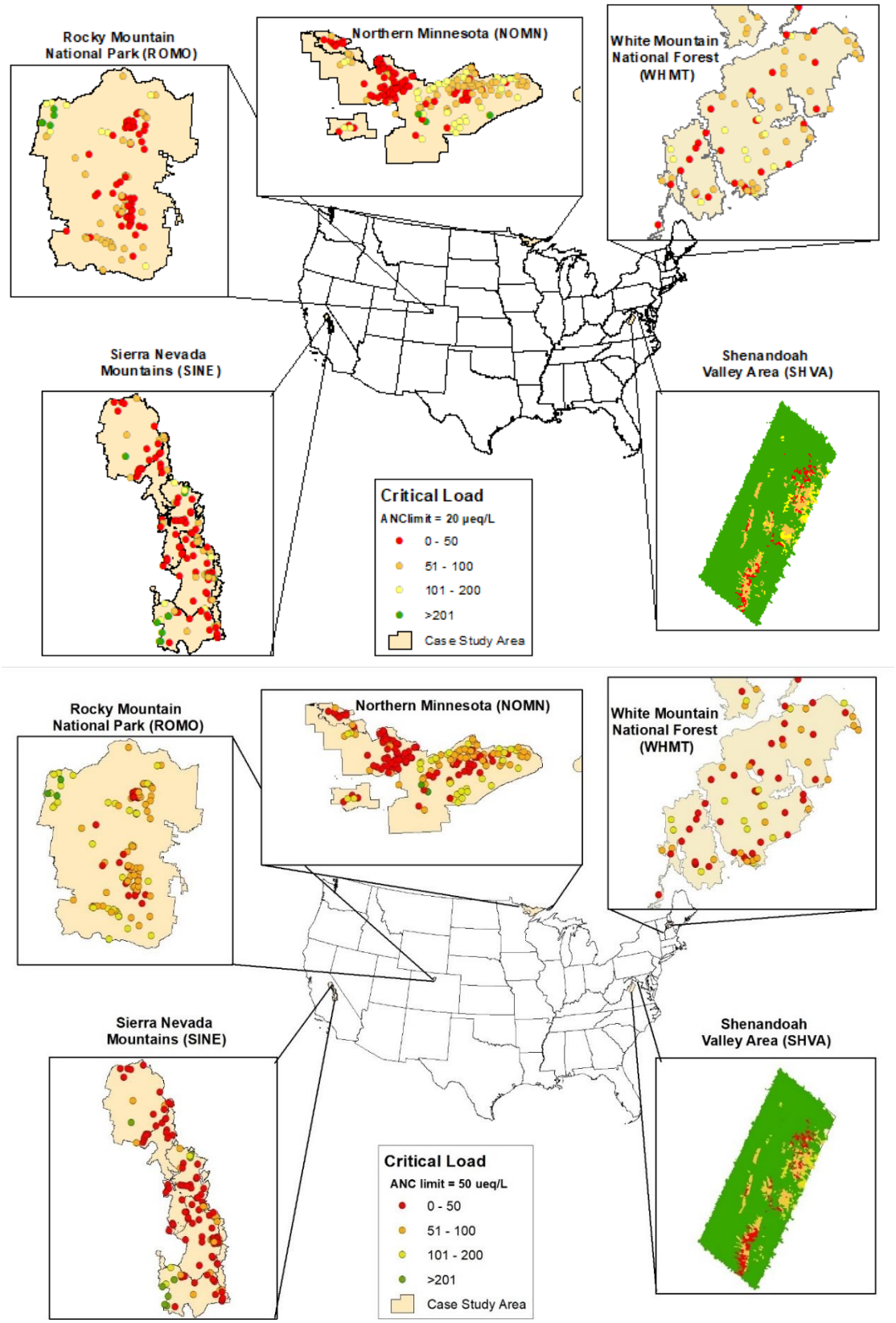

Figure 5A-67. Case study area CL maps for sulfur (meq/m^2-yr) using an ANC threshold of 20 µeq/L (upper) and 50 µeq/L (lower).

5A.2.3.3.2 Case Study Critical Load Exceedances

For the N and S deposition associated with these air quality scenarios, critical load exceedances were calculated for S, and for N and S combined, for each waterbody in each case study area. Exceedances for N and/or S were calculated for all case study areas except for SHVA. Table 5A-52 contains percent exceedances (number waterbodies exceeding the CL divided by the total number of waterbodies with CLs in the case study area times 100) and the absolute number of waterbodies that exceed the CL. All four ANC thresholds were evaluated. Unlike the CLs, exceedances are not consistent among the case study areas. Percent exceedances were similar between CL values determined for S only and for N and/or S deposition. The highest percent exceedances occurred for the ANC value of 50 μeq/L while lower percent exceedances occurred for ANC of 20 μeq/L, as expected, for all scenarios.

Table 5A-52. Number and percent of case study waterbodies estimated to exceed their CLs for specified ANC values and air quality scenario.

Air Quality Scenario μg/m^3	Areas	Sulfur Only		Sulfur and Nitrogen		Sulfur Only		Sulfur and Nitrogen		Sulfur Only		Sulfur and Nitrogen	
		No.	Percent	No.	Percent	No.	Percent	No.	Percent	No.	Percent	No.	Percent
		ANC of 20 μeq/L				*ANC of 30 μeq/L*				*ANC of 50 μeq/L*			
10	ROMO	3	2%	6	5%	6	5%	16	13%	25	21%	37	31%
	SINE*	1	1%	1	1%	3	2%	3	2%	13	9%	13	9%
	NOMN	2	1%	2	1%	2	1%	2	1%	3	2%	4	2%
	WHMT	3	4%	5	7%	9	12%	10	14%	18	24%	19	26%
	SHVA	9	2%			11	2%			20	4%		
12	ROMO												
	SINE*	1	1%	1	1%	9	6%	9	6%	34	24%	34	24%
	NOMN	2	1%	6	3%	2	1%	11	6%	6	3%	21	11%
	WHMT	21	28%	30	41%	25	33%	36	49%	37	50%	48	65%
	SHVA	16	3%			19	4%			68	15%		
15	ROMO												
	SINE*	2	1%	2	1%	11	8%	11	8%	38	27%	38	27%
	NOMN												
	WHMT	23	31%	35	47%	27	36%	41	55%	38	51%	49	66%
	SHVA	156	34%			202	44%			279	60%		

*The air quality and associated deposition estimates for all air quality scenarios in the Sierra Nevada case study are based on a "roll down" approach. The highest $PM_{2.5}$ DVs in the area were rolled down to equal the specified value for each scenario (15, 12 and 10 μg/m^3) and a unit S or N deposition per unit $PM_{2.5}$ concentration (from a regression based on 21-year CMAQ simulation) was applied to derive the associated deposition estimates presented here.

5A.3 KEY UNCERTAINTIES

In this section, we characterize the nature and magnitude of uncertainties associated with this aquatic acidification REA and their impact on the REA estimates. A summary of the overall characterization of uncertainty for the current deposition-related S exposure and aquatic acidification risk analysis is provided in Table 5A-53 below. This summary is followed by subsections describing quantitative analyses that inform our understanding of the variability and uncertainty associated with the CL estimates developed in this assessment and support the uncertainty characterization regarding the influence of a number of factors. Three sets of analyses are presented in the following subsections. The first, described in section 5A.3.1, is a sensitivity analysis using Monte Carlo techniques to quantify CL estimate uncertainty associated with several model inputs. Section 5A.3.2 describes calculation of confidence intervals for NO_3^- flux estimates in New England and Adirondacks lakes and Appalachian streams. Lastly, 5A.3.3 describes an analysis of the variation in CL estimates among the three primary modeling approaches on which the CLs used in this assessment were based.

The mainly qualitative approach used here and in quantitative analyses in other NAAQS reviews,[22] also informed by quantitative sensitivity analyses, is described by WHO (2008). Briefly, with this approach, we have identified key aspects of the assessment approach that may contribute to uncertainty in the conclusions and provided the rationale for their inclusion. Then, we characterized the *magnitude* and *direction* of the influence on the assessment for each of these identified sources of uncertainty. Consistent with the WHO (2008) guidance, we scaled the overall impact of the uncertainty by considering the degree of uncertainty as implied by the relationship between the source of uncertainty and the exposure and risk estimates. A qualitative characterization of *low*, *moderate*, and *high* was assigned to the magnitude of influence and knowledge base uncertainty descriptors, using quantitative observations relating to understanding the uncertainty, where possible. Where the magnitude of uncertainty was rated *low*, it was judged that large changes within the source of uncertainty would have only a small effect on the assessment results (e.g., an impact of few percentage points upwards to a factor of two). A designation of *medium* implies that a change within the source of uncertainty would likely have a moderate (or proportional) effect on the results (e.g., a factor of two or more). A characterization of *high* implies that a change in the source would have a large effect on results (e.g., an order of magnitude). We also included the direction of influence, whether the source of uncertainty was judged to potentially over-estimate ("*over*"), under-estimate ("*under*"), or have an *unknown* impact to exposure/risk estimates.

[22] This approach to uncertainty characterization has been utilized in welfare and health REAs for reviews of the ozone, NO_2, SO_2, and carbon monoxide NAAQS (e.g., U.S. EPA 2014, 2018).

Table 5A-53. Characterization of key uncertainties in exposure and risk analyses for aquatic acidification.

Sources of Uncertainty		Uncertainty Characterization			
		Influence of Uncertainty on Exposure | Risk Estimates*		Knowledge-base Uncertainty	Comments
Category	Element	Direction	Magnitude		
General Aspects of Assessment Design	Representativeness of National-scale analysis	Over	Unknown	Medium	The analysis may overrepresent the more intensely sampled areas which are likely to be the acid-sensitive sites. There is also potential for uncertainty contributed by sites impacted by loading other than atmospheric deposition (e.g., acid mine drainage).
	Representativeness of Ecoregion-scale analysis	Both	Low	Low	Although the delineation of ecoregions takes into account geology and soil type, there is still variation within ecoregions with regard to acid sensitivity (e.g. type of bedrock), which is an important influence on waterbody acidification from deposition. Wide variation in number, and geographic distribution, of CL sites within an ecoregion, contributes to variation among ecoregions with regard to uncertainty in their risk characterizations. Some ecoregions are well represented with many CLs while other have few. The ecoregion-scale analyses focus on ecoregions with at least 50 CLs and this focus reduces somewhat any impact of poorly characterized (or sampled) regions. However, the lack of consideration of spatial distribution (e.g., not necessarily uniform distribution of CL sites within an ecoregion or similar type of distribution across all ecoregions) contributes uncertainty.
	CLs based primarily on steady-state modeling	Over or both	Low	Low	Nearly all CLs for locations outside of the Adirondacks are based on steady-state modeling (a version of SSWC). Many CLs at Adirondack sites are based on dynamic modeling although most in the Adirondacks are based on steady state modeling. Comparison of CLs using SSWC with F-factor to those with the dynamic MAGIC found generally good correlation (r^2 values greater than 0.95, with slight downward bias for New England lakes and upward bias for Appalachian streams, see section 5A.3.3.2). Monte Carlo analyses described in 5A.3.1 indicate potential magnitude of uncertainty ranging from 0.37 to 33.2 meq/m^2/yr (or 0.1 to 5.3 kg S/ha-yr). The higher values were generally in areas with few water quality data and variable runoff, e.g., the Midwest, South and along the CA to WA coast (section 5A.3.1.2).
	Focus on S deposition only	Under	Low	Medium	Although omitting the contribution of N deposition to reduced ANC has the potential to contribute to underestimates of risk (CL exceedances), assessment of the contribution of N deposition (2000-2020) to % exceedances in this assessment indicates relatively negligible contribution (section 5A.2.1). This may be due to lower N deposition since the latter half of the last century (1970s-1990s) and there may also be an influence of the relatively greater contribution of reduced N to total N deposition over the past 20 years (see Chapter 6, section 6.2.1). Uncertainty in the estimation of N contribution to acidification is likely greater than that associated with estimation of S contribution to acidification given that only a subset of deposited N compounds play a direct role. Uncertainty in the factors influencing the amount of N deposition contributing acidity increases the uncertainty for N deposition (e.g., see entry for N leaching estimate below).

Sources of Uncertainty		Uncertainty Characterization			
		Influence of Uncertainty on Exposure | Risk Estimates*		Knowledge-base Uncertainty	Comments
Category	Element	Direction	Magnitude		
	ANC as indicator of acidification risk	Over	Unknown	Medium	ANC is an indicator of water quality acidification risk, such as conditions in which there is increased toxicity due to dissolved Al concentrations and pH. While studies of acidified water bodies in the Northeast have reported associations of different aquatic effects (e.g., species prevalence) with ANC, there is uncertainty in the relationships on a site-specific basis, related to site-specific factors including deposition and acidification history, as well as site geology. The approach used for estimating ANC in this assessment has the potential to overestimate this risk in waterbodies with appreciable organic acids which can bind dissolved Al, reducing toxicity (ISA, Appendix 8, section 8.3.6.2). However, this is only the case when organic acids are high (>5-8 mg/L) in the surface waters. Organic acids are low in most surface waters across the U.S. except for northern regions of New England, New York, and upper midwest. In addition, levels were low during the height of acidification (1980-90's) and have increased as acidic deposition has decreased, although aluminum toxicity has declined (ISA, Appendix 7, section 7.1.5.1.1). In the ecoregion assessment, three ecoregions were omitted from the focus group of 25 in light of a general recognition of naturally occurring acidity, e.g., associated with organic acids, that reduces waterbody response to reduced acid deposition. While this is not considered to play a role in many of the waterbodies historically recognized as sensitive, the extent of such conditions in other areas is unknown. The parameter, ANC, is also recognized as an indicator of risk of episodic acidification events, although the uncertainty associated with this varies among waterbodies based on historical and recovery status.
	Approach for selection of CL	None	Unknown	Low	At waterbody sites for which multiple CL estimates were available, the most recent was selected. When multiple estimates were available for the most recent period, they were averaged. Use of CLs based on the most recent modeling analyses is not expected to directionally contribute uncertainty.
CLs based on Steady State or Dynamic Models	Non-anthropogenic deposition of base cations (BC_{dep})	None or Over	Low-Medium	Low-Medium	Estimates of BCdep (Ca, Mg, Na, K) are based on deposition estimates to the watershed and waterbody. Wet deposition component is based on measurements of precipitation from the NADP deposition network and is well known. However, the dry deposition fraction is not well known and based on an uncertain relationship to wet deposition. When used, the F-factor approach draws on site-specific surface water chemistry data to estimate the faction of base cation deposition from surface water chemistry data. The surface water chemistry data in the CL used estimates vary in collection date from relatively recent to much older (e.g., 2010s to 1980s) and the CL estimates used a range of sample sizes from a single measurement to multiple years of measurements.

Sources of Uncertainty		Uncertainty Characterization			
		Influence of Uncertainty on Exposure | Risk Estimates*		Knowledge-base Uncertainty	Comments
Category	Element	Direction	Magnitude		
	Base cation weathering (BCw) and flux estimates	Both	Medium	Medium	Dynamic and SSWC model applications used relied on estimates of base cation weathering (BCw) rates or base cation flux. The ISA describes the BCw parameter as "one of the most influential yet difficult to estimate parameters in the calculation of critical acid loads of N and S deposition for protection against terrestrial acidification" (ISA, section IS.14.2.2.1). Obtaining accurate estimates of BCw is difficult because weathering is a process that occurs over very long periods of time, and the accuracy of estimates of an ecosystem's ability to buffer acid deposition relies on accurate estimates of weathering rates within the watershed. Dynamic models use calibrated watershed biogeochemical models that estimate BCw using complex biogeochemical relationships based on soil and water quality measurements, among other factors. These models provide the best estimate of BCw because they take into account the complex nature of the watershed and are calibrated to environmental conditions. In the F-factor approach used to estimate base-cation flux in CL estimates based on SSWC modeling, the components of BCw are estimated as part of the total base cation flux from the watershed. This approach has been widely published and analyzed in Canada and Europe, and has been applied in the U.S. (e.g., Dupont et al., 2005 and others). As described in section 5A.1.5.1, this approach is based on quantitative relationships to water chemistry and site-specific surface water chemistry data for key base cations (Ca, Mg, Na, K) fluxes. The surface water chemistry data vary in collection date from relatively recent to much older (e.g., 2010s to 1980s) and the CL estimates used a range of sample sizes from a single measurement to multiple years of measurements. Although the F-factor approach to estimate base-cation flux has been widely published and analyzed in Canada, Europe, and US, the uncertainty in this estimate hasn't been widely analyzed. Monte Carlo analyses described in 5A.3.1 indicate potential magnitude of uncertainty ranging from 0.37 to 33.2 meq/m^2/yr (or 0.1 to 5.3 kg S/ha-yr). The higher values were generally in areas with few water quality data and variable runoff (e.g., the midwest, south and along the CA to WA coast (section 5A.3.1.2).
	Long-term average uptake of base cations in biomass (harvesting) (BC_U)	Over or none	Unknown	Low	This factor in the CL equation is generally set to zero in applications used in this assessment. Loss of base cations occurs when trees are removed from the watershed from logging. In watersheds where logging is important, BCu was set to 0 and is assumed to have a low bias. The subset of CL estimates from Sullivan et al. (2012b) address this bias through the use of nonzero values drawn from McNulty et al. (2007) for sites outside of protected areas (e.g., national parks) that the authors classified as "no harvest."
	N leaching estimate	Both	Unknown	High	In CLs based on both N and S (used in preliminary analyses of this assessment), the amount of N deposition that contributes to acidification was estimated based on water quality measurements of nitrate and annual runoff (section 5A.1.6.2). Estimating the contribution of N deposition to acidification of surface waters is difficult and uncertain because N cycling in an ecosystem is inherently variable and data for modeling are limited across the U.S. The surface water chemistry data also vary in collection date from relatively recent to much older (e.g., 2010s to 1980s). Use of CLs based on older measurements may have no bias or may overestimate current risk as conditions may have improved (or stayed the same). Analyses in section 5A.3.2 indicate flux estimates to have declined over the period from 1990 to 2018 and for higher values in the Adirondack lakes compared to Appalachian streams, with still lower values in New England lakes. The CL estimates in the main assessment focused on S only and thus, didn't depend on this variable.

Sources of Uncertainty		Uncertainty Characterization			
		Influence of Uncertainty on Exposure | Risk Estimates*		Knowledge-base Uncertainty	Comments
Category	Element	Direction	Magnitude		
Exceedance Calculation		Both	Unknown	Medium	The uncertainty of CL exceedances (deposition > CL) is a function of both the deposition and CL estimate uncertainties. Monte Carlo analyses described in 5A.3.1 indicate potential magnitude of uncertainty in CL estimates to range from 0.37 to 33.2 meq/m^2/yr (or 0.1 to 5.3 kg S/ha-yr) with an average value of 7.68 meq S/m2-yr or 1.3 kg S/ha/yr. Uncertainty in the TDep deposition estimates are characterized in Chapter 6, Table 6-13.
TDEP – Estimation of total deposition		See these entries in Chapter 6, Table 6-13.			
* Influence on direction of exposure or risk estimates means would the exposure (deposition estimate) be potentially biased high or low; or would the risk estimate (probability a CL exceedance) be potentially biased high or low. If the element is concluded to contribute uncertainty with the potential to underestimate a CL, this would be represented by Over in the Direction column as it would have a potential to bias high the associated risk estimate.					

5A.3.1 Quantitative Uncertainty Analyses on Model Inputs

The vast majority of CLs relied on in this assessment are derived using the steady-state model. The strength of the CL estimate and the exceedance calculation relies heavily on model inputs, and particularly estimates of the catchment-average base-cation supply (i.e., input of base cations from weathering of bedrock and soils and air), runoff, and surface water chemistry. The uncertainty associated with runoff and surface water measurements is among the uncertainties characterized in Table 5A-53 above, based on previously available information. The analysis described here is focused on analysis of uncertainty in CL estimates associated with uncertainty in the estimates of the catchment supply of base cations to the waterbodies.

The catchment supply of base cations from the long-term weathering of bedrock and soils is the model input that has been previously recognized as having the most influence on the CL calculation and also has the largest uncertainty (Li and McNulty, 2007; ISA, section IS.14.2.2.1)). Although the approach to estimating base-cation supply used in the SSWC model which was employed in most of the CLs used in this assessment, the F-factor approach, has been widely published and analyzed in Canada and Europe, and has been applied in the CONUS (e.g., Dupont et al., 2005), the uncertainty in this estimate is variable and could be large in some cases. The F-factor is commonly used in the SSWC to account for changes in nonmarine base cation concentrations in a waterbody resulting from strong acid anion concentrations in the derivation of a steady-state, long-term estimate of base cation weathering (2008 ISA, pp. 39-40; Henriksen et al., 2002). Other approaches include empirical estimates and dynamic modeling with the MAGIC (section 5A.1.5 above). Use of the F-factor approach is limited to locations for which acid-base stream chemistry measurements are available. Values for F-factors have been reported to vary over time in response to changing phases of acidification and recovery (Sullivan et al., 2012a). Recognizing this source of uncertainty to the CL estimates, a quantitative uncertainty analysis of the state-steady CL model was completed to evaluate the uncertainty in the CL and exceedance estimation that is associated with the estimation of base-cation supply.

5A.3.1.1 Method

A probabilistic analysis using a range of parameter uncertainties was used for CLs determined by the SSWC model using the F-factor approach to assess (1) the confidence interval of the CL, (2) the degree of confidence in the exceedance values and (3) coefficient of variation (CV) of the critical load. The probabilistic framework is Monte Carlo, whereby each steady-state input parameter being assessed varies according to distribution specified as to shape, minimum and maximum (Table 5A-54). The purpose of the Monte Carlo method was to propagate the uncertainty in the steady-state CL estimates by modeling each value many times to describe the distribution around the CL.

Model calculations were performed for each CL waterbody site with values for the F-factor approach selected via Monte Carlo sampling. At each site, enough model simulations were performed (i.e., 5,000 times) to capture the range of behaviors represented by the SSWC model parameters analyzed (see equation 5A-2, section 5A.1.5.1, for model details). The parameters analyzed were surface water runoff (m/yr), dissolved surface water concentrations of seven chemicals (Table 5A-54). The distributions sampled for these parameters were determined by various methods. For runoff (Q), the minimum and maximum values for annual runoff (m/yr) during the period 1972-2016, available from Wieczorek et al. (2018) were used for each waterbody.[23] The distribution for each of the chemical concentrations at each waterbody site was defined by a normal distribution with the minimum and maximum equal to the minimum and maximum concentration reported for that waterbody, for each waterbody where 6-years of water quality data was available. For waterbodies with fewer than six years of water quality data, the minimum and maximum values were based on a range determined from regional long-term water quality data. Decade averages for the water quality parameters were calculated for sites with long-term data within the region of the site without sufficient data and the minimum and maximum values were used for the decade that matched the water quality data used to calculate the CL. The decades ranged from 1980-90 to 2010-2020. Water quality data used were from the EPA's Long-term Monitoring (LTM) program and that are part of the NCLD.[24]

Table 5A-54. Parameters varied in the Monte Carlo analysis.

Parameter	Units	Distribution Used for Monte Carlo Sampling
Surface water runoff, Q	m/yr	Normal
Calcium, Ca	µeq/L	Normal
Magnesium, Mg	µeq/L	Normal
Chlorine, Cl	µeq/L	Normal
Sodium, Na	µeq/L	Normal
Potassiun (K)	µeq/L	Normal
NO_3^-	µeq/L	Normal
SO_4^{2-}	µeq/L	Normal

The Monte Carlo analysis for the parameters in Table 5A-2 was done in R. A total of 14,943 waterbodies in the CONUS were analyzed. Results of this analysis are described in section 5A.3.1.2 below.

[23] Values were drawn from the U.S. Geological Survey data in Version 3.0 (January 2021) of Select Attributes for NHDPlus Version 2.1 Reach Catchments and Modified Network Routed Upstream Watersheds for the Conterminous United States. This data source is Wieczorek et al. (2018), Select Attributes for NHDPlus Version 2.1 Reach Catchments and Modified Network Routed Upstream Watersheds for the Conterminous United States: U.S. Geological Survey data release, https://doi.org/10.5066/F7765D7V.

24 See https://www.epa.gov/power-sector/monitoring-surface-water-chemistry#tab-6 and obtain data from https://doi.org/10.23719/1518546. Date of data download was 10/21/2020. For NCLD see https://nadp.slh.wisc.edu/clad-national-critical-load-database/

5A.3.1.2 Results

Based on the Monte Carlo analysis for the F-factor approach parameters, we have described the uncertainty around the CL associated with these parameters in terms of the confidence interval around the mean result from the Monte Carlo simulations for each waterbody site. Figure 5A-68 indicates locations for which the variation in the CL estimate, based on the relative size of the range of Monte Carlo outputs, is relatively larger (red and orange dots) and smaller (blue and green).

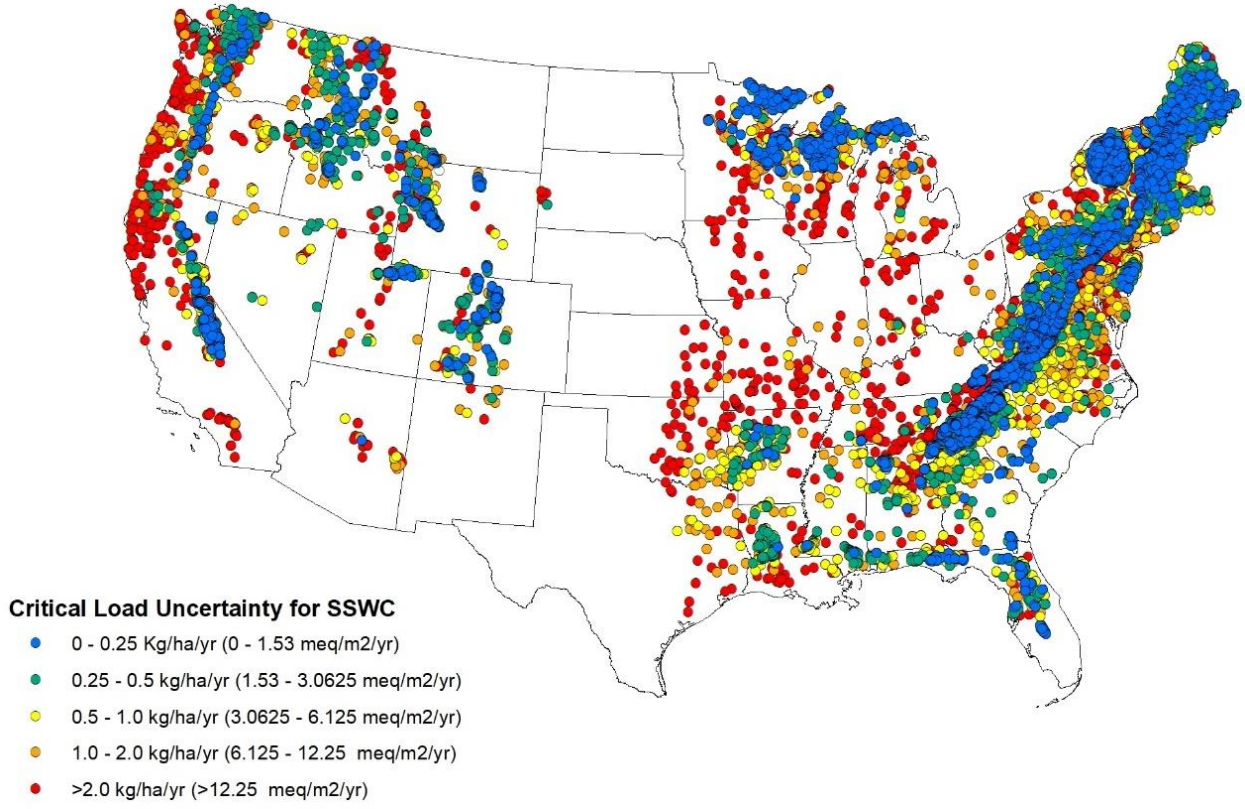

Figure 5A-68. Critical load uncertainty analysis for 14,943 values across the CONUS of the SSWC model. Blue and green dots have the lowest confidence interval and orange, and red dots have the highest confidence interval.

The range of the confidence interval size, based on the 5th to 95th percentile, was 0.37-33.2 meq/m^2-yr or 0.1-5.3 kg S/ha-yr. Sixty-one percent of CL values had a low confidence level of less than 3.0325 meq/m^2-yr or 0.5 kg S/ha-yr, while 26% had levels greater than 6.25 meq/m^2-yr or 1.0 kg S/ha-yr (Table 5A-55). Low confidence intervals were associated with CLs determined with long-term water quality data and low variability in runoff measurements. CL values determined by a single water quality measurement and in areas where runoff is variable (e.g., the western U.S.) had high uncertainty. CLs with the lowest uncertainty occurred in the

eastern U.S., particularly along the Appalachian Mountains, upper Midwest, and Rocky Mountains (Figure 5A-68). Less certain CLs were found in the Midwest and South and along the CA to WA coast. Most of the CLs in the Midwest are based on a single or few water quality measurements while variability in runoff in CA to WA coast account for those high uncertainty values. On average the magnitude of the confidence interval for all SSWC CLs was 7.68 meq S/m^2-yr or 1.3 kg S/ha-yr, giving a confidence level of ±3.84 meq/m^2-yr or ±0.65 kg S/ha-yr.

Table 5A-55. Results of the Monte Carlo analysis for uncertainty broken down by confidence interval.

Range of Confidence interval kg/ha-yr	#. Values	Percent	
0.0 – 0.25	5462	37%	37%
0.25 – 0.5	3612	24%	61%
0.5 – 1.0	1994	13%	74%
1.0 – 2.0	903	6%	80%
>2.0	2972	20%	100%
Total	14943		

Table 5A-56 shows the average and 5th to 95th percentiles of the spread of the confidence interval around the mean of the Monte Carlo simulation CLs for sites in each ecoregion. Fifty-one ecoregions had a sufficient number of sites analyzed to allow for the calculation of a 5th and 95th percentile. Ecoregions in the Appalachian Mountains on average (e.g., Northern Appalachian and Atlantic Maritime Highlands (5.3.1), Blue Ridge (8.4.4), Northern Lakes and Forests (5.2.1), and North Central Appalachians (5.3.3) and Rockies (e.g. Sierra Nevada (6.2.14), Southern Rockies (6.2.14), and Idaho Batholith (6.2.15) had lower uncertainty (smaller confidence intervals around the mean CL from the Monte Carlo simulations), while Northeastern Coastal Zone (8.1.7), Cascades (6.2.7), Coast Range (7.1.8), Interior Plateau (8.3.3), and Klamath Mountains/California High North Coast Range (6.2.11) had on average higher uncertainty.

Table 5A-56. Results of the Monte Carlo analysis for uncertainty by ecoregion.

Ecoregion		No. Values	Average Confidence Interval, CI, on the Mean (5th – 95th percentile CI)	
Code	Name		kg S/ha-yr	meq/m^2-yr
5.3.1	Northern Appalachian and Atlantic Maritime Highlands	2804	0.59 (0.05 – 2.07)	3.71 (0.32 – 12.96)
8.4.4	Blue Ridge	2500	0.32 (0.06 – 0.9)	2 (0.39 – 5.61)
8.4.1	Ridge and Valley	1497	1.64 (0.05 – 8.16)	10.25 (0.33 – 50.98)
5.2.1	Northern Lakes and Forests	894	0.47 (0.02 – 2.04)	2.94 (0.12 – 12.76)
8.3.4	Piedmont	573	1.29 (0.2 – 3.24)	8.09 (1.24 – 20.28)
6.2.12	Sierra Nevada	566	0.41 (0.03 – 1.66)	2.57 (0.18 – 10.39)
6.2.10	Middle Rockies	552	0.95 (0.08 – 5.08)	5.95 (0.53 – 31.76)

Ecoregion		No. Values	Average Confidence Interval, CI, on the Mean (5th – 95th percentile CI)	
Code	Name		kg S/ha-yr	meq/m²-yr
6.2.14	Southern Rockies	444	0.58 (0.1 – 2.1)	3.62 (0.64 – 13.16)
8.3.5	Southeastern Plains	413	1.59 (0.15 – 5.63)	9.94 (0.95 – 35.2)
8.4.2	Central Appalachians	399	1.31 (0.08 – 3.4)	8.18 (0.47 – 21.27)
8.1.8	Acadian Plains and Hills	371	1.2 (0.09 – 4.17)	7.47 (0.54 – 26.09)
8.1.7	Northeastern Coastal Zone	323	2.38 (0.19 – 7.54)	14.87 (1.18 – 47.14)
8.3.1	Northern Piedmont	265	4.1 (0.79 – 11.5)	25.6 (4.96 – 71.89)
8.5.4	Atlantic Coastal Pine Barrens	233	1.1 (0.17 – 3.52)	6.87 (1.06 – 21.98)
6.2.7	Cascades	229	3.68 (0.05 – 2.86)	22.97 (0.29 – 17.89)
5.3.3	North Central Appalachians	222	0.6 (0.09 – 1.99)	3.75 (0.54 – 12.47)
8.1.3	Northern Allegheny Plateau	217	1.46 (0.29 – 4.77)	9.11 (1.79 – 29.79)
6.2.15	Idaho Batholith	212	0.51 (0.13 – 1.75)	3.21 (0.8 – 10.95)
6.2.5	North Cascades	169	1.08 (0.15 – 4.73)	6.75 (0.96 – 29.55)
8.3.7	South Central Plains	157	1.19 (0.32 – 3.09)	7.45 (2.03 – 19.34)
8.5.3	Southern Coastal Plain	149	0.76 (0.1 – 2.89)	4.72 (0.6 – 18.09)
8.4.9	Southwestern Appalachians	127	1.2 (0.18 – 4.71)	7.52 (1.15 – 29.46)
7.1.8	Coast Range	119	5.88 (1.82 – 15.45)	36.77 (11.37 – 96.59)
8.5.1	Middle Atlantic Coastal Plain	118	2.55 (0.26 – 9.04)	15.96 (1.63 – 56.48)
6.2.13	Wasatch and Uinta Mountains	114	1.19 (0.15 – 7.11)	7.46 (0.95 – 44.44)
8.1.4	North Central Hardwood Forests	101	2.3 (0.07 – 4.89)	14.4 (0.45 – 30.59)
6.2.3	Northern Rockies	96	1.14 (0.19 – 4.84)	7.13 (1.18 – 30.27)
8.3.3	Interior Plateau	89	5.44 (0.54 – 12.54)	34.01 (3.36 – 78.36)
6.2.11	Klamath Mountains/California High North Coast Range	85	6.85 (0.43 – 18.46)	42.82 (2.67 – 115.34)
8.1.1	Eastern Great Lakes Lowlands	72	2.69 (0.23 – 8.69)	16.83 (1.43 – 54.29)
6.2.9	Blue Mountains	65	1.33 (0.26 – 4.22)	8.3 (1.62 – 26.37)
8.4.5	Ozark Highlands	61	5.77 (1.22 – 9.5)	36.07 (7.6 – 59.39)
8.4.8	Ouachita Mountains	51	0.94 (0.2 – 3.41)	5.88 (1.26 – 21.29)
8.3.6	Mississippi Valley Loess Plains	41	3.1 (0.26 – 24.02)	19.39 (1.63 – 150.14)
8.4.7	Arkansas Valley	39	1.31 (0.21 – 4.98)	8.2 (1.32 – 31.11)
7.1.7	Puget Lowland	39	2.03 (0.29 – 5.77)	12.71 (1.81 – 36.08)
8.4.3	Western Allegheny Plateau	37	2.03 (0.41 – 4.89)	12.69 (2.55 – 30.55)
8.1.6	Southern Michigan/Northern Indiana Drift Plains	36	2.9 (0.75 – 5.21)	18.12 (4.66 – 32.56)
6.2.4	Canadian Rockies	32	2.5 (0.2 – 7.23)	15.59 (1.22 – 45.2)
6.2.8	Eastern Cascades Slopes and Foothills	32	1.52 (0.21 – 4.84)	9.51 (1.33 – 30.24)
9.4.5	Cross Timbers	31	3.72 (1.58 – 11.31)	23.24 (9.89 – 70.66)
9.2.3	Western Corn Belt Plains	27	3.91 (1.55 – 9.16)	24.43 (9.67 – 57.28)
13.1.1	Arizona/New Mexico Mountains	27	3.22 (0.28 – 10.53)	20.12 (1.74 – 65.79)
8.4.6	Boston Mountains	26	0.89 (0.23 – 4.12)	5.56 (1.42 – 25.72)
11.1.1	Central California Foothills and Coastal Mountains	25	10.79 (0.5 – 54.47)	67.41 (3.1 – 340.46)
9.2.4	Central Irregular Plains	24	3.08 (1.1 – 4.94)	19.27 (6.89 – 30.88)
7.1.9	Willamette Valley	24	3.43 (0.95 – 7.06)	21.45 (5.97 – 44.11)
11.1.3	Southern California Mountains	22	10.21 (1.5 – 20.12)	63.84 (9.4 – 125.78)
8.5.2	Mississippi Alluvial Plain	21	3.85 (0.95 – 9.94)	24.09 (5.91 – 62.1)
10.1.3	Northern Basin and Range	20	1.92 (0.35 – 8.81)	12.01 (2.18 – 55.05)

Ecoregion		No. Values	Average Confidence Interval, CI, on the Mean (5th – 95th percentile CI)	
Code	Name		kg S/ha-yr	meq/m²-yr
8.3.2	Interior River Valleys and Hills	19	4 (1.57 – 10.46)	25 (9.78 – 65.39)
10.1.5	Central Basin and Range	17	N/A *	N/A *
8.2.4	Eastern Corn Belt Plains	16	N/A	N/A
9.5.1	Western Gulf Coastal Plain	16	N/A	N/A
8.1.5	Driftless Area	15	N/A	N/A
8.1.10	Erie Drift Plain	14	N/A	N/A
8.3.8	East Central Texas Plains	11	N/A	N/A
8.2.1	Southeastern Wisconsin Till Plains	11	N/A	N/A
9.4.4	Flint Hills	9	N/A	N/A
9.4.2	Central Great Plains	5	N/A	N/A
10.1.4	Wyoming Basin	4	N/A	N/A
9.4.7	Texas Blackland Prairies	3	N/A	N/A
5.2.2	Northern Minnesota Wetlands	2	N/A	N/A
11.1.2	Central California Valley	2	N/A	N/A
10.1.8	Snake River Plain	2	N/A	N/A
10.1.2	Columbia Plateau	2	N/A	N/A
8.2.3	Central Corn Belt Plains	2	N/A	N/A
9.3.1	Northwestern Glaciated Plains	2	N/A	N/A
10.1.6	Colorado Plateaus	1	N/A	N/A
* N/A indicates there was not a sufficient number of sites in the analysis to support calculation of percentiles.				

5A.3.2 Uncertainty Analysis for N Leaching Estimates

An analysis of uncertainty associated with NO_3^- flux used to estimate N leaching into lakes or streams was performed using water quality data from EPA's Long-term Monitoring (LTM) program[25] over the past 28 years. In EPA's LTM program, lakes or streams are sampled weekly to quarterly depending on the site and individual project. Annual flux of NO_3^- was calculated using annual concentration of NO_3^- for a given monitoring site and multiplied by annual runoff, in m/yr (Wieczorek et al., 2018) for the watershed and year. Confidence intervals were calculated for monitoring sites for a given region (i.e., New England, Adirondacks Mountains, and Appalachian Mountains) and for four time periods (i.e., 1990-2018, 1990-1999, 2000-2009, 2010-2018).

The results of this analysis are summarized by region and time period in Table 5A-57. Nitrate flux varied between regions with Adirondacks lakes having the highest annual fluxes and New England Lakes with the lowest fluxes. Average values ranged from 0.36 to 11.71 meq/m^2-yr as NO_3^-(0.01 to .37 kg N/ha-yr). The ranges of confidence interval for the NO_3^- flux differed across the monitoring sites from 0.15 to 1.62 meq/m^2-yr as NO_3^- (0.01 to 0.05 kg N/ha-yr). A

[25] The EPA's Long-Term Monitoring program tracks changes in surface water chemistry in the four regions shown below, known to be sensitive to acid rain: New England, the Adirondack Mountains, the Northern Appalachian Plateau, and the central Appalachians (https://www.epa.gov/power-sector/monitoring-surface-water-chemistry#tab-6). Data from this program are available at: https://doi.org/10.23719/1518546.

combined S and N confidence interval was ± 3.87 to 4.20 meq/m^2-yr which is equivalent to 0.61 to 0.672 kg S/ha-yr or 0.54 to 0.58 kg N/ha-yr.

Table 5A-57. Uncertainty analysis of NO_3^- flux estimates based on data from EPA's Long-term Monitoring Program.

	Average (meq/m^2-yr)	S.D. (meq/m^2-/yr)	5th to 95th (meq/m^2-yr)	Magnitude & Confidence Interval (meq/m^2-yr)
New England Lakes				
All Years	0.7	1.05	0.01 – 2.87	0.15 (0.62 – 0.78)
1990 to 1999	0.8	1.17	0.00 – 3.10	0.30 (0.64 – 0.95)
2000 to 2009	0.92	1.18	0.01 – 3.88	0.29 (0.78 – 1.07)
2010 to 2018	0.36	0.59	0.01 – 1.48	0.15 (0.29 – 0.44)
Adirondacks Lakes				
All Years	8.82	7.79	0.13 – 23.52	0.77 (8.44 – 9.21)
1990 to 1999	11.71	9.01	0.72 – 27.83	1.62 (10.89 – 12.52)
2000 to 2009	9.28	7.11	0.68 – 21.2	1.16 (8.70 – 9.86)
2010 to 2018	5.73	6.01	0.00 – 17.91	1.03 (5.21 – 6.24)
Appalachian Streams				
All Years	3.27	5.77	0.03 – 13.68	0.52 (3.00-3.53)
1990 to 1999	5.05	7.29	0.43 – 20.18	1.14 (4.48 – 5.61)
2000 to 2009	2.43	4.75	0.00 – 11.61	0.73 (2.06 – 2.79)
2010 to 2018	2.27	4.30	0.00 – 10.82	0.70 (1.92 – 2.62)

5A.3.3 Variation in Critical Load Estimates Associated with Modeling Approach

To consider the influence of modeling approach on CL estimates, we compared estimates derived using three types of approaches: (1) the steady-state approach, based on the SSWC model with F-Factor approach for estimating base cation weathering; (2) the steady-state approach with statistical regression model for estimating base cation weathering; and, (3) the dynamic model, MAGIC. The CLs used in this REA are nearly all based on the first of these (SSWC with F-Factor approach), although many of the CLs in the Adirondacks were derived using the MAGIC model. The analyses described here provide a sense of the variation in CL estimates based on these three approaches.

5A.3.3.1 Method

Critical loads used in the national assessment analysis used different methods (see methods for more details). To understand differences in the CLs calculated with different methods, waterbodies where methods overlap were compared. There are three main CL approaches that have been applied in the literature, all based on watershed mass-balance approach where acid-base inputs are balanced. The three approaches include: (1) SSWC model and F-Factor (SSWC F-Factor) that is based on quantitative relationships to water chemistry

(Scheffe et al., 2014; Lynch et al., 2022), (2) Steady State model with Statistical Regression Model (Regional Regression) that extrapolated weathering rates across the landscape using water quality or landscape factors (Sullivan et al., 2012b; McDonnell et al., 2014), and (3) Dynamic Models (MAGIC) (U.S. EPA, 2009). Critical load values were compared among the three models applying these approaches to determine model biases.

Comparisons of CLs among the approaches (e.g., SSWC F-Factor, Regional Regression, and MAGIC) were completed for the lakes in New England and the Adirondacks and streams in the Appalachian Mountains that each had CLs based on three different approaches (drawn from NCLD). A total of 114 and 77 CLs were compared between SSWC Factor and MAGIC approaches for New England and Adirondacks lakes, respectively. A total of 1129 CLs were compared between SSWC Factor and Regional Regression based on CLs from Sullivan et al. (2012a) for lakes in the Adirondacks. For streams in Appalachian Mountains, 66 CLs were compared between SSWC Factor and MAGIC approaches and 43 between SSWC Factor and Regional Regression based on CLs from McDonnell et al. (2014).

5A.3.3.2 Results

Results from the comparison between different CL methods are summarized below for lakes in New England and the Adirondacks and streams in the Appalachian Mountains. For New England and Adirondacks lakes, the MAGIC and the SSWC F-Factor (Lynch et al., 2022; Scheffe et al., 2014) CL values were comparable with a R^2=0.979 and R^2=0.9587 and RMSE of 15 and 21 meq/m^2-yr, respectively (Figure 5A-69).

Across CLs for all sites in the three regions, the Regional Regression (Sullivan et al., 2014) CL estimates were strongly correlated with those from the SSWC F-Factor model, with R^2= 0.9815 and a bias towards higher values for the Regional Regression approach (Figure 5A-70a). For all CLs in the three regions within the range of 0 to 150 meq/m^2-yr, the correlation was slightly lower (r^2=0.8922) and the regression coefficient closer to one (1.0365) (Figure 5A-70b).

For streams in the Appalachian Mountains, general agreement was found between the SSWC F-Factor, Regional Regression, and MAGIC approaches; with the MAGIC approach showing better correlation (than the Regional Regression approach) with the SSWC F-factor., For example, CLs determined by the MAGIC approach were highly correlated with CLs derived with the SSWC F-factor approach, with a R^2=0.9887 and RMSE of 24 meq/m^2-yr (Figure 5A-71a)**.** However, the correlation was not as strong (R^2=0.8861) between CLs based on Regional Regression approach (McDonnell et al., 2014) and the SSWC F-Factor model (Lynch et al., 2022; Scheffe et al., 2014), with the SSWC F-factor CLs generally lower than those based on the Regional Regression approach as indicated by a regression coefficient of 0.7396 (Figure 5A-71b). We additionally note that McDonnell et al. (2014) reported a highly correlated relationship

(R^2 = 0.92 and RMSE = 9-11 meq/m^2-yr) with the MAGIC approach. Overall, generally good agreement has been found between the three methods used to calculate CLs that were used in this assessment, indicating that they would be expected to produce comparable results when used together.

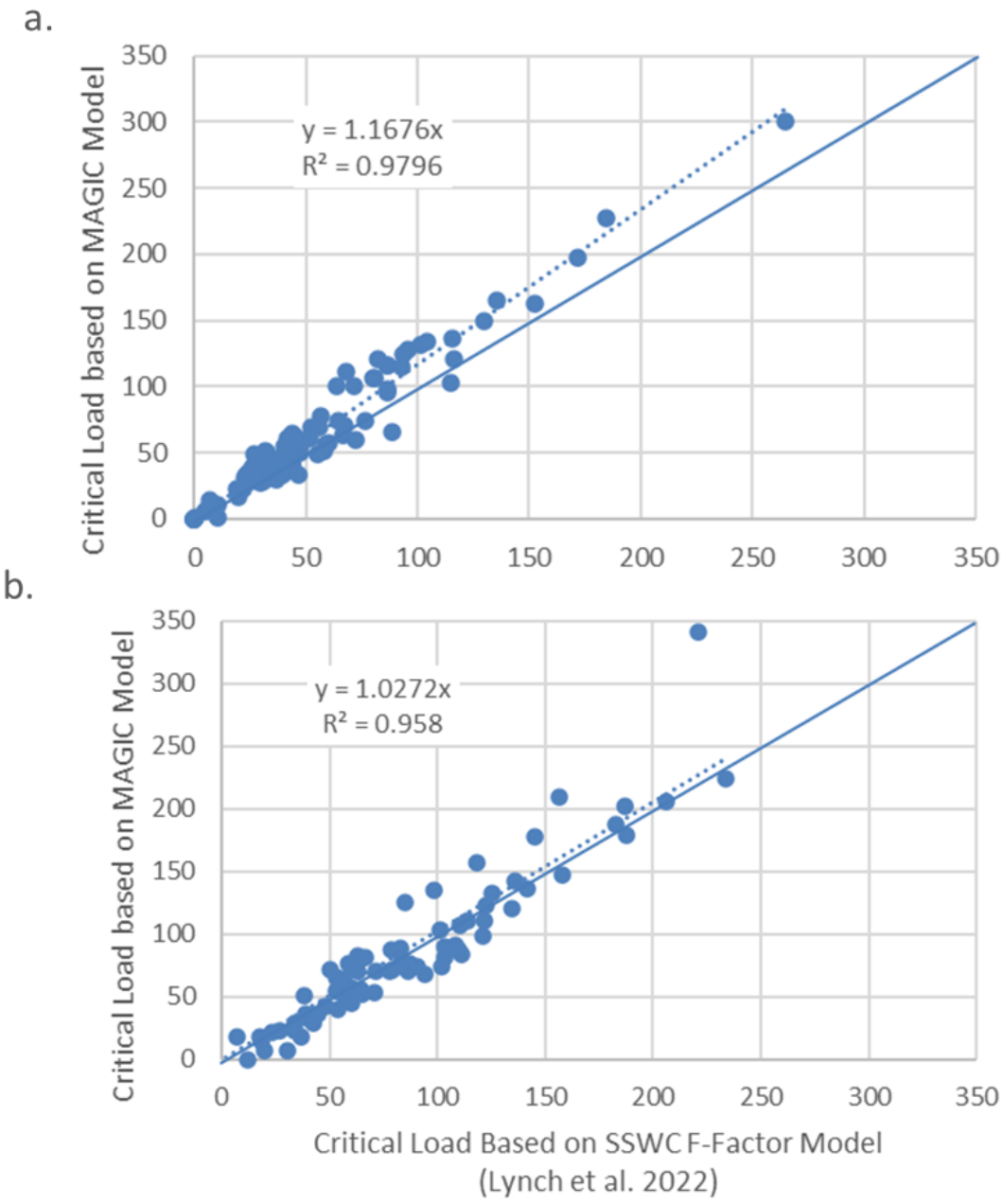

Figure 5A-69. Critical load comparison between values based on MAGIC (y-axis) and values based on the SSWC F-factor (Lynch et al., 2022) for New England lakes (a.) and Adirondack lakes (b.). Units are meq/m^2-yr.

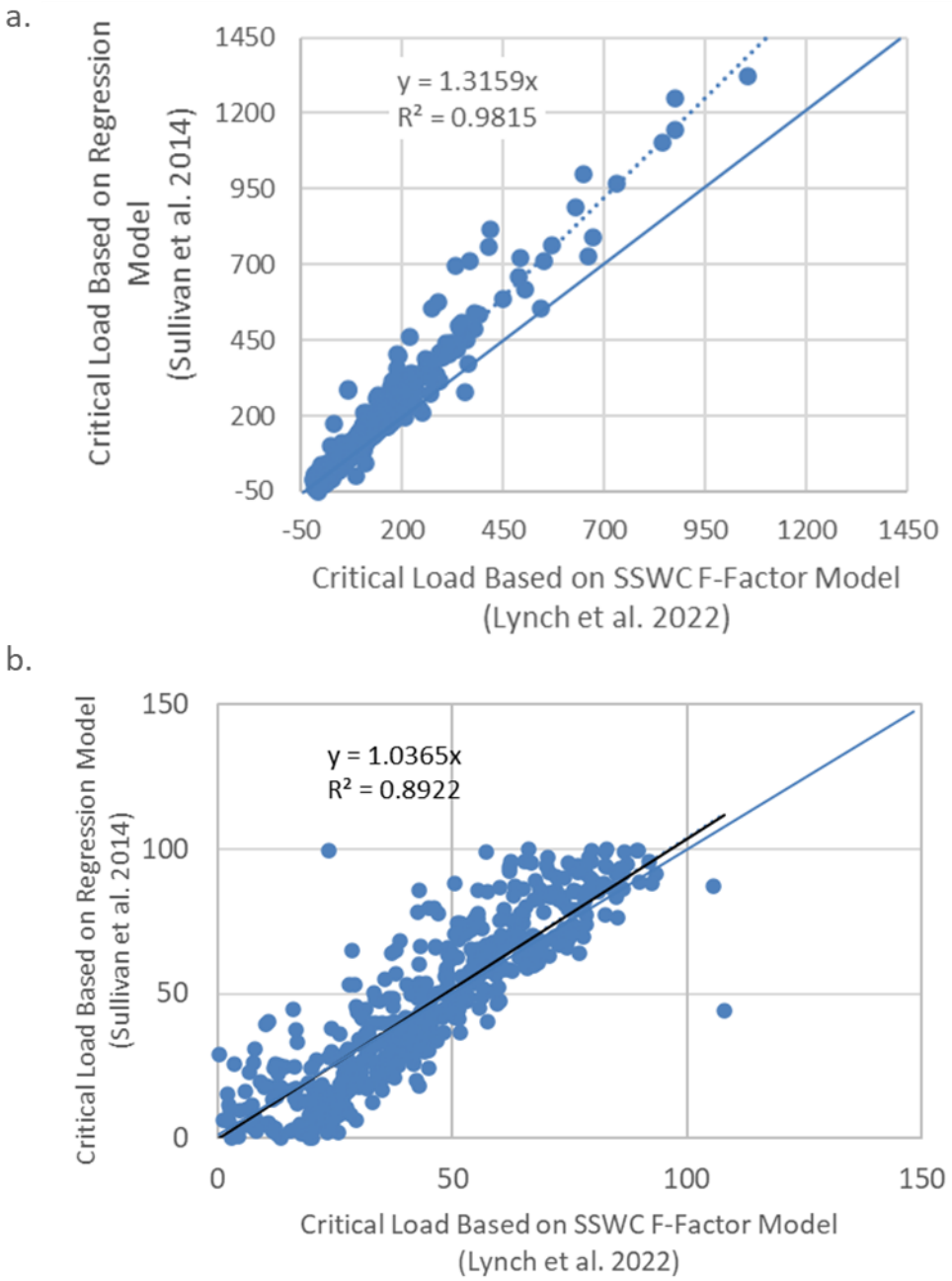

Figure 5A-70. Critical load comparison between values based on Regional Regression model (Sullivan et al., 2014) (y-axis) and values based on the SSWC F-factor model (Lynch et al., 2022) for the full range of CLs (a) and for the range from 0 to 150 meq/m^2-yr (b). Units are meq/m^2-yr.

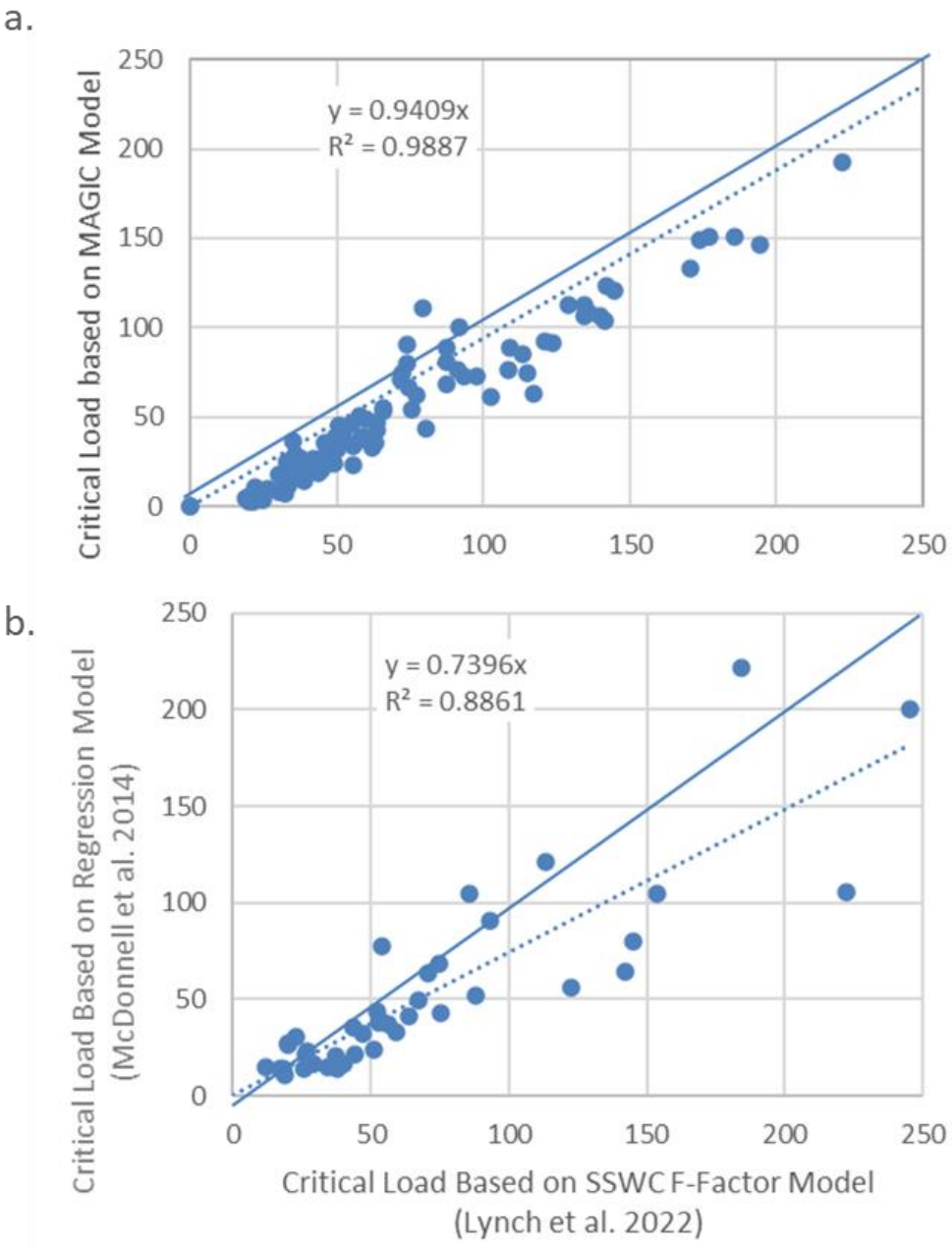

Figure 5A-71. Critical load comparisons: (a.) between values based on MAGIC (y-axis) and values based on the SSWC F-factor model (Lynch et al., 2022) (x-axis); and (b.) between values based on Regional Regression model (McDonnell et al., 2014) (y-axis) and values based on the SSWC F-factor model (Lynch et al., 2022) (x-axis). Units are meq/m^2-yr.

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APPENDIX 5B

ADDITIONAL DETAIL RELATED TO KEY TERRESTRIAL ECOSYSTEM STUDIES

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2B. Species-specific Sample Distribution across Ecoregions for Species with Statistically Significant Associations of Survival with N/S, from Horn et al. (2018) Supplemental Information Dataset

5B.1 INTRODUCTION

This appendix summarizes salient aspects of key studies investigating responses of terrestrial ecosystem components (trees, communities of herbs and shrubs, and lichens) to sulfur and nitrogen deposition, and direct effects of the pollutants in ambient air. The effects may relate to ecosystem acidification (e.g., acidification of soils in which plants are growing) or nutrient enrichment (e.g., through changes in competitive advantages of nitrogen-limited species) or both. The studies described here vary in the extent to which they clarify which factors may be eliciting the responses. Two general types of studies are described in the sections that follow: controlled addition experiments and observational (or gradient) studies. Each has strengths, limitations and uncertainties associated with interpretation.

The strengths of the controlled addition study design include its ability to elucidate N- or S–related factors and circumstances (e.g., chemical form, duration, concentration) that elicit a response in the exposed plants (e.g., changes in growth rates of individual species, changes in productivity of a forest plot, changes in community composition). The scope of impacts that can be studied, however, is generally limited in the species included, and the size of terrestrial community. Observational studies, in contrast, can include a large number and range of species and terrestrial communities, including species less amenable to maintenance in controlled experimental conditions. These studies, also called gradient studies as they provide for consideration of observations across a gradient of pollutant concentrations, provide for the assessment of numerous species and communities across large areas, including across ecoregions.[1] Further, controlled addition studies, which generally include controls that have not received additions, may be limited to assessment of responses to the addition of the specific study chemicals. An observational study by its very nature involves the combined impact of historical and contemporaneous atmospheric deposition in the study areas, which then poses challenges to disentangling the effects of historic versus recent deposition and of the various chemicals deposited, as well as the effects of the soil chemistry and geology.[2] Further, the

[1] Ecoregions are areas where ecosystems (and the type, quality, and quantity of environmental resources) are generally similar. The ecoregion framework referenced in this document is derived from Omernik (1987) and from mapping done in collaboration with EPA regional offices, other Federal agencies, state resource management agencies, and neighboring North American countries. Designed to serve as a spatial framework for ecosystems and ecosystem components, ecoregions denote areas of similarity in the mosaic of biotic, abiotic, terrestrial, and aquatic ecosystem components with humans being considered as part of the biota.

[2] In context of 2015 ozone NAAQS review, and regarding potential use for predictive purposes in that review of a single-species O_3 gradient study involving tree seedlings planted in fields with transplanted soil at locations along a gradient in O_3 concentrations, CASAC, while noting it to provide important results, cautioned care in consideration for predictions in other circumstances of this single study that used a gradient methodology without experimental control of the pollutant exposures (Frey et al., 2014).

observational studies do not generally include measurements or assessments of the site soil chemistry or geology. Rather, they utilize atmospheric deposition estimates at assessment sites as surrogates for exposure conditions. These various strengths and limitations inform consideration of the studies below.

5B.2 TREE GROWTH AND SURVIVAL

As described in the ISA, acidic deposition, which can be comprised of S and N compounds, can contribute to acidification of soils in which trees grow (ISA, section IS.5). Deposition of N can also contribute to N enrichment of soil, which can increase the growth of N-limited trees. In a mixed forest, this can contribute to competitive advantages (depending on species' growth rates), and potentially reducing the growth rate of out-competed species (ISA, section IS.5.2). The relationship between deposition and these effects depends on soil status with regard to acidification and N content, and accordingly is influenced by historic deposition and the soil characteristics important to soil responses. As noted in the ISA, "[i]n areas where N and S deposition has decreased, chemical recovery must first create physical and chemical conditions favorable for growth, survival, and reproduction" for biological recovery to occur (ISA, p. IS-102). For example, although fewer studies have tracked potential recovery of terrestrial than aquatic ecosystems, modeling studies in the southern Appalachian Mountains "suggest current stress and recovery likely to take decades even under scenarios of large reductions in S deposition" (ISA, p. 4-99). In the subsections below, we provide details of several key studies in the current ISA that evaluate relationships between N and S deposition on tree growth and survival.

5B.2.1. Addition Studies

Several experiments involving S or N additions have been reported in the ISA focused on study areas in the eastern U.S. These studies involve appreciable annual additions of S and/or N compounds to experimental forest plots. While some study durations are limited to fewer than five years, others extend appreciably longer than 10 years, providing the time to affect chemical pools within the soil and the associated soil characteristics linked to acidification or nutrient enrichment effects (e.g., Ca:Al ratio or NO_3^- leaching). Among the studies summarized in Table 5B-1 below are addition studies that found species-specific results for growth and survival for several eastern species including oaks, spruce, maples and pines. (Magill et al., 2004; McNulty et al., 2005; Pregitzer et al., 2008; Wallace et al., 2007). Further, some multiyear S/N addition (>20 kg/ha-yr) experiments with a small set of eastern species including sugar maple, aspen, white spruce, yellow poplar, and black cherry, have not reported growth effects (Bethers et al., 2009; Moore and Houle, 2013; Jung and Chang, 2012; Jensen et al., 2014).

Table 5B-1. Experimental addition studies assessing tree growth and/or survival.

Location, Reference	Description	Additions	Tree specific Findings
Michigan (Pregitzer et al., 2008)	Four study areas across a 500 km gradient in temperature and N deposition in NW Michigan. Forests (approx 90 years old) dominated by sugar maple (82% by basal area). Study assessed soil biogeochemical properties, microbial communities, tree growth/ mortality.	30 kg N $ha^{-1}yr^{-1}$ for 10 years starting in 1994 (as $NaNO_3$). Background deposition ranged from 6.8 to 11.8 kg N $ha^{-1}yr^{-1}$.	Increased growth (total live woody biomass) and mortality. Total deposition estimates: 36.8-41.8 kg/ha-yr.
Mt. Ascutney, VT (McNulty et al., 2005)	Six study plots in montane spruce-fir forests. Assessed soil biogeochemical properties, microbial communities and tree growth and mortality.	15.7 and 31.4 kg N $ha^{-1}yr^{-1}$ over 14 years starting in 1988 (as NH_4Cl). Background deposition was 10 kg N $ha^{-1}yr^{-1}$	Reductions in total live basal area (low N-↓18%; high N ↓40% vs control↑9%), indicating reduced growth rates; increased red spruce mortality in high N.
Bear Brook, ME (Elvir et al., 2003; Bethers et al., 2009)	Two experimental watersheds (1 control and 1 treatment), each with softwood, mixed wood, and hard wood forest. Studies assessed soil biogeochemical properties, microbial communities and tree growth.	25.2 kg N $ha^{-1}yr^{-1}$ and 28.8 kg S $ha^{-1}yr^{-1}$ (as $(NH_4)_2SO_4$) starting in 1989; assessed after 10 yrs. Initial background deposition was 8.4 kg N $ha^{-1}yr^{-1}$ and 14.4 kg S $ha^{-1}yr^{-1}$.	Increased growth rates for sugar maple, but not for red spruce. No effect on sugar maple seedling density.
Northern Quebec, Canada (Moore and Houle, 2013)	Eight year N addition (approximately 3x and 10x estimates of concurrent deposition), beginning in 2001, across 9 plots in boreal forests with sugar maple, yellow birch and American beech. Studies assessed soil chemistry, foliar chemistry and tree growth and crown dieback.	26 and 85 kg N $ha^{1}yr^{-1}$ (from ammonium nitrate additions as NH_4NO_3) Background wet deposition of 8.5 kg N $ha^{-1}yr^{-1}$	After 8 years, no effect on sugar maple basal area growth or crown dieback.
Harvard Forest, MA (Magill et al., 2004)	Eight plots, four in a red pine plantation and four in a hardwood forest stand dominated by red and black oak, were assessed for tree growth and mortality.	50 and 150 kg N $ha^{-1}yr^{-1}$ for 14 years starting in 1988 (as NH_4NO_3). Background deposition was 9 kg N $ha^{-1}yr^{-1}$	Increased growth (stand-level biomass), but no change in mortality in the hardwood forest. Decreased growth and increased mortality in the red pine plantation.
Canada (Jung and Chang et al., 2012)	At study plots near Atasca oil sands, assessed above ground tree biomass. Main canopy species were quaking aspen and white spruce. Also included balsam fir, balsam poplar, black spruce and paper birch	30 kg N/ha-yr, 30 kg S /ha-yr and 30 kg N+30 kg S /ha-yr from 2006-2009	Biomass was increased in N-only treatment and was highest in the N+S treatment. Understory biomass unaffected. No evidence of increased NO_3^- leaching
Millbrook, NY (Wallace et al., 2007)	Six pairs of plots in an upland mixed-oak forest dominated by chestnut oak, northern red oak and hickories at the Institute of Ecosystem Studies where studies assessed NO_3^- leaching, tree growth and mortality.	100 kg N $ha^{-1}yr^{-1}$ (1996 to 1999), then 50 kg N $ha^{-1}yr^{-1}$ (2000 to 2003) (as NH_4NO_3). Background deposition was 10 kg N $ha^{-1}yr^{-1}$	Increased growth rates across species (oaks and hickories) and increased mortality in oaks.
Fernow Forest, WV (May et al., 2005; Jensen et al., 2014)	Two paired watersheds, one control and one treatment. The most abundant species were red maple, tulip poplar and black cherry. Studies assessed soil biogeochemical properties and tree growth and mortality.	35 kg N $ha^{-1}yr^{-1}$ and 40 kg S $ha^{-1}yr^{-1}$ starting in 1989 (as $(NH_4)_2SO_4$) Background deposition was approximately 15 kg N $ha^{-1}yr^{-1}$ and 20 kg S $ha^{-1}yr^{-1}$	Reduced growth (stem diameter) in all 3 species (red maple, tulip poplar and black cherry) based on measurements taken in 1999 and 2001 (after 10 years of treatments). No difference in growth (basal area) for tulip poplar and black cherry after 22 yrs.

5B.2.2. Gradient or Observational Studies

The evidence newly available in this review includes observational or gradient studies that investigated the existence of statistically significant associations of tree growth and survival or mortality with S or N deposition (Table 5B-2; ISA, Appendix 5, section 5.5.2 and Appendix 6, sections 6.2.3.1, 6.3.3 and 6.6.1). In general, these studies utilized measurements of tree growth and survival or mortality across multiyear intervals at designated plots, and estimates of average S and/or N deposition (or in some cases, emissions estimates) in the same locations. Statistical models were employed in the analyses and took into account the influence of different sets of additional factors (e.g., related to climate, other air pollutants, topography and stand characteristics).

Tables 5B-2 and 5B-3 below summarize these studies, some of which focused on regions within a state and others which encompassed multistate regions. The three larger studies utilized data from the USFS Forest Inventory and Analysis (FIA) program in which measurements are taken at multiyear intervals at designated plots in forests across the U.S. The three studies have utilized USFS-FIA data for different, but overlapping, study areas (Figure 5B-1) and species. More detailed descriptions of these studies and their findings are provided in sections 5B.2.2.1 through 5B.2.2.3 below.

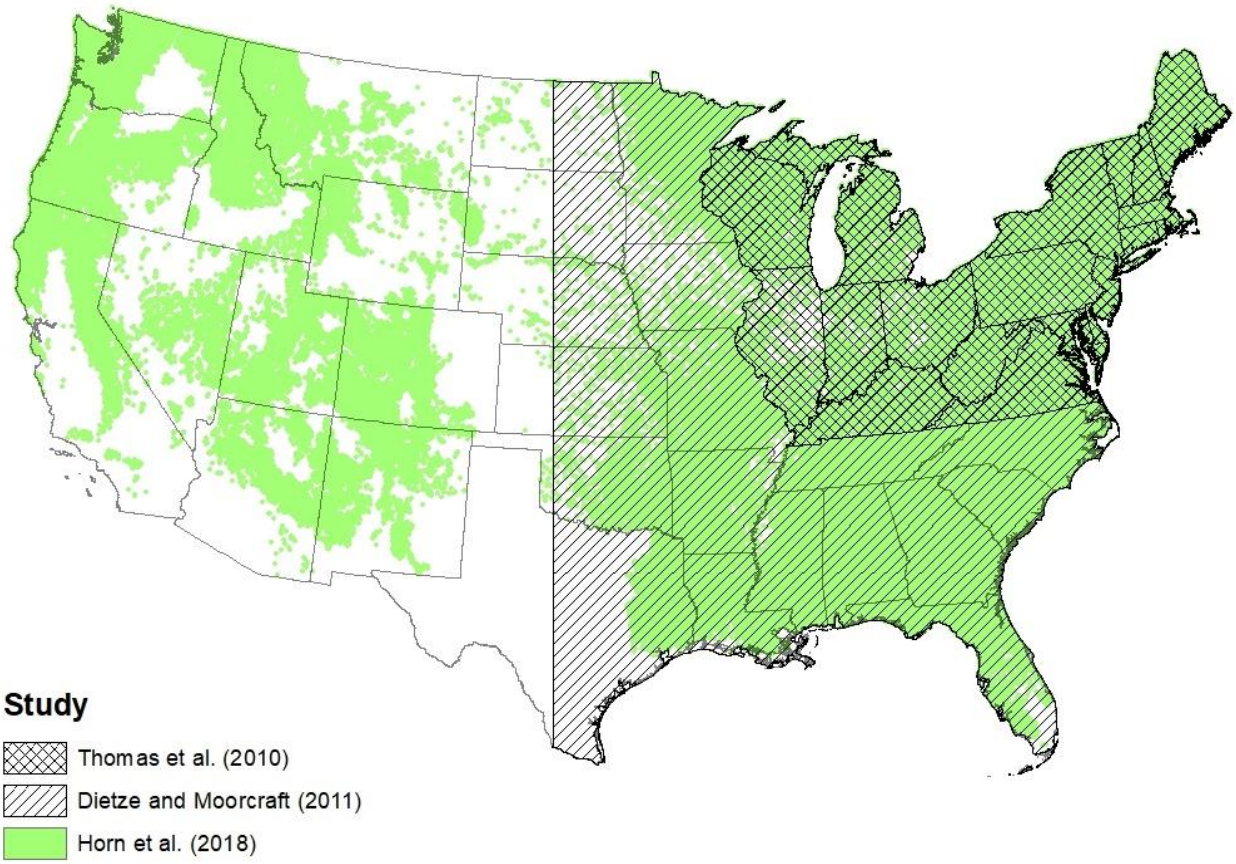


Figure 5B-1. Study areas of three observational studies utilizing FIA plot data. The western extent of Dietze and Moorcroft (2011) is a rough approximation.

Other observational studies in the recently available evidence have investigated relationships of tree growth with estimates of SO_X and N oxide emissions. For example, increases in eastern red cedar growth in West Virginia have been associated with reductions in SO_2 emissions and increases in atmospheric CO_2 concentrations (Thomas et al., 2013). In a North Carolina high-elevation forest, increases in red spruce radial growth since the late 1980s has been associated with declining SO_X and N oxide emissions from SE utilities, as well as increasing temperatures and CO_2 (Soulé, 2011). Recent studies in areas of Europe where SO_2 concentrations are generally higher than in the U.S. have also reported increased growth of some conifer species (e.g., silver fir) to be related to reductions in SO_2 concentrations (ISA, Appendix 3, section 3.2).

Table 5B-2. Recent gradient/observational studies of associations between tree growth and survival or mortality and S or N deposition: smaller-scale studies.

Study	Description	Summary
	Smaller Regional Scales	
Bedison and McNeil (2009)	32 plots in northern hardwood and subalpine spruce-fir dominated forest plots in Adirondack Park, NY. Trees were measured in1984 and 2004. The spatial pattern of inorganic N deposition in wet deposition was estimated across the plot locations by multiple regression. Analyses performed for growth of both individual species and all individuals within each plot. Potential influence of S deposition was not assessed.	At the species level, positive associations of growth with N deposition were found for maple, spruce and fir species, with the largest growth increases in red maple, balsam fir and red spruce. Responses varied by forest type and size class.
Sullivan et al. (2013)	Study focused on 50 plots in western Adirondack region with commonly occurring sugar maple and a 10-fold range of Ca availability (based on previous stream and soil studies). Plant measures included DBH of all trees > 10 cm within plots, assessment of sugar maple canopy condition and vigor, dendrochronology of sugar maple trees, and seedling and sappling counts in subplots. Soil chemistry measurements included base saturation, exchangeable calcium, exchangeable magnesium and soil pH. Total S and inorganic (nitrate and ammonium) N deposition estimated using empirically based GIS model. Average annual (based on the period 2000-2004) N deposition was calculated as the product of estimated average annual precipitation from PRISM5, based on 30-year normals (1970-2000) and kriged S and N precipitation chemistry from NADP locations. Dry deposition of SO_4-S, HNO_3-N, and particulate NO_3-N and NH_4-N across Adirondack region calculated as products of air concentrations, based on the average of 2000-2004 CASTNET air chemistry data, and vegetation cover deposition velocities per CASTNET protocols.	- Plots with lower soil base saturation did not have sugar maple regeneration, with the proportion of sugar maple seedlings dropping off substantially from at/above approximately 60% for base saturation levels at/above 20% to at/below approximately 20% for base saturation at/below about 10%. - Canopy vigor was positively correlated with soil pH and exchangeable Ca, Mg. - Mean growth rates (BAI) were positively correlated with exchangeable Ca and base saturation at the watershed level. Sugar maple distribution negatively associated with estimated average 2000-04 N+S deposition (750-1120 eq/ha/yr)

Table 5B-3. Recent gradient/observational studies of associations between tree growth and survival or mortality and S or N deposition: larger-scale FIA data studies.

Study	Description	Summary
Larger Regional and National Scales *(and using USFS FIA data)*		
Thomas et al. (2010)	Assessed 24 of the most common northeastern tree species using 20,067 FIA plots in 19 states from 1978 to 2001, with the measurement interval varying from 8.3 to 14.4 across states. Tree growth and survival were assessed with regard to association with N deposition (mean annual total N deposition, 2000-04).	Growth of 11 species was positively associated with N deposition (including all species with arbuscular mycorrhizal fungi associations). Growth of 3 species was negatively associated with N deposition. Survival of 8 species was negatively associated with N deposition, with positive associations for 3 species.
Dietze and Moorcroft (2011)	Assessed influence of patterns of SO_4^{2-} and NO_3^- wet deposition (1994-2005 average), O_3 (1996-2006) and climate, topographic and tree stand factors on observed variation in tree mortality at FIA plots in the eastern and central U.S. from 1971 to 2005, binning the 267 species into 10 plant functional types.	Mortality in 7 of the functional groups was positively associated with both SO_4^{2-} and O_3; and negatively associated in 1 group. Mortality in 9 of the 10 functional groups was negatively associated with NO_3^-, and positively associated in 1 functional group.
Horn et al. (2018)	At USFS/FIA plots across the continental U.S., analyzed potential for associations of growth and survival across a measurement interval (of generally 10 years) with estimates of average N and S deposition for the same interval, all within the period, 2000-2013. Other factors included in the analysis were temperature, precipitation, and terms representing the influence of tree size and competition on growth and survival. Deposition estimates were drawn from TDep dataset of NADP's Science Committee on Total Deposition for the measurement interval of each plot. The analyses focused on 71 species that met criteria for sample size (>2000 trees for both growth and survival datasets) and for collinearity (correlation among the independent variables) of N or S, separately, with the other three independent variables (S or N, temperature and precipitation) for growth or survival (Variance Inflation Factor < 3).	Growth in 31 species was negatively associated with S deposition. Survival in 40 species was negatively associated with S deposition. Growth in 20 species was positively associated with N deposition and in 2 species (yellow birch and eastern hemlock) was negatively associated. Growth in 17 other species was positively associated with N deposition at lower levels and negatively associated at higher levels. Survival of 1 species was positively associated with N deposition and in 6 species was negatively associated. Survival in 25 other species was positively associated with N deposition at lower N deposition and negatively associated at higher levels.

5B.2.2.1. Dietze and Moorcroft (2011)

The study by Dietze and Moorcroft (2011) statistically analyzed patterns of tree mortality in the eastern and central U.S. using FIA data from 1971 to 2005. The total sample size was 3.4 million tree measurements and 750,000 plot level measurements. Mortality was quantified as a binary metric (lived or died) based on resampling of FIA plots after intervals of 5 to 15 years.

Climate data were extracted from the database maintained by the PRISM database.[3] Using data from 1971 to 2000, the annual average precipitation, average monthly minimum temperature across December, January and February, and the average monthly maximum temperature across June, July and August were calculated. Air quality data were obtained from the National Atmospheric Deposition Program (NADP) for estimates of wet deposition (in kg $ha^{-1}yr^{-1}$) for ammonium (NH_4^+), nitrate (NO_3^-), hydrogen ion (H^+) and sulfate (SO_4^{2-}) for the period of 1994-2005 and from the EPA's AIRDATA database for ozone for the period of 1996-2006. The ranges of sulfate and nitrate wet deposition estimates across the study area were 4 to 30 kg/ha-yr and 6 to 16 kg/ha-yr, respectively (Dietze and Moorcroft, 2011). There were 267 tree species sampled in the study region. The species were classified into 10 different plant functional types to facilitate analyses (see Attachment 1). The mortality analysis utilized a logistic regression model for binary mortality probability, relating the mortality probability (live or dead) to a linear model of the covariates.

All 13 covariates[4] were found to be statistically significant predictors of mortality for more than one of the plant functional types. Sulfate deposition demonstrated a significant positive effect on mortality in seven of the 10 plant functional groups and a slight negative effect in one group (Table 5B-4). Nitrate deposition demonstrated a significant negative effect on mortality in 9 of the 10 plant functional groups and a positive effect in the tenth. Of note is that ozone exhibited the same pattern of effects as SO_4^{2-} (Table 5B-4). The authors also noted correlations between the nitrate and sulfate wet deposition estimates (correlation coefficient of 0.82), and that the highest deposition estimates were for the Ohio River valley and the northeastern United States (Dietze and Moorcroft, 2011).

[3] The PRISM (Parameter-elevation Regressions on Independent Slopes Model) database is maintained by the PRISM Climate Group, who compile data from multiple monitoring networks and develop spatial climate datasets to investigate short- and long-term climate patterns. https://prism.oregonstate.edu/

[4] There were 13 covariates in 4 categories: climate (mean annual precipitation, mean summer maximum temperature, mean winter temperature), air pollutants (NO_3^-, SO_4^{2-}, O_3), topography (topographic convergence index, elevation, slope, radiation index), and stand characteristics (stand basal area, stand age, and focal tree DBH).

Table 5B-4. Influence of three air pollutants on pattern of tree mortality for 10 plant functional groups in the eastern and central U.S. (drawn from Dietze and Moorcroft, 2011).

Plant Functional Group	Sulfate, wet deposition	Nitrate, wet deposition	Ozone
Early Successional. Hardwood	Pos	Neg	Pos
Evergreen Hardwood	Pos	Neg	Pos
Hydric	Pos	Neg	Pos
Late Successional Conifer	Neg	Neg	Neg
Late Successional Hardwood	Pos	Neg	Pos
Midsuccessional Conifer		Neg	
Northern Midsuccessional Hardwood		Pos	
Northern Pine	Pos	Neg	Pos
Southern Midsuccessional Hardwood	Pos	Neg	Pos
Southern Pine	Pos	Neg	Pos

In this study, which was limited to the eastern and central U.S., the deposition metrics were based on wet deposition estimates for SO_4^{2-}, as an indicator of acid deposition,[5] and NO_3^-, as an indicator of wet deposition of total N (Dietze and Moorcroft, 2011).[6] As noted by the authors, "[t]he impacts of both acidification and nitrogen deposition on tree mortality result from cumulative, long-term deposition, and the patterns presented here should be interpreted in that light," further noting that "these relationships are not intended to assess the impacts of interannual variability in deposition nor the efficacy of NO_3^- or SO_4^{2-} regulation" (Dietze and Moorcroft, 2011). Different patterns and associations might be found for analyses utilizing total deposition (wet and dry) and for species and locations in the western U.S., with its differing species, soils, climate and historic deposition patterns. In order to utilize all the measurements, including those for species with lower sample sizes, the tree species were categorized into plant functional groups; accordingly, variation in mortality at species level was not assessed.

5B.2.2.2. Thomas et al. (2010)

The study by Thomas et al. (2010) statistically analyzed relationships of growth and survival to N deposition for 24 commonly occurring tree species in a 19-state region of the U.S. The study region included USFS FIA program plots in 19 states, bounded by Maine in the Northeast to Virginia and Kentucky in the South, and west to Wisconsin and Illinois. Data were extracted for the 24 tree species at 20,067 plots. Two measurements were taken at these plots

[5] Preliminary analyses indicated stronger relationship for tree mortality with SO_4^{2-} than with hydrogen ion (Dietze and Moorcroft, 2011).

[6] Preliminary analyses indicated a stronger relationship for tree mortality with NO_3^- than with NH_4 or total N (Dietze and Moorcroft, 2011).

during the period from the 1978 to 2001, with the measurement interval varying across the 19 states from 8.3 to 14.4 years (Thomas et al., 2010, Supplemental Information).

Nitrogen deposition was estimated using NADP wet deposition estimates and CASTNET dry deposition estimates for the period from 2000 through 2004. Total N deposition estimates at the study plots for this period ranged from 3 to 11 kg N/ha-yr (Thomas et al., 2010, Supplemental Information). Precipitation and temperature were calculated from PRISM with plot specific values for the span of years from first measurement to second measurement. The statistical analyses tested a suite of alternate regression models for growth and survival response to N deposition, precipitation and temperature. The Akaike Information Criteria (AIC) were used to select the most parsimonious model (i.e., the best model fit for the fewest parameters).

Variation in tree growth for 14 of the 24 species was found to be significantly associated with N deposition, with positive associations (greater growth at sites with greater N deposition) found for 11 species and negative associations for three species. All three species with negative associations were evergreen conifers (red pine, red spruce, and white cedar) that varied widely in the amount of growth variation per kg N/ha-yr from -9% for red pine to -0.1% and -0.01% for the other two species, respectively (Thomas et al., 2010). Three of the four most abundant species (red maple, sugar maple and northern red oak) exhibited strong positive associations. The largest variation in growth per unit variation in the N deposition metric was observed for black cherry, tulip poplar, scarlet oak, white ash and balsam fir (18 to 12.3% difference in growth per kg N/ha-yr).

With regard to probability of tree survival, variation in survival probability across the study area was significantly associated with the N deposition metric for 11 of 24 species examined. The association was negative for eight species, with the largest survival variation per kg N/ha-yr observed for scarlet oak (-1.67%) and quaking aspen (-1.3%). The association was positive for three species (red maple, paper birch, and black cherry), with only one of the three having a survival variation per kg N/ha-yr above 1%, black cherry (Thomas et al., 2010).

The authors also suggest that the type of mycorrhizal fungi association with the tree species may influence its response to N deposition as all five species with arbuscular mycorrhizal fungi associations had positive associations of growth with N deposition and all 8 of the species with negative associations of survival with N deposition had ectomycorrhizal fungi associations (Thomas et al., 2010). Mycorrhizal fungi are important for supplying nutrients and water to plants, influencing soil C sequestration, and producing mushrooms (ISA, p. ES-16). Mycorrhizal fungi have long been observed to be sensitive to increased forest N availability (ISA, Appendix 6, section 6.2.3.2).

Not included in the analysis were several factors with the potential to influence tree growth and survival, including competition, soil chemistry, S deposition and ozone. Accordingly,

there was also no analysis of collinearity between such parameters. Most notably, there was no assessment of the extent of N deposition correlation with S deposition and/or ozone. The study area and species list was the most limited of the three observational studies relying on USFS-FIA data.

5B.2.2.3. Horn et al. (2018)

The most recent analysis utilizing the USFS-FIA data, by Horn et al. (2018) also covers the largest area. This study relies on tree measurements taken for approximately 1.4 million trees across approximately 70,000 FIA plots. The plots are scattered across 47 states of the contiguous U.S., excluding Wyoming[7] (Figure 5B-2; Horn et al., 2018, Supplemental Data). The eastern U.S. has many more plots than the West and the areas with highest densities of plots (and associated measurements) include Wisconsin, northern Michigan and Minnesota and New England (Figure 5B-2).[8]

The study investigated associations between variation in tree growth and survival and atmospheric deposition of N and S across the plots for each species using an approach somewhat similar to Thomas et al. (2010). The tree growth and survival measurements were those collected by the FIA generally within the years from 2000 to 2016, with the remeasurement interval for each plot from which measurements were used in the analysis varying by state and inventory cycle from 8.8 to 12.1 years (Horn et al., 2018, Supplemental Data). The most common measurement interval across all plots in the study dataset was 10 years (Horn et al., 2018).

[7] The lack of plots in Wyoming resulted because when the researchers obtained the FIA in January 2017, although there were FIA plots in Wyoming, there were no re-measured plots which is a requirement to assess rates of growth and survival.

[8] This observation is the result of there being more plots in the eastern US due to greater forested area. Within all U.S. forested areas, plot density is the same by the FIA design (Bechteld and Patterson, 2005).

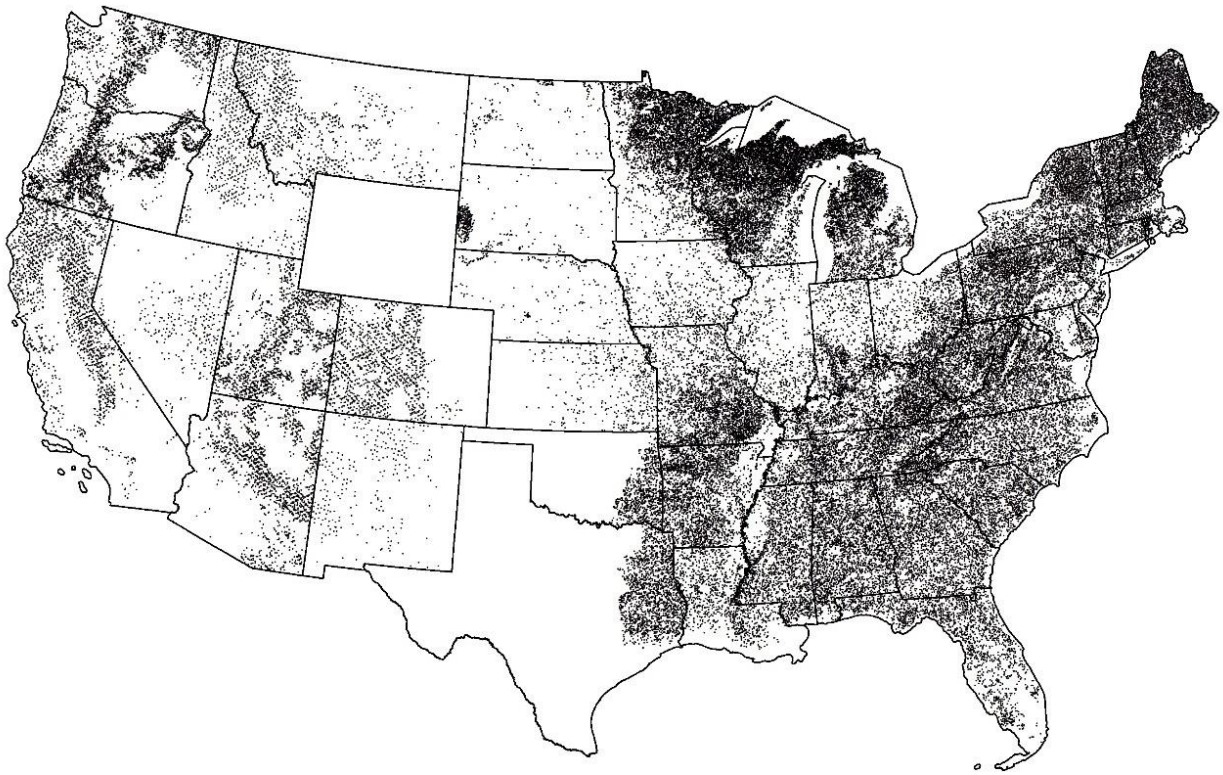


Figure 5B-2. Location of FIA plots, based on survival analysis of Horn et al. (2018).

Individual tree data were available for a total of 151 species, with 94 species meeting the study threshold of 2000 individual trees for both growth and survival data (Horn et al., 2018). Tree growth values were in terms of biomass gains based on measurements of individual trees at the USFS FIA plots during initial and follow-up visits. Survival was assessed by observing whether a tree observed on an initial visit was still alive at the follow-up visit (e.g., survived or not). Thus, survival is a probability metric of the tree surviving and the relationship of survival (y/n) with the average deposition at that site across years between visits was statistically analyzed (along with other co-factors like temperature, precipitation, size, competition, and N or S deposition).

The N and S deposition estimates for each plot's measurement interval were derived from spatially modeled N and S deposition estimates available from the U.S. National Atmospheric Deposition Program's Total Deposition Science Committee (stored on the U.S. EPA's FTP server). Average N deposition and S deposition for each plot were derived from the annual deposition estimates for the years included in the measurement interval (from year of first measurement to year of follow-up measurement) for the plot. The plot-level deposition estimates

were assigned to all the trees in that plot. Temperature and precipitation values were obtained from PRISM Climate Group[9] and assigned to individual plot values, as for N and S deposition.

In addition to temperature and precipitation, other parameters analyzed in the statistical models included tree size and competition. A total of 5 different models of growth as a function of various sets of the 7 parameters were examined: 1) a full model with the size, competition, climate, S deposition, and N deposition terms; 2) a model with all terms except the N deposition term; 3) a model with all terms except the S deposition term; 4) a model with all terms but without S and N deposition terms; and 5) a null model that estimated a single parameter for the mean growth parameter. For survival, a total of 9 different models were examined, the same 5 as for growth plus additional models using 2 different size estimates. S deposition was constrained to have a flat or decreasing response while N deposition could have flat, increasing or decreasing effects. The models selected to describe growth and survival for each species were the simplest models (i.e., the one with the fewest parameters) that were within 2.0 AIC units of the best model (i.e. the model with the lowest AIC) following Thomas et al. (2010).

To quantify collinearity of N and S deposition against other environmental variables, the study calculated variance inflation factors (VIF). This was done for each tree species and for both growth and survival. While VIF values from 3 to 10 have been presented in the literature as a threshold for high collinearity, the authors used VIF < 3 as a criterion for species inclusion (Horn et al., 2018). The growth and/or survival models for 71 of the 94 species analyzed met this criterion. Although not utilized in selecting the model for each species, correlation coefficients were calculated for N and S deposition across the plots assessed for that species (Horn et al., 2018, Supplemental Information).

Of the 71 species, growth of 31 and survival probability for 40 were negatively associated with the S deposition metric values. For 21 species, both growth and survival were negatively associated with S deposition. No statistically significant association was observed for growth or survival in 5 of the 71 species (Horn et al., 2018). With regard to N, among the statistically significant models for growth and survival for some species were hump-shaped relationships, with positive associations in the lower part of the range of N deposition estimates for a species and negative associations in the upper part of the range. This was the case for growth and N deposition for 17 species and for survival of 25 species. Growth for two species and survival for six was negatively associated with the N deposition metric across their ranges. Conversely, positive associations across the full range were found for growth of 20 species and

[9] The PRISM climate group at Oregon State University, supported by the USDA, collects climate data and applies modeling techniques to develop publicly available datasets covering the period from 1895 to the present. The Parameter-elevation Relationships on Independent Slopes Model (PRISM) is an interpolation method used in developing the data. (PRISM Climate Group, Oregon State University, https://prism.oregonstate.edu).

survival of one species, black locust (Robinia pseudoacacia),[10] which was also among the 20 species with positive growth associations.

Multiple factors with potential impacts on tree growth/survival were not assessed, including ozone and others, such as disturbance history (Latty et al., 2004) and insect infestation (Eshleman et al., 1998, 2004). Further, the influence of soil characteristics on growth or survival was also not analyzed. Whether these factors may be correlated with the N/S deposition metrics values and any effect on the reported associations is unknown. Significantly, the study does not account for the influence at the FIA plots of higher historical deposition. So the extent to which observed associations relate to historically higher deposition is unclear. Thus, the extent to which relationships reported for N and S deposition could have had unaccounted for influences of these variables and associated impacts is unknown.

The authors express strongest confidence in findings from this gradient analysis for the Eastern U.S., noting the smaller gradients in deposition and smaller number of different species at western plots (Horn et al., 2018). Plots for some species (e.g., Utah juniper, Douglas fir) were only in the West (Table 5B-5), FIA plots for some other species are predominantly in the Eastern U.S. (northeast, mid-atlantic or south), or in the Midwest (e.g., upper Great Lakes areas). Given the lesser confidence for species only at western plots, we have focused discussion below on the species for which the sample sites were not limited to the western U.S.

Table 5B-5. Species with significant growth or survival associations with S or N deposition for which FIA sites are only in western states (drawn from Horn et al., 2018).

All FIA assessment sites in western states	
Genus species	**Common name**
Juniperus osteosperma	Utah juniper
Lithocarpus densiflorus	Tanoak
Pinus monophyla	Singleleaf pinyon
Pseudotsuga menziesii	Douglas fir
Tsuga heterophylla	Western hemlock

Examination of the correlation coefficients additionally indicates relatively high N/S correlations for some species, complicating interpretation. For example, across the 71 species, the two highest correlation coefficients are those for eastern hemlock (0.78) and American beech (0.76), and four of the six species with the next highest coefficients are also for species whose ranges are concentrated in the eastern U.S. (pond cypress [0.71], yellow birch [0.7], sugar maple [0.67],and pitch pine [0.66]) (Horn et al., 2018, Supplemental Information). Differences in

[10] More than 90% of sample sites for this species were in ecoregions 8.1 – 8.4, with more than 50% in 8.4 (Ozark, Ouachita-Appalachian Forests), regions heavily impacted by SO_2 and acid deposition in the past (ISA, Figure 2-70); the N/S correlation coefficient for these sampling sites was 0.18 (Horn et al., 2018, Supplemental Figures).

quantitative relationships among species may reflect, in part, differences in geographic distribution of sampling locations, with some species' sites largely concentrated in just a couple of ecoregions (e.g., paper birch in the far north Great Lakes and Appalachians). Thus, differences in geographic distributions of the species contribute to differences in ranges of deposition history, geochemistry, etc. and may contribute to findings reported for some species.

Across sites of species with statistically significant associations of growth or survival with the S deposition metric, the median average measurement-interval S deposition value,[11] with a few exceptions, ranged from 5 to 12 kg S $ha^{-1}yr^{-1}$. Focusing first on association for growth, the median S deposition metric values for the species for which growth was negatively associated with S deposition (excluding the two species with samples only in the west) ranged from 4 to 12 kg S $ha^{-1}yr^{-1}$, with values below 5 kg S $ha^{-1}yr^{-1}$ for two species, paper birch and white spruce (for which 75-80% of sites were in the Northern Forests ecoregion[12]), and above 10 kg S $ha^{-1}yr^{-1}$ for two species, black locust and sweet birch, which have 70% to more than 90% of their sites in the Eastern Temperate Forests ecoregion[13] (Figure 5B-3; distribution of measurement sites shown in Attachment 2A).

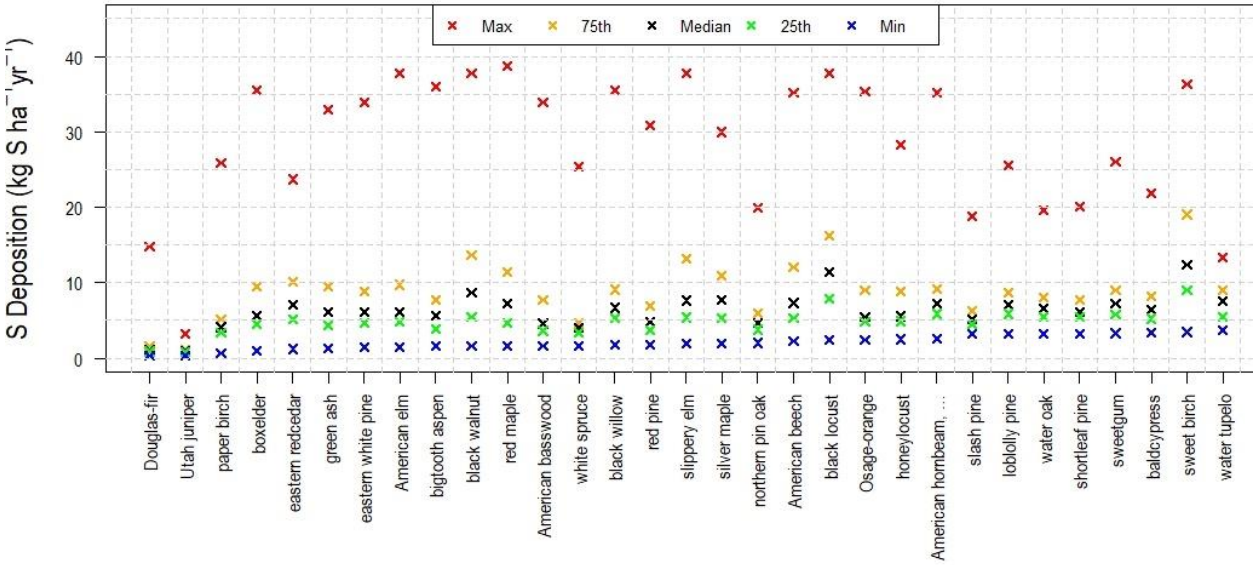


Figure 5B-3. Average measurement interval S deposition at sites of species with negative growth associations with S deposition metric (drawn from Horn et al., 2018).

[11] Median average measurement-interval S and N deposition values cited in this document are rounded to whole numbers.

[12] The Northern Forests is the level 1 ecoregion (5.0), which in the U.S. is located in northern Michigan, Wisconsin and Minnesota (https://www.epa.gov/eco-research/ecoregions-north-america).

[13] Eastern Temperate Forests is the level 1 ecoregion (5.0), which includes most of the eastern U.S. (https://www.epa.gov/eco-research/ecoregions-north-america).

The median deposition metric values for the 40 species for which survival probability was negatively associated with S deposition ranged from 3 to 12 kg S ha $ha^{-1}yr^{-1}$ (Figure 5B-4). Values for ten species were at or above 10 and for two were below 5 kg S ha $ha^{-1}yr^{-1}$. The two values below 5 were for paper birch, for which nearly 80% of the measurement sites were in the Northern Forests ecoregion, and quaking aspen, for which more than 60% of the sites were in the Northern Forests ecoregion and another 16% were in the Southern Rockies and Wasatch and Uinta Mountains (see sample distribution in Attachment).

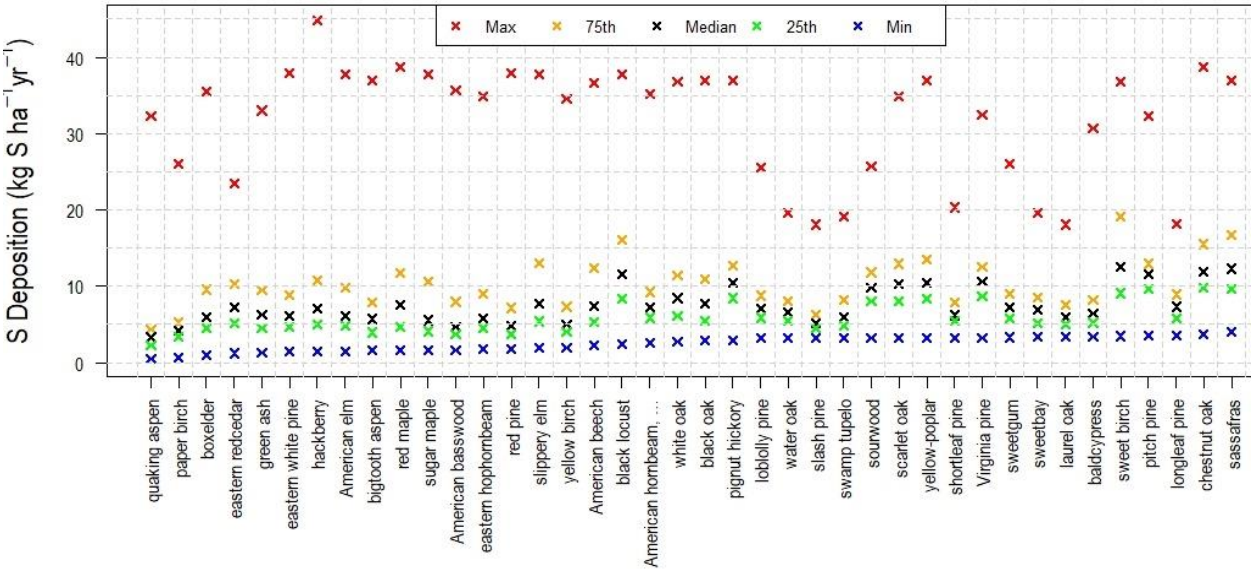


Figure 5B-4. Average measurement-interval S deposition at sites of species with negative survival associations with S deposition metric (drawn from Horn et al., 2018).

With regard to N deposition, of the 39 species with significant associations of growth with N deposition, the association was negative across the full deposition range of their sites for two species, pitch pine and bur oak. These species' sites were predominantly in the Atlantic coastal pine barrens and northern plains and forests, respectively. The median deposition across all sites of these species were 9 and 10 kg N ha $ha^{-1}yr^{-1}$ (Figure 5B-5). The median deposition values for the two other species, with hump shaped functions that were negative at the median,[14] were 7 and 8 kg N ha $ha^{-1}yr^{-1}$, respectively (Figure 5B-5).

[14] Given its role as a measure of central tendency of a dataset, the nature of the association for hump shape models at the median is what is described in the groupings here.

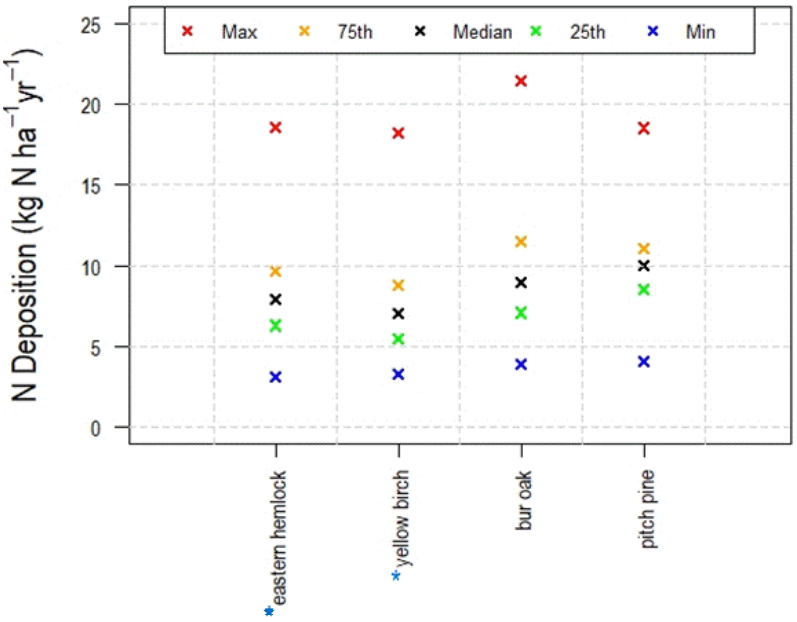


Figure 5B-5. Average measurement-interval deposition at sites of species with negative associations of growth with N deposition metric at median (drawn from Horn et al., 2018). Blue asterisks indicate species with hump shape associations.

Of the remaining 35 species with significant associations of growth with measurement-interval N deposition, the association was positive across the full deposition range of their sites for 20 species. The median N deposition metric values for the 17 nonwestern species[15] of these 20 species ranged from 7 kg N ha ha^{-1}yr^{-1} (for a number of species) up to 12 kg N ha ha^{-1}yr^{-1} for silver maple, hackberry and black walnut (Figure 5B-6). For the 15 species with significant associations of growth with measurement-interval N deposition that were positive at the median average measurement-interval deposition for the species, one was a western species, western hemlock (Table 5B-5). The median average measurement-interval deposition metric values for the other 14 species ranged from 7 to 11 kg N ha ha^{-1}yr^{-1} (Figure 5B-6).

[15] Three western species, Utah juniper, Douglas fir and western hemlock (Table 5B-5) had positive growth association across range of N deposition metric values.

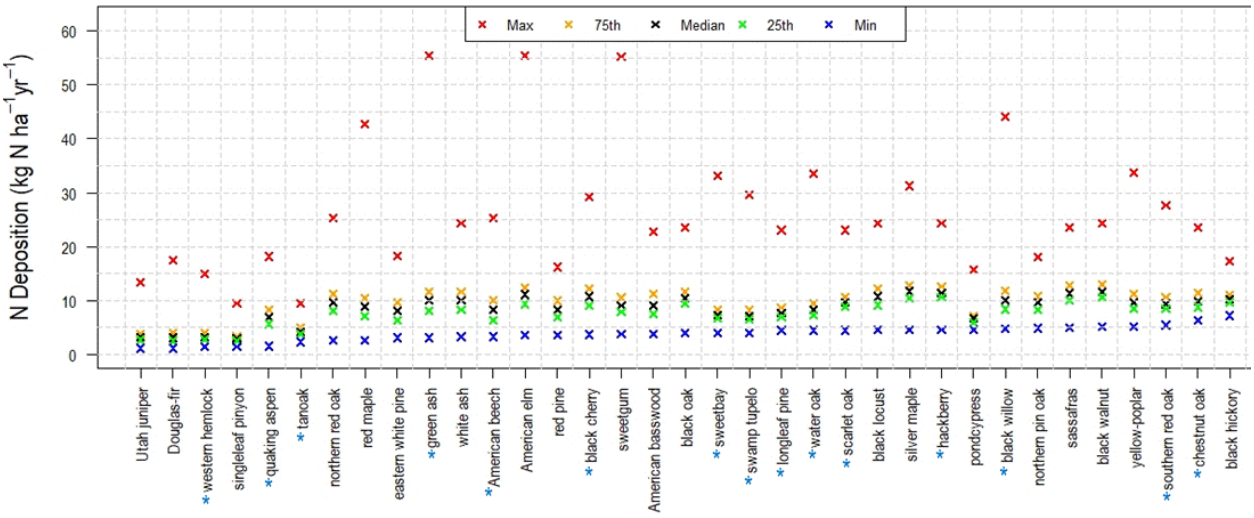


Figure 5B-6. Average measurement-interval deposition at sites of species with positive associations of growth with N deposition metric at median (drawn from Horn et al., 2018). Blue asterisks indicate species with hump shape associations.

Of the six species with negative associations of survival with the N deposition metric across the full range of the N deposition metric (water oak, southern red oak, winged elm, scarlet oak, mockernut hickory and American elm), the median deposition values ranged from 8 to 11 kg N ha ha^{-1}yr^{-1}.(Figure 5B-7). The median deposition values for all of the 21 other species with hump shape functions that were negative at the median deposition value ranged from 3 to 11 kg N ha ha^{-1}yr^{-1} (Figure 5B-7; see blue asterisks). The values for the 19 species for which sample sites were not limited to the western U.S. ranged from 7 to 12 kg N ha ha^{-1}yr^{-1}. The four values below 9 were for quaking aspen (75% sites in Northern Forests, Wasatch and Uinta Mountains and Southern Rockies), slash pine (~60% sites in southern coastal plain), eastern hemlock (~50% sites in Northern Forests and ~30% in Mixed Wood Plains) and red pine (nearly 70% in Northern Forests).

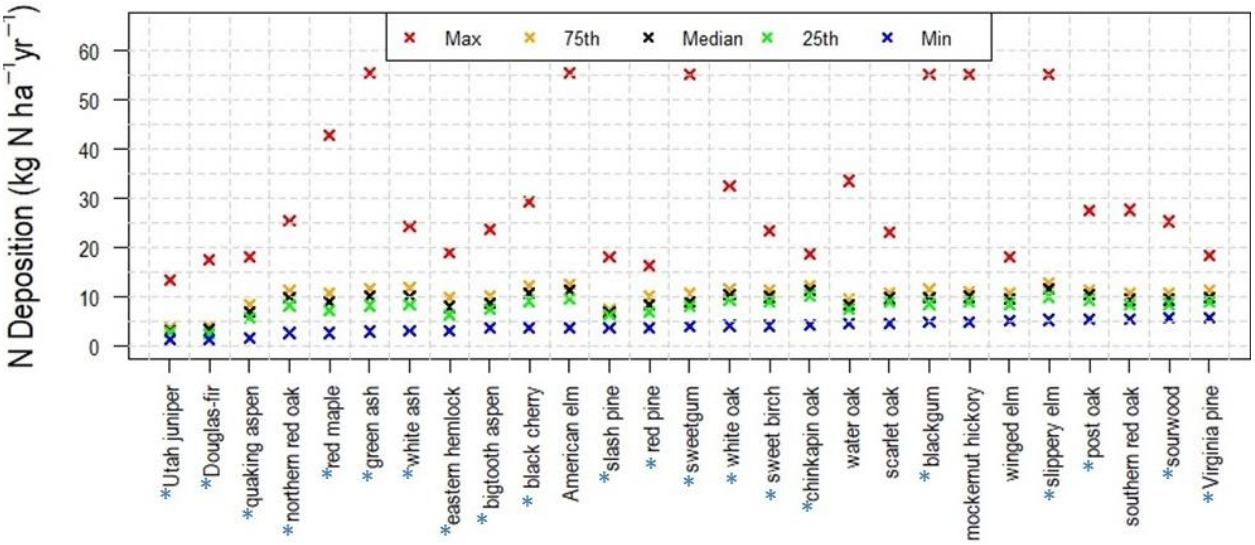


Figure 5B-7. Average measurement-interval deposition at sites of species with negative associations of survival with N deposition metric (drawn from Horn et al., 2018). Blue asterisks indicate species with hump shape associations.

Turning to positive associations of survival with N, there was 1 species (black locust) with a positive associations of survival with N across the full deposition range with a median deposition of 11 kg N ha ha^{-1}yr^{-1} (Figure 5B-8). The median deposition values for the 4 species with hump-shaped associations that were positive at the median ranged from 7 to 12 kg N ha ha^{-1}yr^{-1}. The two values below 10 were for paper birch, for which nearly 80% of the measurement sites were in the Northern Forests ecoregion, and American beech with more than 50% of sites in Northern Forests (and N/S correlation coefficient of 0.76).

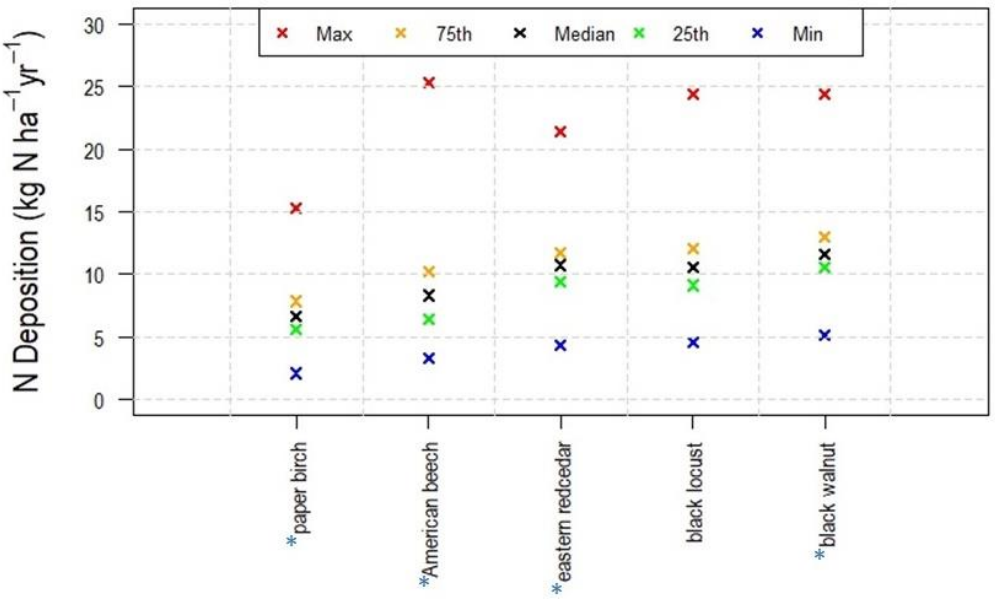


Figure 5B-8. Average measurement-interval deposition at sites of species with positive associations of survival with N deposition metric (drawn from Horn et al., 2018). Blue asterisks indicate species with hump shape associations.

5B.2.3. Tree Growth and Survival: Key Observations, Uncertainties and Limitations

Looking across the array of experimental addition studies and the three recent observational (or gradient) studies, we note a number of key observations and associated uncertainties and limitations:

Experimental Addition Studies of Tree Growth/Survival

- Some studies additionally reported soil chemistry and/or tree cellular responses, which can inform interpretation of responses that may relate to geology and soil chemistry in those locations.
- S or S + N addition: Some multiyear S or S+N addition experiments (>20 kg/ha-yr) with a small set of eastern species, including sugar maple, aspen, white spruce, yellow poplar, black cherry, have not reported detrimental growth effect (Table 5B-1; Bethers et al., 2009; Moore and Houle 2013; Jung and Chang, 2012; Jensen et al., 2014). Some reported increased growth (25.2 kgN + 28.8 kg S/ha-yr for 10 years [Bethers et al., 2009]), while one reported reduced growth in three species after 10 years that resolved in two of the species after 22 years (Jensen et al., 2014).
- N addition: Several studies found mixed results for growth and survival for several eastern species including oaks, spruce, maples and pines (Table 5B-1; Magill et al., 2004; McNulty et al., 2005; Pregitzer et al., 2008; Wallace et al., 2007).

Observational/Gradient Studies of Tree Growth/ Survival

- Newly available in this review are three large observational studies of tree growth/survival and S/N deposition.
- Although ozone was analyzed in one of the three studies, soil characteristics and other factors with potential to impact tree growth and survival (other than climate) were not assessed.
- S deposition: Two large studies that analyzed growth and/or survival measurements in 94 and 267 species, respectively, at sites across the country, or in the eastern half of the country, describe negative associations of tree survival and growth with the S deposition metric for nearly half the species individually and negative associations of tree survival for 9 of the 10 species' functional type groupings (Dietze and Moorcroft, 2011; Horn et al., 2018). Survival for the same 9 species groups was also negatively associated with long-term average ozone (Dietze and Moorcroft, 2011).
  - The S deposition metrics were derived from estimates for total S or sulfate in overlapping time periods of roughly 10 years and include areas, particularly in the eastern U.S., that have experienced decades of much higher deposition.
    - The full range of average SO_4^{2-} deposition estimated for the 1994-2005 time period and eastern U.S. study area assessed by Dietze and Moorcroft (2011) is 4 to 30 kg S $ha^{-1}yr^{-1}$.
    - The full range of average total S deposition estimates for the 2000-2013 time period and sites across the U.S. assessed by Horn et al. (2018) is 0.2 to 45 kg S $ha^{-1}yr^{-1}$ (Horn et al., 2018, Supplemental Information).
      - The median S deposition for sites of nonwestern species with neg associations with growth or survival ranged from 5 to 12 kg S $ha^{-1}yr^{-1}$, with few exceptions (Horn et al., 2018).
  - The extent to which the differences in growth or survival across sites with different deposition estimates relate to historically higher deposition at the sites (e.g., *versus* the deposition metrics analyzed) is unknown. There are few available studies describing recovery of historically impacted sites (e.g., ISA, section IS.4.1, IS.5.1, IS.11.2).
- N deposition: Three large studies that analyzed growth and/or survival measurements in 24 to 267 species at sites in the northeastern or eastern U.S., or across the country, describe associations of tree survival and growth with several N deposition metrics (Dietze and Moorcroft, 2011; Thomas et al., 2010; Horn et al., 2018).
  - The N deposition metrics were derived from estimates for total N or nitrate in overlapping time periods and include areas that have experienced decades of much higher deposition.
    - The full range of average NO_3^- deposition estimated for the 1994-2005 time period) and eastern U.S. study area assessed by Dietze and Moorcroft (2011) is 6 to 16 kg N $ha^{-1}yr^{-1}$.

- The full range of average total N deposition estimates for the 2000-2013 time period and sites across the U.S. assessed by Horn et al. (2018) is 0.9 to 55 kg N $ha^{-1}yr^{-1}$ (Horn et al., 2018, Supplemental Information).
  - The median N deposition for sites of nonwestern species for which associations with growth or survival were negative (either over full range or at median for species) ranged from 7 to 12 kg N $ha^{-1}yr^{-1}$ (Horn et al., 2018).
  - The median N deposition for sites of nonwestern species for which associations with growth or survival were positive (either over full range or at median for species) ranged from 7 to 12 kg N $ha^{-1}yr^{-1}$ (Horn et al., 2018).
- The extent to which the associations of growth or survival with site-specific N deposition estimates relate to historic patterns of N or S deposition at the sites (e.g., *versus* the specific magnitude of the N deposition metrics analyzed) is unknown.

Only a very small subset of the 71 species of Horn et al. (2018) have been previously studied with regard to S deposition and growth or survival, although the study by Dietze and Moorcroft (2011) included these species in its groupings by plant functional type (Table 5B-6). With regard to relationships of tree growth or survival with N deposition metrics, some of the Horn et al. (2018) species were also assessed in the study by Thomas et al. (2010), as well as all of the species being included in the groupings of Dietze and Moorcroft (2011). Table 5B-6 indicates a similarity in the findings, particularly of Horn et al. (2018) and Dietze and Moorcroft (2011), although the time period and estimation approach for S and N deposition differ.

Given the role of deposition in causing soil conditions that affect tree growth and survival, and a general similarity of spatial variation of recent deposition to historic deposition, the similarity in the two studies' findings may indicate the two different metrics to both be reflecting geographic variation in impacts stemming from historic deposition. Although the spatial patterns are somewhat similar, the magnitudes of S and N deposition in the U.S. have changed appreciably over the time period covered by these studies. An example of this is illustrated by the patterns of wet deposition of SO_4^{2-} and NO_3^- in Figures 5B-9 and 5B-10, respectively, and patterns of total S and N deposition in Figures 5B-11 and 5B-12, respectively. The appreciable differences in magnitude across the time periods contribute uncertainty to interpretations related to specific magnitudes of deposition associated with patterns of tree growth and survival.

Differences in findings of Thomas et al. (2010) may be related to the much shorter N deposition time period used, as compared to those of Horn et al. (2018) and Dietze and Moorcroft (2011). The findings of unimodal or hump-shape associations for Horn et al. (2018) for species with positive or negative associations in Thomas et al. (2010) may also reflect

different time periods assessed. The time period for the deposition metric in Thomas et al. (2010), 2000-2004, overlaps with the earliest five years of the longer time period within which the measurement intervals for Horn et al. (2018) fall. Further, the occurrence of negative and positive survival or growth associations from Thomas et al. (2010) and Horn et al. (2018) for species in a plant functional grouping for which Dietze and Moorcroft (2011) found negative association may reflect difference in study areas, e.g., early successional hardwood, which had a positive association of survival with N, includes quaking aspen for which Thomas et al. (2010) reported negative survival association. The study area of Thomas et al. (2010) was limited to the Northeast, however, while aspen is prevalent in the Northern Forests ecoregion, which is included in Dietze and Moorcroft (2011) study area.

Table 5B-6. Significant associations in the three studies using USFS tree measurements.

Species	S Deposition		N Deposition		
	Dietz and Moorcroft (2011) (SO_4^{2-}, wet, 1994-2005)	Horn et al. (2018) (total S, ~2000-2013)	Dietze and Moorcroft (2011) (NO_3^-, wet, 1994-2005)	Thomas et al. (2010) (total N, 2000-2004, FIA data,1970s-90s)	Horn et al. (2018) (total N, ~2000-2013)
	Positive (↑) or negative (↓) association for growth (G) or survival[A] (Su)				
Early Successional Hardwood	↓Su		↑Su		
Betula alleghaniensis, yellow birch		↓Su		Small ↓Su	↓G
Betula lenta		↓Su ↓G			U Su
Betula papyrifera, paper birch		↓Su ↓G		Small ↑Su	U Su
Gleditsia triacanthos		↓G			
Liquidambar styraciflua		↓Su			U Su
Maclura pomifera		↓G			
Populus grandidentata, bigtooth aspen		↓Su ↓G		Small ↓Su	U Su
Populus tremuloides, quaking aspen		↓Su		↓Su ↑G	U Su U G
Prunus serotina, black cherry				↑Su ↑G	U Su U G
Salix nigra		↓G			U G
Late Successional Hardwood	↓Su		↑Su		
Acer negundo, boxelder		↓Su ↓G			
Acer rubrum, red maple		↓Su ↓G		small ↑Su ↑G	↑G
Acer saccharum, sugar maple		↓Su		↑G	
Acer saccharinum, silver maple		↓G			↑G
Carpinus caroliniana, American hornbeam		↓Su ↓G			
Oxydendrum arboreum, sourwood		↓Su			U Su
Tilia americana, American basswood		↓Su ↓G		Small ↓Su	↑G
Northern Midsuccessional Hardwood			↓Su		
Celtis occidentalis, hackberry		↓Su			U G
Fraxinus americana, white ash				↑G	U Su ↑G
Fraxinus pennsylvanica, green ash		↓Su ↓G			U Su U G
Juglans nigra, black walnut		↓G			U Su
Quercus alba, white oak		↓Su			U Su
Quercus ellipsoidalis, northern pin oak		↓G			↑G
Quercus rubra, northern red oak				small ↓Su ↑G	U Su ↑G
Quercus velutina, black oak		↓Su			↑G
Sassafras albidum, sassafras		↓Su			↑G
Ulmus americana, American elm		↓Su↓G			↓Su ↑G
Ulmus rubra, slippery elm		↓Su ↓G			
Hydric	↓Su		↑Su		
Nyssa aquatica		↓G			
Nyssa biflora		↓Su			U G
Taxodium ascendens					↑G
Taxodium distichum		↓Su ↓G			

Species	S Deposition		N Deposition		
	Dietz and Moorcroft (2011) (SO_4^{2-}, wet, 1994-2005)	Horn et al. (2018) (total S, ~2000-2013)	Dietze and Moorcroft (2011) (NO_3^-, wet, 1994-2005)	Thomas et al. (2010) (total N, 2000-2004, FIA data,1970s-90s)	Horn et al. (2018) (total N, ~2000-2013)
	Positive (↑) or negative (↓) association for growth (G) or survival[A] (Su)				
Southern Midsuccessional Hardwood	↓Su		↑Su		
Carya alba, mockernut hickory					↓Su
Carya glabra, pignut hickory		↓Su		↑G	
Carya texana, black hickory					↑G
Liriodendron tulipifera, yellow poplar		↓Su		↑G	↑G
Nyssa sylvatica, black gum					U Su
Quercus coccinea, scarlet oak				↓Su ↑G	U Su U G
Quercus falcata, southern red oak					↓Su
Quercus laurifolia, laurel oak		↓Su			
Quercus muelenbergii, chinkapin oak					U Su
Quercus nigra, water oak		↓Su ↓G			↓Su U G
Quercus prinus, chestnut oak		↓Su		Small ↓Su	U G
Quercus stellata, post oak					U Su
Ulmus alata, winged elm		↓Su			
Evergreen Hardwood	↓Su		↑Su		
Magnolia virginia		↓Su			U G
Midsuccessional Conifer			↑Su		
Picea rubens, red spruce				Small ↓G	
Picea glauca, white spruce		↓G			
Pseudotsuga menziesii, Douglas fir		↓G			U Su ↑G
Late Successional Conifer	Weak ↑ Su		↑Su		
Abies balsamea, balsam fir				↑G	
Juniperus virginiana, eastern redcedar		↓Su ↓G			U Su
Thuja occidentalis, northern white cedar				Small[B] ↓G	
Tsuga canadensis, eastern hemlock		↓G			
Northern Pine	↓Su		↑Su		
Pinus resinosa, red pine		↓Su ↓G		↓G	U Su, ↑G
Pinus regida, pitch pine		↓Su			U G
Pinus strobus, eastern white pine		↓Su ↓G		small ↓Su small ↑G	↑G
Southern Pine	↓Su		↑Su		
Pinus echinata, shortleaf pine		↓Su ↓G			
Pinus elliotti, slash pine		↓Su ↓G			U S
Pinus palustris, longleaf pine		↓Su			U G
Pinus taeda, loblolly pine		↓Su ↓G			
Pinus virginiana, Virginia pine		↓Su			U Su

[A] For Dietze and Moorcroft (2011), an up arrow is shown for survival if they reported a negative association with mortality.
[B] For Thomas et al. (2010), "small" used when growth or survival response per unit N is <1%e.
For Horn et al. (2018) "U" used for unimodal (or hump-shaped) associations (positive at lower deposition values and negative at higher).

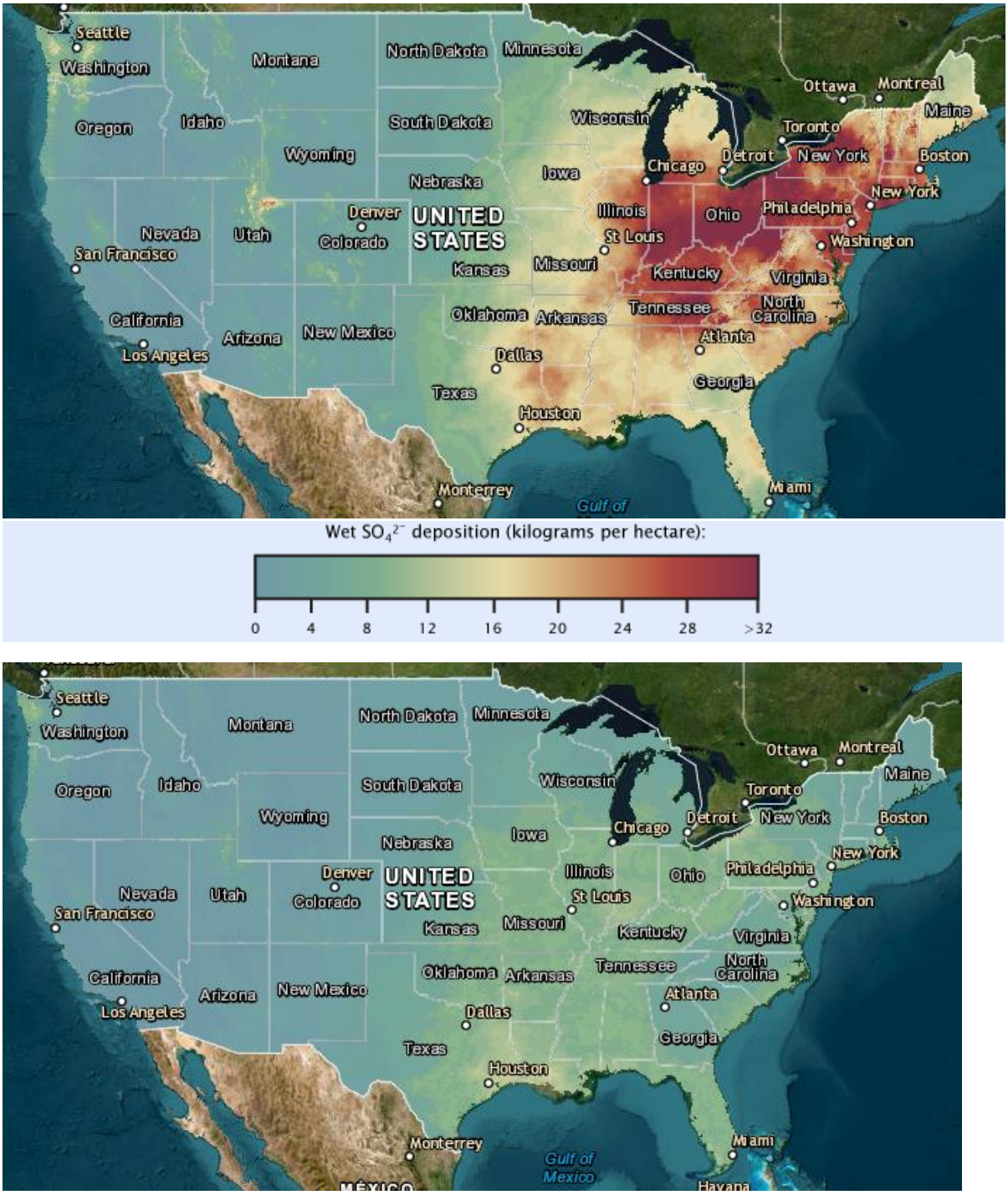


Figure 5B-9. Annual mean wet SO_4^{2-} deposition in the U.S. for 1989-1991 (top panel) and 2014-2016 (bottom panel) (U.S. EPA, 2023; NADP, 2018).

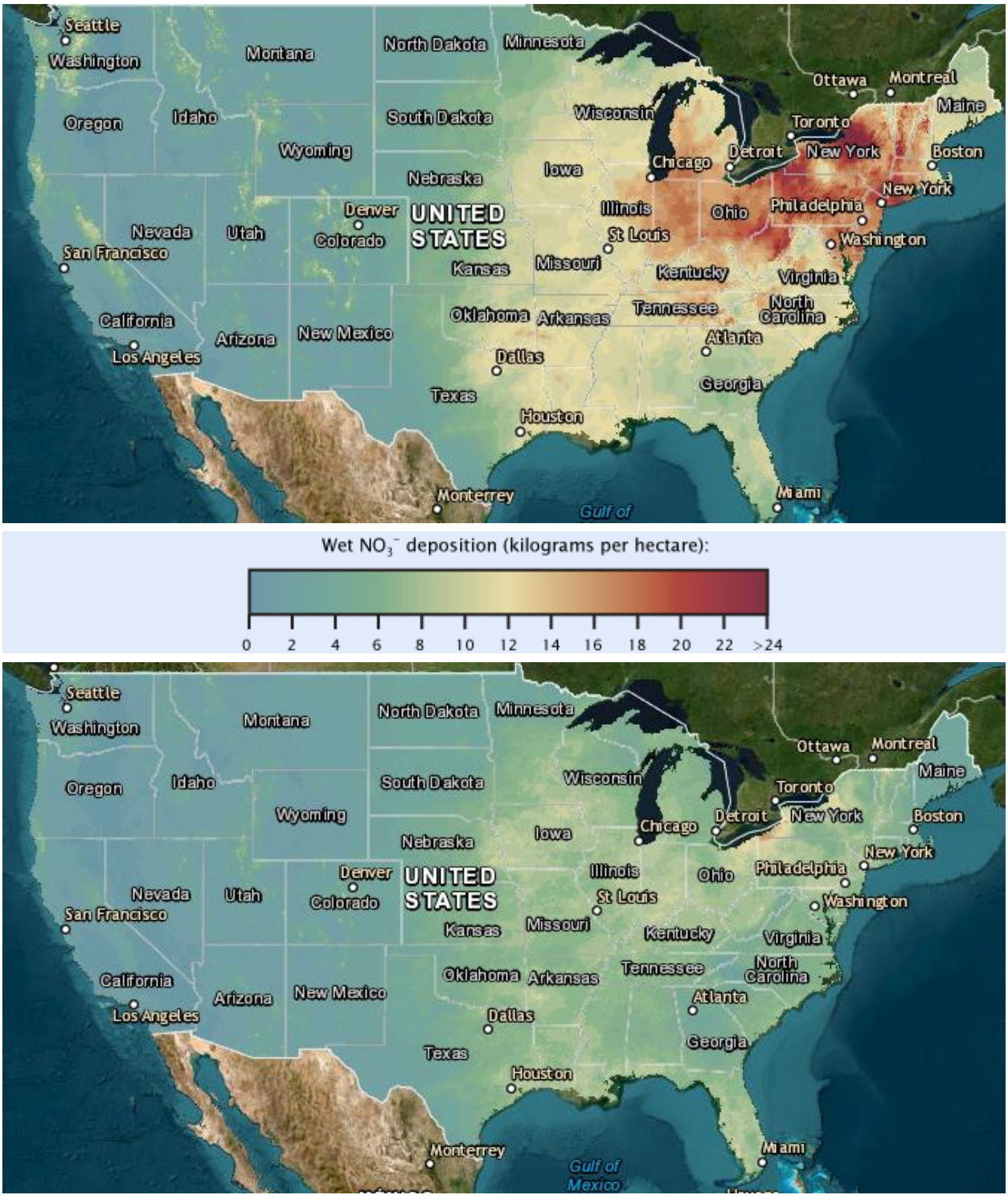
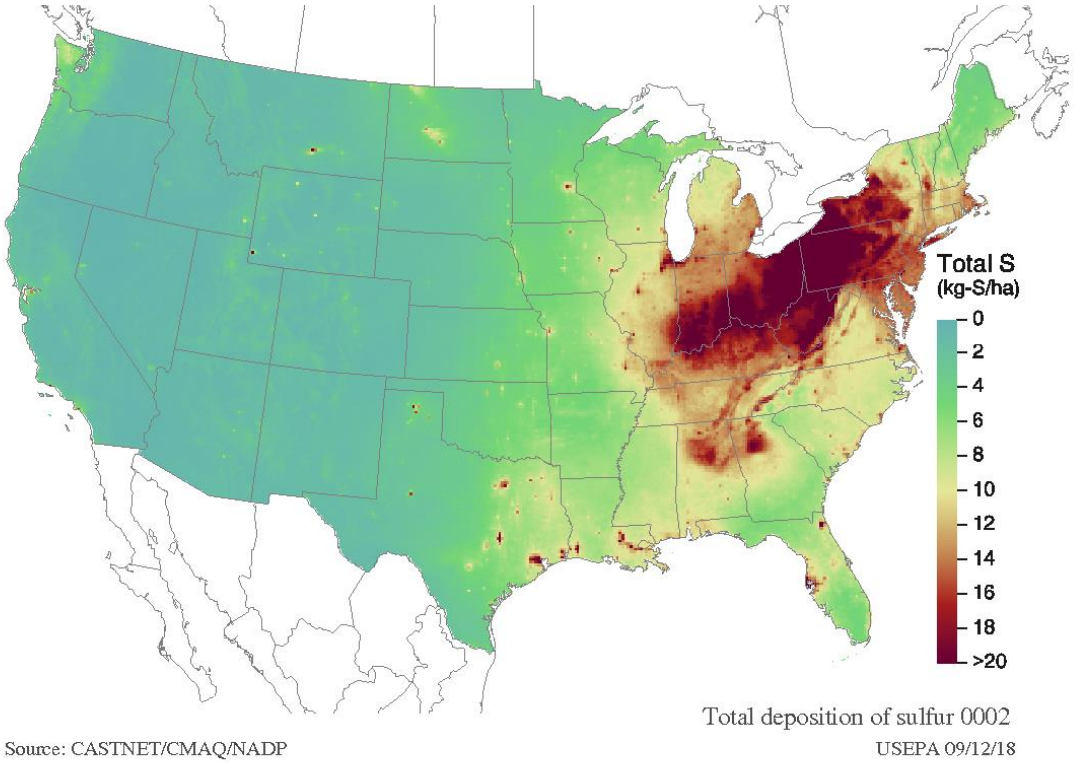


Figure 5B-10. Annual mean wet NO_3^- deposition in the U.S. for 1989-1991 (top panel) and 2014-2016 (bottom panel) (U.S. EPA, 2023; NADP, 2018).



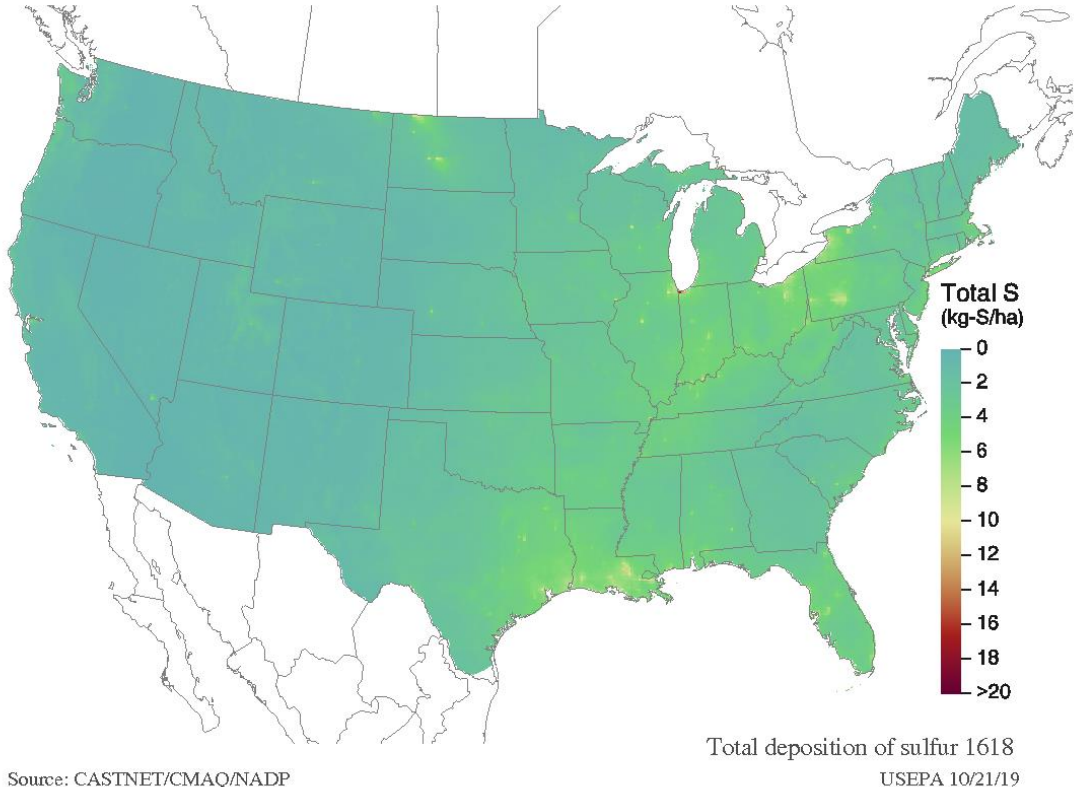


Figure 5B-11. Wet plus dry deposition of total sulfur over 3-year periods. Top: 2000-2002; Bottom: 2016-2018. Drawn from the ISA, Figure 2-70.

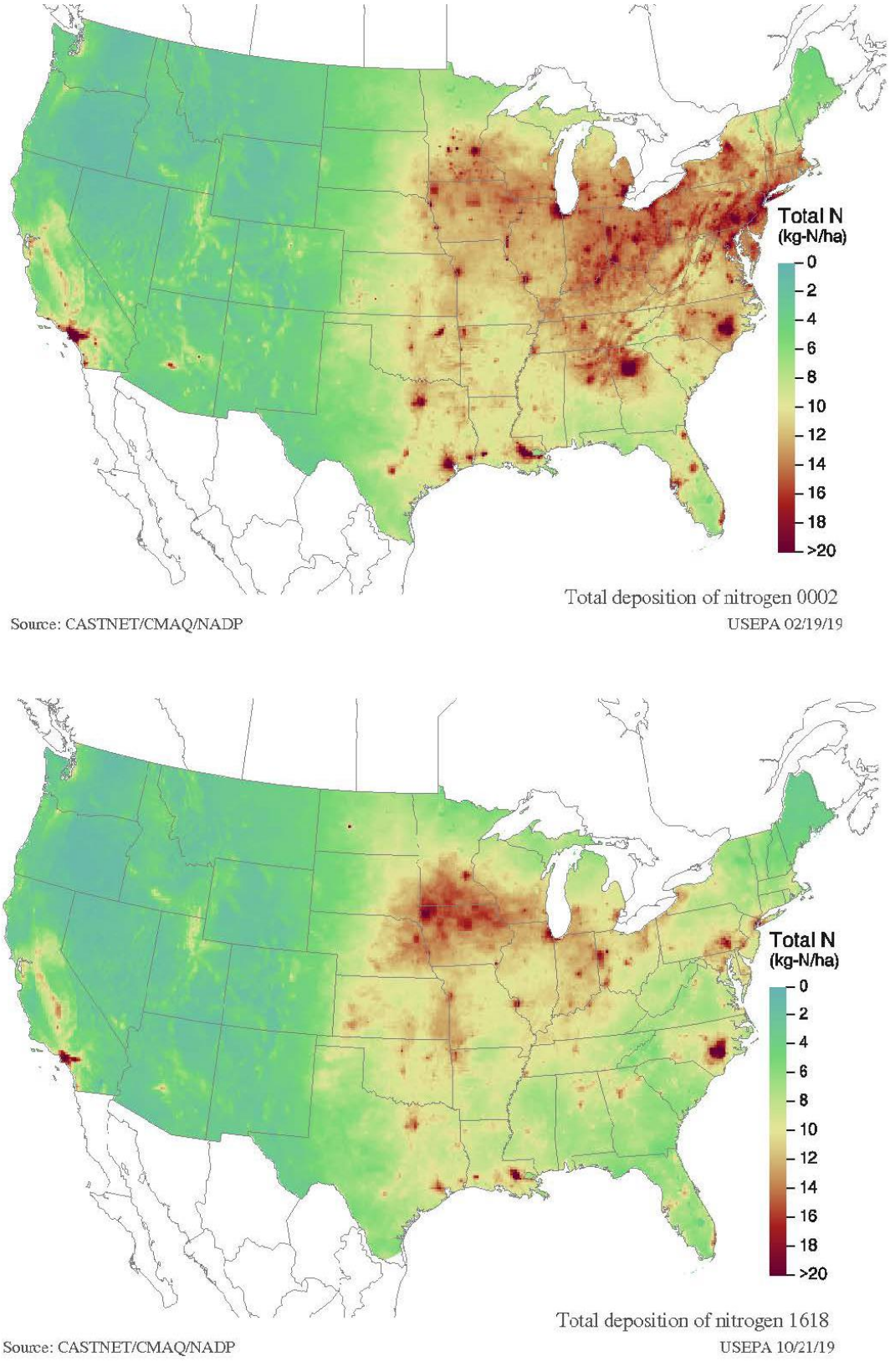


Figure 5B-12. Wet plus dry deposition of total nitrogen over 3-year periods. Top: 2000-2002; Bottom: 2016-2018. Drawn from the ISA, Figure 2-51.

5B.3 SPECIES RICHNESS OF HERB AND SHRUB COMMUNITIES

The subsections below summarize salient aspects of studies that have assessed herb and shrub community metrics and their relationship to N deposition. The addition studies in section 5B.3.1 below evaluated the impact of fertilizer treatments using ammonium nitrate. Section 5B.3.2 summarizes the few recent observational studies that statistically analyze variation in species richness metrics with variation in N deposition, while also providing detailed information regarding the largest such study (Simkin et al., 2016). We note that as species richness is the number of species and does not convey information about species composition, an increase in species richness may reflect only the addition of new species or a combination of additions and subtractions, with a net positive result. The extent to which the observational studies account for potential influence of S deposition varies.

5B.3.1. Experimental Addition Studies

A number of experimental addition studies focused on N (e.g., through addition of ammonium nitrate fertilizer) are discussed in the ISA and summarized in Table 5B-7 below.

Table 5B-7. Experimental addition studies assessing herb and shrub community responses.

Location	Description	Additions	Findings
Joshua Tree National Park, in Mojave desert, CA (Allen et al., 2009)	Assessed biomass and % cover responses of native and non-native grasses to two fertilization levels at four sites	5 and 30 kg N $ha^{-1}yr^{-1}$ as ammonium nitrate (NH_4NO_3) fertilizer over 2 years Ambient air deposition was estimated to be approximately 5 – 8 kg N $ha^{-1}yr^{-1}$	In 1st year, non-native grass biomass increased significantly at three of the four study sites receiving 30 kg N/ha/yr. No significant change with 5 kg N/ha/yr; of with either dose in 2nd year. No change in % cover. Native grass species richness increased with 30 kg N/ha-yr at 1 site that authors judged related to lower nonnative species presence.
Prairie grasslands in Cedar Creek Ecosystem Science Reserve, MN (Clark and Tillman, 2008)	Study plots in two prairie-like successional grasslands and one native savanna grassland. The soils were limed to maintain constant pH (and avoid acidification).	10, 20, 34, 54 and 95 kg N $ha^{-1}yr^{-1}$ (ammonium nitrate addition) over 23 years (1982 to 2004). Background wet deposition of N was estimated to have averaged 6 kg N $ha^{-1}yr^{-1}$ wet deposition.	Species numbers declined with increasing chronic addition, including at the lowest addition (10 kg N/ha/yr). In a subset of plots for which additions were ceased after 10 years, relative species numbers increased, converging with controls after 13 years. Little recovery species composition was observed.
Dry sedge meadow in Rocky Mountain National Park, CO	Five replicate plots (20 total) in a dry meadow community. Study assessed plant species richness, cover of vascular plants, above	5, 10 and 30 kg N $ha^{-1}yr^{-1}$ (ammonium nitrate addition) over 4 years starting in 2006.	No significant effect on plant species richness or diversity. No significant effect on foliar % N or above ground biomass. Based on *Carex rupestris* increasing in cover from 34 to 125% in response to additions, authors estimated 3 kg

Location	Description	Additions	Findings
(Bowman et al., 2012)	ground biomass, and soil chemistry.	Background deposition was estimated to be 4 kg N $ha^{-1}yr^{-1}$	N/ha-yr as deposition associated with an increase in *C rupestris* cover and 9 - 14 kg N/ha-yr with NO_3^- leaching in soil solution.
Santa Margarita Ecological Reserve, Riverside, California (Vourlitis, 2017)	Study of long term effects of N deposition on native and exotic plant cover in coastal sage scrub communities. 4 control and 4 addition plots (10 x 10 m)	50 kg N $ha^{-1}yr^{-1}$ over 13 years. Background deposition estimated at 4 – 6 kg N $ha^{-1}yr^{-1}$	Increase in the native shrub *Artemesia californica* in the 4th and 5−9th yr of the 13-yr experiment; decrease in the native shrub *Salvia mellifera* in the 4th and 11−13th yr; increase in the exotic plant *Brassica nigra* in the 11−13th yr
Santa Margarita Ecological Reserve, Riverside, California and Sky Oaks Field Station, San Diego County, CA (Vourlitis and Pasquini, 2009)	Study of effects of N deposition on plant community composition in coastal sage scrub and chaparral communities. 4 control and 4 addition plots (10 x 10 m) at each site (16 total)	50 kg N $ha^{-1}yr^{-1}$ for 5 years as granular NH_4NO_3 (2003–2006) or $(NH_4)2SO_4$ (2007–2008). Background deposition estimated as 6-8 kg N $ha^{-1}yr^{-1}$	Dry season addition of N significantly changed community composition in coastal sage scrub communities, but not in chaparral communities
Great Basin, California (Concilio and Loik, 2013)	Study effects of elevated N deposition on sagebrush steppe communities in 54 paired plots (half control, half with additions).	50 kg N $ha^{-1}yr^{-1}$ for 4 years starting in 2007. Background deposition estimated as 1 – 3 kg N $ha^{-1}yr^{-1}$	Community composition (native species diversity and abundance of the invasive grass *Bromus tectorum*) differed by disturbance history (e.g. fire), but was not affected by N deposition.
Sevilleta National Wildlife Refuge, New Mexico (Collins et al., 2017)	Study of the effect of nighttime warming, winter precipitation and N deposition in 40 plots (3.0 x 3.5 m each) randomly crossed across treatment effect.	20 kg N $ha^{-1}yr^{-1}$ for 7 years starting in 2006. A wildfire burned the plots after the second year. Ambient air deposition was approximately 3 kg N $ha^{-1}yr^{-1}$ [A]	Native desert grass communities were affected by N deposition in the 3 years following the fire, but not in the two years preceding the fire or the last year of the experiment.
Arches National Park, Colorado Plateau, Utah (McHugh et al., 2017)	Study of community composition in a semi-arid grassland	0, 2, 5 and 8 kg N $ha^{-1}yr^{-1}$ for 2 years starting in 2011. Background deposition was estimated as 2 – 3 kg N $ha^{-1}yr^{-1}$	No significant change in community composition or species richness, but did find a strong connection between composition and soil microbial community structure.

[A] As the background deposition was not reported in this publication, we have estimated it as the 2007-09 average deposition based on TDep version 2018.02, using EPA's CL Mapper Tool at: https://www.epa.gov/gcx/about-cl-mapper.

5B.3.2. Gradient or Observational Studies

Recent gradient studies have included analyses investigating the potential of N enrichment in southern California to alter plant community composition through increases in the presence of invasive annual species (ISA, Appendix 6, section 6.3.6). A recent study by Cox et al. (2014) utilized a landscape-level analysis of vegetation change since the 1930s to investigate risk of conversion of coastal sage scrub vegetation to exotic annual grassland and any association with N deposition. The authors concluded that sites with 2002 N deposition estimates (based on CMAQ modeling [Tonnesen et al., 2007]) less than 11 kg N $ha^{-1}yr^{-1}$ were less likely to have converted from Coastal sage scrub to non-native grasslands (ISA, Appendix 6, section 6.3.6; Cox et al., 2014). The authors also evaluated the circumstances associated with recovery of coastal sage scrub communities from exotic annual grassland that was observed in the 1930s maps, and reported that plots in areas where surrounding plots had little or no exotic grassland and 60% cover by coastal sage scrub had increased probability of recovery (Cox et al., 2014). A second study across the same gradient of 2002 N deposition estimates (6.6 to 20.2 kg N $ha^{-1}yr^{-1}$) reported similar observations, finding that sites with N deposition above 10 kg N/ha-yr had lower native species richness (Fenn et al., 2010).

One of the largest studies, by Simkin et al. (2016), analyzed relationships between observed variation in herb and shrub species richness and average N deposition, soil pH, and annual average temperature and precipitation at more than 15,000 forest, woodland, shrubland and grassland sites in multiple regions of the U.S. (Figure 5B-13; Table 5B-8). The study categorized sites into open-canopy and closed-canopy communities and, in a "national" analysis, investigated quantitative relationships between site variation in species richness, assessed over the 23-year period from 1990 to 2013, and in estimates of average N deposition for the "modern" period of 1985 to 2011 (Simkin et al., 2016, Supplemental Information, SI Methods).

Table 5B-8. Key aspects of analysis by Simkin et al. (2016)

Study Area	Community assessments	N Deposition estimates	Other variables considered
Northwestern U.S. (predominantly WA, OR, far north CA, western MT, NV, UT), northeastern CO, MI, mid-Atlantic (MD, VA) and Southeast (NC, SC, GA, FL)	Assessments… >15,000 sites	10-yr average (2002-11) dry deposition from CMAQ added to 27-year average (1985-2011) wet deposition from NADP.	Soil pH, precipitation and temperature (1981-2010)

The site assessments were drawn from seven databases of biological survey data sources, with varied distribution across the states represented. For example, more than a third of the sites were in Minnesota and the Pacific Northwest (WA and OR) and another third in the Carolinas and Virginia; about 100 sites are in the northeastern U.S. (Simkin et al., 2016, Supplemental Information, Table S1; Figure 5B-13).

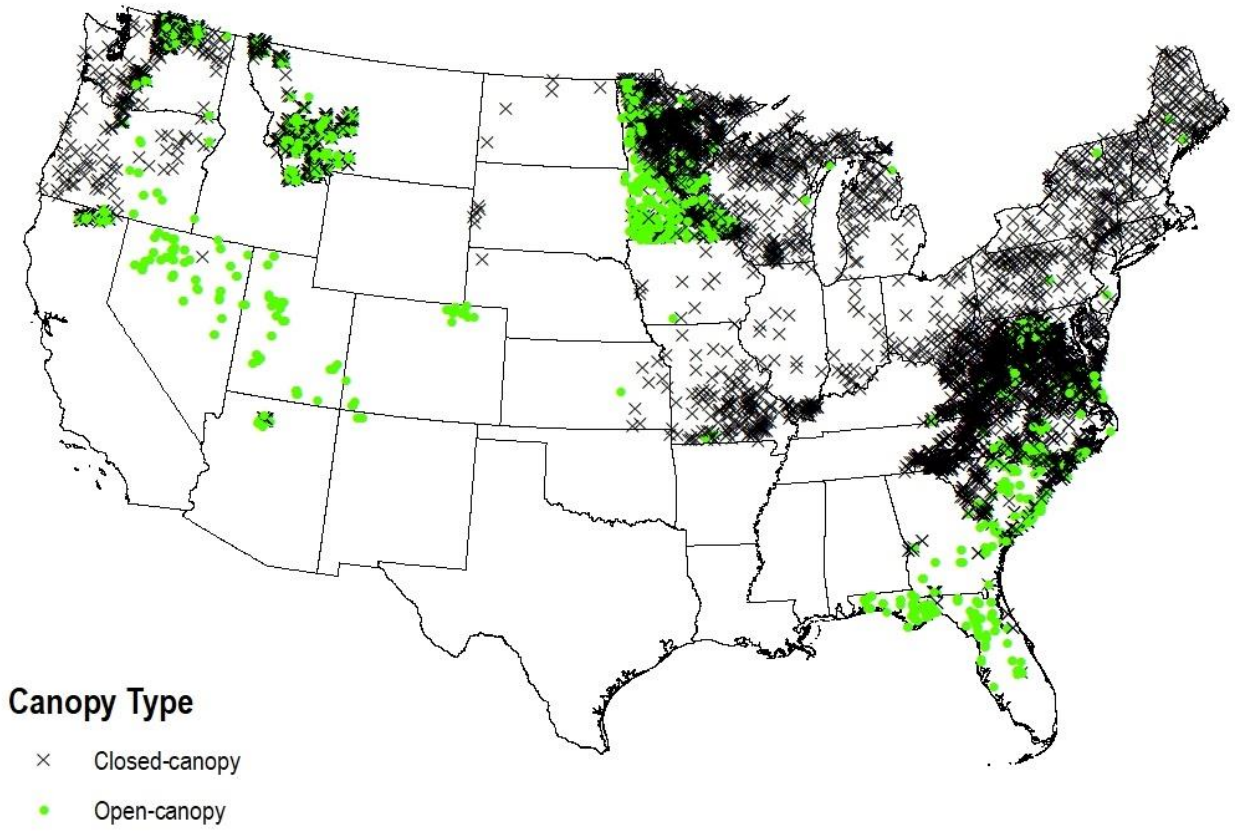


Figure 5B-13. Sites included in analysis by Simkin et al. (2016). Based on dataset available at https://datadryad.org/stash/dataset/doi:10.5061/dryad.7kn53

When sites were grouped as closed-canopy (forested) sites versus open-canopy (woodland, shrubland and grassland) sites, a statistical relationship was observed for variation in herbaceous species richness (number of herbaceous species) with variation in N deposition (and soil pH, followed by temperature and precipitation). Different quantitative relationships were observed for the two categories of sites. In open-canopy ecosystems, there was a positive relationship between herbaceous species richness and N deposition at the low end of the deposition range (sites with higher N deposition had more species), then a negative relationship with N deposition at higher deposition rates, with the deposition magnitude at the inflecting point varying with pH (Simkin et al., 2016). For example, in soils with pH of 4.5, the inflection point was 6.5 kg N $ha^{-1}yr^{-1}$ and in pH 7 soils, it was 8.8 kg N $ha^{-1}yr^{-1}$. In closed-canopy ecosystems, the variation in forest understory species richness with variation in N deposition was more strongly dependent on soil pH. At closed-canopy sites with low pH (4.5), a negative relationship was observed for species richness with N deposition above 11.6 kg N $ha^{-1}yr^{-1}$. At closed-canopy sites with soil pH greater than 8.0, no negative association of species richness with N deposition was observed across the full range of N deposition estimates, which extended up to about 20 kg N/ha-yr (Simkin et al., 2016).

The statistical models for the two categories of sites were then applied to the pH, temperature and precipitation for each site to predict N deposition values expected to be associated with a difference in species count from the predicted optimal for a site of that soil pH, temperature and precipitation. For the forested (closed-canopy sites), the inflection points above which a lower species richness would be expected ranged from 7.9 to 19.6 kg N $ha^{-1}yr^{-1}$, across pH, temperature and precipition of the assessed sites, with a mean of 13.4 kg N $ha^{-1}yr^{-1}$. Across the open-canopy sites, these N deposition inflection points ranged from 7.4 to 10.3 kg N $ha^{-1}yr^{-1}$, with a mean of 8.7 kg N $ha^{-1}yr^{-1}$ (Simkin et al., 2016).

Simkin et al. (2016) also performed regional gradient analyses for a set of sites for which the data were judged sufficient. This involved 44 gradients for a subset of 26 vegetation types that spanned a range in N deposition estimates the authors judged to be adequate. Of the 44 gradients, a negative association of species richness with N dep was observed at 16 (36.5%), a positive association at 8 (18%), and no association found for the remaining 20 (45%). Among the 8 gradients showing positive associations, most had N deposition estimates averaging at or below 3 kg N $ha^{-1}yr^{-1}$. Overall, a negative association of species richness with N deposition estimates was more common for gradients involving soil that was acidic, or had higher precipitation or warmer temperatures (Simkin et al., 2016).

In summary, the national-scale analysis of herbaceous species richness by Simkin et al. (2016) indicated that N deposition effects on forest closed-canopy species richness is highly dependent on soil pH (ISA, Appendix 6, section 6.3.3.2). At open-canopy sites (e.g., grasslands, shrublands, and woodlands) with low rates of N deposition (e.g., below 6.5 kg N $ha^{-1}yr^{-1}$ for soil pH of 4.5 and below 8.8 kg N $ha^{-1}yr^{-1}$ for soil pH of 7), relatively higher N deposition was generally associated with higher plant species richness (Simkin et al., 2016; ISA, Appendix 6, section 6.3.5). The open-canopy site-level N deposition above which a negative association was found for species richness with N deposition (higher deposition lower species count) ranged from 7.4 to 10.3 kg N ha^{-1} yr^{-1}, with an average of 8.7 kg N ha^{-1} yr^{-1}. At forested sites, relatively higher N deposition was associated with higher plant species richness for sites with soil pH of 4.5 and N deposition estimates below 11.6 kg N $ha^{-1}yr^{-1}$. With N deposition above this level the association was negative (higher deposition, lower species richness). At forested sites with the most basic soil (pH of 8.2), there was no value of N deposition that was negatively associated with species richness. At both the national and gradient analyses, few sites with N deposition estimates at or below 3 kg N $ha^{-1}yr^{-1}$ showed a negative relationship of species richness with N deposition (Simkin et al., 2016).

Study limitations with regard to interpretations specific for N deposition include that no other pollutants with potential to affect species richness (and which may covary in many places with N deposition), including sulfate and ozone, were considered. Further, the “modern” N

deposition estimates (1985-2011) were correlated with both shorter duration more recent estimates and with longer duration historical estimates, introducing uncertainty with regard to the particular deposition of interest with greatest influence on the results. This correlation coupled with the variation in magnitude of the deposition estimates for the various periods also contributes uncertainty regarding identification of what might be termed N deposition thresholds that might contribute to different types of relationships with species richness. Further, the study does not provide information on the species that are absent versus present, or their role in the community, across the varying species richness values. Additionally, site distribution varied across parts of the U.S. (as a result of combining species richness assessment surveys conducted in different contexts, for different purposes). For example, the most densely sampled closed canopy areas were in the southern Appalachians and Virginia, and Minnesota, areas of historically high and low deposition, respectively (Figure 5B-13). With regard to herb and shrub communities, there was appreciable representation in Minnesota and virtually no representation in Mediterranean California or the Great Plains. The potential influence of the relative distribution of sites across areas of greater versus lesser historical deposition is unclear.

5B.4 LICHEN COMMUNITY COMPOSITION

Lichens absorb N, S, and other elements from the air and from material deposited on their surfaces. Accordingly, lichens can be sensitive to air pollution and are frequently used as indicators of air quality, and associated deposition (2008 ISA, section 3.3.5.1), on forest ecosystems. Shifts in lichen community composition to greater presence of more N tolerant species have been associated with areas that have received high acidifying deposition and high concentrations of SO_2, N oxides and reduced N, such as the eastern U.S. (2008 ISA, section 3.2.2.3).

Research in the late 1970s-early 1980s reported inverse associations of lichen cover with atmospheric oxidants in the San Bernardino Mountains just outside Los Angeles, California. Studies in this region have reported a reduction in lichen species by about 50% since the early 1900s, with elevated HNO_3 identified as a contributor to lichen community declines in the Los Angeles basin dating back to the 1970s. Studies since the 2008 ISA indicate these communities have not yet recovered (ISA, Appendix 3, section 3.3). Surveys of urban and industrial areas in the 1970s and 80s (e.g., in urban areas of Great Britain) also identified SO_2 as a factor in lichen community declines observed lichen deaths (ISA, Appendix 3, section 3.2; Hutchinson et al.,

1996[16]). The relative influences of airborne versus deposited air pollutants in such impacts is unclear.

5B.4.1. Studies Investigating Direct Effects of Pollutants in Ambient Air

Sulfur oxides and oxides of N have been associated with effects on lichens (ISA, Appendix 3, section 3.2 and 3.3). In laboratory experiments involving daily HNO_3 exposures, with peaks near 50 ppb, over durations of 18 to 78 days, effects on lichen photosynthesis were reported, among other effects (ISA, Appendix 6, section 6.2.3.3; Riddell et al., 2012). Based on studies extending back to the 1980s, HNO_3 has been suspected to have had an important role in the dramatic declines of lichen communities that occurred in the Los Angeles basin (ISA, Appendix 3, section 3.4; Nash and Sigal, 1999; Riddell et al., 2008; Riddell et al., 2012). For example, lichen transplanted from clean air habitats to analogous habitats in the Los Angeles basin in 1985-86 were affected in a few weeks by mortality and appreciable accumulation of H^+ and NO_3^- (ISA, Appendix 3, section 3.4; Boonpragob et al., 1989).

Air monitoring data summarized in Chapter 2 indicate areas of the U.S. experiencing appreciably higher annual mean NO_2 concentrations in the 1980s compared to more recent years (Figure 2-22). For example the 95th percentile of U.S. sites ranged from just over 50 ppb to just over 60 ppb during the 1980s (Figure 2-22). During the 1980s and earlier, the Los Angeles metropolitan statistical area had some of the highest annual average NO_2 concentrations. For example, the annual average NO_2 concentration in Los Angeles was 0.078 ppm in 1979, 0.071 ppm in 1980, 0.058 ppm in 1985 and 0.057 ppm in 1989 (U.S. EPA, 1983, 1987, 1991). Concentrations of O_3 in Los Angeles were also quite high during this time (U.S. EPA, 1983, 1987, 1991); however, while O_3 impacts on plants are well established, research with lichens indicates a lesser sensitivity. This contributes to the evidence for NO_2, and particularly, HNO_3, as "the main agent of decline of lichen in the Los Angeles basin" (ISA, Appendix 3, p. 3-15).

Co-occurring elevations in SO_2 and ozone contribute uncertainty to identification of a threshold concentration of N oxides likely to elicit lichen community changes such as those that occurred in the Los Angeles basin. More recent studies indicate variation in eutrophic lichen abundance to be associated with variation in N deposition metrics (ISA, Appendix 6, section 6.2.3.3). The extent to which these associations are influenced by residual impacts of historic air quality is unclear.

[16] The publication by Hutchinson et al. (1996) cited in ISA, cites to Seaward (1987) as the support for its characterization; the characterization summarized here is also drawing on the specific details provided by Seaward (1987).

5B.4.2. Observational Studies Investigating Relationships with Atmospheric Deposition

Several recent studies have reported negative associations of lichen community composition/abundance and N deposition (and S deposition) metric values in areas of the Northwest, California and at some sites in the northeast (Table 5B-9; ISA, Appendix 6, section 6.5). For example, analyses of surveys in 1990s report species richness differences among sites in the Pacific NW to vary with estimates of N deposition (and N-$PM_{2.5}$) across sample sites ranging from approximately 8.2 to <1 kg N $ha^{-1}yr^{-1}$ and 10 to <1 kg dissolved inorganic N $ha^{-1}yr^{-1}$ (Geiser et al., 2010; Root et al., 2015, Appendix B, Table B.1). The study by Geiser et al. (2010) analyzed relationships between lichen community composition and several N deposition metrics at sites in Western Oregon and Washington forests. At other sites in the western U.S., Root et al. (2015) analyzed relationships between lichen community/abundance metrics and lichen N concentrations and N deposition estimates extrapolated from lichen N concentrations. Statistical modeling was used to identify N deposition estimates associated with a change in lichen community/abundance metric(s) for sites in 2 ecoregions. Both papers utilized a linear regression approach. Geiser et al. (2010) used the regression to relate community composition to an "air score," while Root et al. (2015) used it to relate a community-based index to air concentrations of nitrogen in fine PM, which was then related to N deposition.

There are several limitations associated with use of these studies' findings for purposes of interpreting potential risk to lichens of recent N deposition. For example, the estimates of deposition utilized different methods than the current commonly accepted methods. The potential role of other unaccounted environmental factors (including ozone, SO_2 and S deposition) has not been addressed in these observational/gradient, uncontrolled studies, and there is a scarcity of controlled N addition experiments that might augment conclusions. The significance of findings of the western studies is unclear for other areas of the U.S., and there is uncertainty concerning the independence of any effect of the deposition levels analyzed from residual effects of past N deposition. Further, the extent to which these observations reflect communities still exhibiting impacts of much higher pollution of the 1970s-80s is unknown. Although some studies have investigated historical impacts, there remain uncertainties as to the extent to which impacts on lichen communities noted in recent studies reflect recent N deposition. And there are few controlled N addition experiments that might augment or inform interpretation of the findings of observational/gradient studies. Other studies in Europe and Canada have not reported such associations with relatively large N deposition gradients.

Table 5B-9. Lichen endpoints and associated deposition estimates.

Description	Deposition Estimates	Findings
Cleavitt et al. (2011) analyzed 4 plots distributed across a gradient in estimated S deposition in Acadia National Park, ME	12 to 18 kg S/ha-yr	Rather than relate deposition to lichen distribution, this study reported that throughfall chemistry influenced bark pH and that influenced the suitability of tree boles as habitat for lichen. Epiphytic lichen species richness and presence of pollution-sensitive epiphytes were greater on red maple trees, which have a higher pH in the bark relative to red spruce trees.
Cleavitt et al. (2015) analyzed 24 sites in 4 Class I areas in Northeastern U.S. (Lye Brook Wilderness, VT, Great Gulf and Presidential Range-Dry River Wiildernesses, NH, and Acadia National Park, ME); assessed multiple metrics for lichen status associations with concurrent (2-yr ave) and cumulative (2000-13) S and N deposition estimates. Cumulative and 2-yr average recent N deposition were tightly correlated (r2=0.90 p , 0.0001);cumulative and recent S deposition were not correlated. Aerosol NO_3^- declined from ~0.7-0.9 to ~0.25-0.5 ug/m^3 across 14-yr period.	Total S deposition of ~6-15 kg S/ha-yr across the 4 areas in 2000; with subsequent reductions to ~3-6 kg S/ha-yr by 2013. Total N deposition of ~4-15 kg N/ha-yr across 4 areas In 2000; with subsequent reductions to ~3-8 kg N/ha-yr (Cleavitt et al., 2015, Figure 4).	Negative associations of lichen species richness, abundance of N-sensitive species, and poorer thallus condition with annual mean and cumulative N deposition. Cumulative dry deposition of S yielded best fit to decreases in thallus condition, poorer community-based S Index values, and absence of many S-sensitive species, indicating stronger role for legacy of historical deposition than recent deposition patterns. "Lichen metrics were generally better correlated with cumulative deposition than annual deposition." "In our study, dry S deposition related more closely to patterns in lichen metrics than total or wet S deposition. Dry deposition of S may be more harmful to lichens, both because it has the potential to become highly concentrated when the thallus is rehydrated, and because it largely originates from SO_2, which has a long history of toxicity to lichens."
Geiser et al. (2010) analyzed data at sites in Western OR and WA forests, calculating different N metrics (total, dry and wet N deposition; wet $NO_3^- + NH_4^+$ deposition; and $PM_{2.5}$-N, dry N deposition for specified breakpoint in "air scores." Statistical modeling of FIA plot air scores based on aspects of lichen community composition and lichen N/S concentrations (assessed 1994-2002) for data subset, considering elevation, precip (1961-90), hardwood basal area (Geiser and Neitlich, 2007). Then model used to predict scores for remaining plots. Range of scores divided into six bins from "best" (lowest bin) to "worst" (highest bin).	Average 1990-99 N deposition estimated from CMAQ modeling (0.8 – 8.2 kg/ha-yr across all sites); NADP wet deposition and IMPROVE particulate N for 1994-2002	For breakpoint between 3rd and 4th air scores,total N deposition ranged from about 3 to 9 kg N/ha-yr The score equal to the breakpoint between the 3rd and 4th bins ("fair" and "degraded") was associated with 33-43% fewer oligotrophic species and 3 to 4 fold more eutrophic species than scores in the "best" bin. Per Geiser & Neitlich 2007 for same areas: "Ozone is potentially adversely affecting Pacific Northwest lichens." "Ambient [air] concentrations of NO_X often correlate with SO_2, making it difficult to separate SO_2 effects on lichen communities from NO_X effects."
Root et al. (2015) analyzed data for sites in WA, northern ID, NW MT, OR and far NE CA for relationship between lichen community metrics (assessed 1993-2011) and lichen N concentrations (samples 1993-2001) and N deposition estimated from lichen N. Created lichen index relating lichen N to species frequency (excluding uncommon species and species with "ambiguous relationships").	Inorganic N deposition extrapolated from lichen N concentrations, estimated to range from 0.174 to 9.49 kg N/ha-yr across sampling plots	Based on a judgment that "[l]ichen communities did not appear to be strongly impacted by N concentration below 0.378 ug N/m^3/year" which was the lowest N-$PM_{2.5}$ concentration near "known N pollution sources," and the associated lichen N concentration estimated by linear regression, the throughfall N deposition was estimated to be 2.5 kg Ha-yr. Throughfall N deposition estimated from the lichen index value estimated for the chosen N-$PM_{2.5}$ and its estimated relationship with throughfall N, was estimated to be 1.5 kg N/ha-yr.

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Attachments

Attachment 1
Species by Plant Functional Group
Drawn from Dietze and Moorcroft (2011) "Tree mortality in the eastern and central United States: patterns and drivers"

Plant Functional Group	Genus Species	Common Name	Genus Species	Common Name
Early Successional Hardwood - Large positive influence of SO_4^{2-} deposition on mortality - negative influence of NO_3^- deposition on mortality	*Ailanthus altissima*	ailanthus	*Populus alba*	silver poplar
	Albizia julibrissin	Mimosa	*Populus balsamifera*	balsam poplar
	Alnus	alder	*Populus deltoides*	eastern cottonwood
	Betula	Birch	*Populus deltoides sub monilifera*	plains cottonwood
	Betula alleghaniensis	yellow birch	*Populus grandidentata*	bigtooth aspen
	Betula lenta	sweet birch	*Populus tremuloides*	quaking aspen
	Betula nigra	river birch	*Prosopis pubescens*	screwbean mesquite
	Betula papyrifera	paper birch	*Prunus*	cherry
	Betula populifolia	gray birch	*Prunus americana*	American plum
	Bursera simaruba	gumbo limbo	*Prunus aviumPRAV*	sweet cherry
	Catalpa	catalpa	*Prunus nigra*	Canada plum
	Catalpa bignoniodes	southern catalpa	*Prunus pensylvanica*	pin cherry
	Catalpa speciosa	northern catalpa	*Prunus serotina*	black cherry
	Elaeagnus angustifolia	Russian-olive	*Prunus virginiana*	chokecherry
	Ficus aurea	Florida strangler fig	*Robinia pseudoacacia*	black locust
	Gleditsia triacanthos	honeylocust	*Salix*	willow
	Gymnocladus dioicus	Kentucky coffeetree	*Salix alba*	white willow
	Larix laricina	tamarack	*Salix bebbiana*	Bebb willow
	Larix spp	Larch spp	*Salix caroliniana*	costal plain willow
	Liquidambar styraciflua	sweetgum	*Salix nigra*	black willow
	Maclura pomifera	Osage-orange	*Salix sepulcralis*	weeping willow
	Melia azedarach	Chinaberrytree	*Sideroxylon lanuginosum ssp. lanuginosum*	gum bully
	Paulownia tomentosa	paulownia	*Vernicia fordii*	tung-oil-tree
	Populus	poplar		
Evergreen Hardwoods - Large positive influence of SO_4^{2-} deposition on mortality - negative influence of NO_3^- deposition on mortality	*Avicennia germinans*	Black-mangrove	*Magnolia grandifolia*	southern magnolia
	Casuarina lepidophloia	belah	*Magnolia virginiana*	sweetbay
	Cinnamomum camphora	camphor tree	*Melaleuca quinquenervia*	melaleuca
	Conocarpus erectus	buttonwood mangrove	*Persea borbonia*	redbay
	Eucalyptus	eucalyptus	*Quercus margarettiae*	dwarf live oak
	Eucalyptus grandis	grand eucalyptus	*Quercus virginiana*	live oak
	Gordonia lasianthus	loblolly-bay	*Rhizophora mangle*	American mangrove
	Ilex opaca	American holly	*Umbellularia californica*	California laurel
	Laguncularia racemosa	white -mangrove		
Hydric - Large positive influence of SO_4^{2-}	*Carya aquatica*	water hickory	*Planera aquatica*	water elm
	Citrus	Citrus	*Populus heterophylla*	swamp cottonwood
	Eugenia rhombea	red stopper	*Quercus lyrata*	overcup oak

Plant Functional Group	Genus Species	Common Name	Genus Species	Common Name
deposition on mortality - negative influence of NO_3^- deposition on mortality	*Gleditsia aquatica*	waterlocust	*Sabal palmetto*	cabbage palmetto
	Metopium toxiferum	Florida poisontree	*Salix amygdaloides*	peachleaf willow
	NULL	palm, other	*Taxodium ascendens*	pondcypress
	Nyssa aquatica	water tupelo	*Taxodium distichum*	baldcypress
	Nyssa biflora	swamp tupelo	*Thrinax morrisii*	key thatch palm
	Nyssa ogeche	Ogechee tupelo		
Late Successional Conifer - negative influence of NO_3^- deposition on mortality - weakly negative influence of SO_4^{2-} deposition on mortality	*Abies balsamea*	Balsam fir	*Juniperus virginiana var silicicola*	Southern redcedar
	Chamaecyparis thyoides	Atlantic white-cedar	*Thuja occidentalis*	northern white-cedar
	Juniperus	juniper	*Tsuga*	hemlock
	Juniperus ashei	Ashe juniper	*Tsuga canadensis*	eastern hemlock
	Juniperus scopulorum	Rocky Mountain juniper	*Tsuga caroliniana*	Carolina hemlock
	Juniperus virginiana	eastern redcedar		
Late Successional Hardwood - Large positive influence of SO_4^{2-} deposition on mortality - negative influence of NO_3^- deposition on mortality	*Acer*	Maple	*Carpinus caroliniana*	hornbeam
	Acer barbatum	Florida maple	*Castanea dentata*	American chestnut
	Acer leucoderme	chalk maple	*Cornus florida*	Flowering dogwood
	Acer negundo	boxelder	*Diospyros*	persimmon
	Acer nigrum	black maple	*Diospyros virginiana*	common persimmon
	Acer pensylvanicum	striped maple	*Fagus grandifolia*	beech
	Acer platanoides	Norway maple	*Halesia*	silverbell
	Acer rubrum	red maple	*Halesia carolina*	Carolina silverbell
	Acer saccharinum	silver maple	*Halesia parviflora*	two-wing silverbel
	Acer saccharum	sugar maple	*Oxydendrum arboreum*	sourwood
	Acer spicatum	mountain maple	*Platanus*	sycamore
	Aesculus	buckeye	*Sapindus saponaria var drummondii*	western soapberry
	Aesculus flava	yellow buckeye	*Tilia*	basswood
	Aesculus glabra	Ohio buckeye	*Tilia americana*	american basswood
	Aesculus glabra var arguta	Texas buckeye	*Tilia americana var caroliniana*	Carolina basswood
	Alnus glutinosa	European alder	*Tilia americana var. heterophylla*	American basswood
Midsuccessional conifer - negative influence of NO_3^- deposition on mortality	*Abies*	fir spp.	*Picea glauca*	white spruce
	Abies concolor	white fir	*Picea mariana*	black spruce
	Abies fraseri	Fraser fir	*Picea pungens*	Blue spruce
	Picea	spruce	*Picea rubens*	red spruce
	Picea abies	Norway Spruce	*Pseudotsuga menziesii*	Douglas-fir
Northern Midsuccessional Hardwood - positive influence of NO_3^- deposition on mortality	*Amelanchier*	serviceberry	*Morus alba*	white mulberry
	Amelanchier arborea	Downy serviceberry	*Morus rubra*	red mulberry
	Carya	hickory	*Ostrya virginiana*	eastern hophornbeam
	Carya cordiformis	bitternut hickory	*Quercus alba*	white oak
	Carya ovalis	red hickory	*Quercus bicolor*	swamp white oak
	Carya ovata	shagbark hickory	*Quercus ellipsoidalis*	northern pin oak
	Celtis laevigata var reticulata	netleaf hackberry	*Quercus ilicifolia*	scrub oak
	Celtis occidentalis	hackberry	*Quercus macrocarpa*	bur oak

Plant Functional Group	Genus Species	Common Name	Genus Species	Common Name
	Cladrastis kentukea	yellowwood	*Quercus palustris*	pin oak
	Crataegus	hawthorn	*Quercus prinoides*	swarf chinakapin oak
	Crataegus crus-galli	cockspur hawthorn	*Quercus rubra*	northern red oak
	Crataegus mollis	downy hawthorn	*Quercus velutina*	black oak
	Fraxinus americana	white ash	*Sassafras albidum*	sassafras
	Fraxinus nigra	black ash	*Sorbus americana*	American mountain-ash
	Fraxinus pennsylvanica	green ash	*Sorbus aucuparia*	European mountain-ash
	Fraxinus profunda	pumpkin ash	*Ulmus*	elm
	Juglans	walnut	*Ulmus americana*	American elm
	Juglans cinera	butternut	*Ulmus pumila*	Siberian elm
	Juglans nigra	black walnut	*Ulmus rubra*	slippery elm
	Malus	apple spp.	*Ulmus thomasii*	rock elm
	Malus coronaria	sweet crabapple	*Unknown*	Unknown dead hardwood
	Malus ioensis	prairie crabapple		
Northern Pine - Large positive influence of SO_4^{2-} on mortality - negative influence of NO_3^- on mortality	*Pinus banksiana*	jack pine	*Pinus rigida*	pitch pine
	Pinus nigra	Austrian pine	*Pinus strobus*	white pine
	Pinus ponderosa	Ponderosa pine	*Pinus sylvestris*	Scotch pine
	Pinus resinosa	red pine		
Southern Midsuccessional Hardwood - Large positive influence of SO_4^{2-} deposition on mortality - negative influence of NO_3^- deposition on mortality	*Asimina triloba*	pawpaw	*Morus*	mulberry
	Carya alba	mockernut hickory	*Nyssa sylvatica*	blackgum
	Carya carolinae-septentrionalis	southern shagbark hickory	*Quercus*	oak spp. -- Deciduous
	Carya glabra	pignut hickory	*Quercus buckleyi*	Buckley oak
	Carya illinoinensis	pecan	*Quercus coccinia*	scarlet oak
	Carya laciniosa	shellbark hickory	*Quercus falcata*	southern red oak
	Carya myristiciformis	nutmeg hickory	*Quercus imbricaria*	shingle oak
	Carya pallida	sand hickory	*Quercus incana*	bluejack oak
	Carya texana	black hickory	*Quercus laevis*	turkey oak
	Castanea mollissima	chinese chestnut	*Quercus laurifolia*	laurel oak
	Castanea pumila	Chinkapin	*Quercus margarettiae*	runner oak
	Castanea pumila var ozarkensis	Ozark chinkapin	*Quercus marilandica*	blackjack oak
	Celtis	hackberry	*Quercus michauxii*	swamp chestnut oak
	Celtis laevigata	sugarberry	*Quercus muehlenbergii*	chinkapin oak
	Cercis canadensis	eastern redbud	*Quercus nigra*	water oak
	Cotinus obovatus	smoketree	*Quercus oglethorpensis*	Oglethorpe oak
	Fraxinus	ash	*Quercus pagoda*	cherrybark oak
	Fraxinus caroliniana	Carolina ash	*Quercus phellos*	willow oak
	Fraxinus quadrangulata	blue ash	*Quercus prinus*	chestnut oak
	Liriodendron tulipifera	yellow-poplar	*Quercus shumardii*	Shumard's oak
	Magnolia	magnolia	*Quercus similis*	Delta post oak
	Magnolia acuminata	cucumbertree	*Quercus sinuata var sinuata*	Durand oak
	Magnolia fraseri	mountain magnolia	*Quercus stellata*	post oak

Plant Functional Group	Genus Species	Common Name	Genus Species	Common Name
	Magnolia macrophylla	bigleaf magnolia	*Triadica sebifera*	Chinese tallowtree
	Magnolia tripetala	umbrella magnolia	*Ulmus alata*	winged elm
	Malus angustifolia	southern crabapple	*Ulmus crassifolia*	cedar elm
			Ulmus serotina	September elm
Southern Pine - Large positive influence of SO_4^{2-} deposition on mortality - negative influence of NO_3^- deposition on mortality	*Pinus clausa*	Sand pine		
	Pinus echinata	shortleaf pine		
	Pinus elliottii	slash pine		
	Pinus glabra	spruce pine		
	Pinus palustris	longleaf pine		
	Pinus pungens	Table Mountain pine		
	Pinus serotina	pond pine		
	Pinus taeda	loblolly pine		
	Pinus virginiana	Virginia pine		

Attachment 2A
Species-specific Sample Distribution across Ecoregions for Species with Statistically Significant Associations of Growth with N/S from Horn et al. (2018) Supplemental Information Dataset

Key:

NA_L3 = North American Ecoregion, code for level 3

US_L3NAME = Name of Ecoregion at level 3

See: https://www.epa.gov/eco-research/ecoregions

Median = Tree-specific median S and/or N deposition for the species samples

Assoc = U= unimodal, ↑=positive, ↓=negative

N/S = elation coefficient for N and S deposition values for the species samples

Count = number of species' tree samples assessed in all plots in that ecoregion

% = percent of species' tree samples in that ecoregion

NA_L3 CODE	US_L3NAME	**boxelder** Median S=6 Assoc S-↓ N/S = 0.14		**red maple** Median N=9, S=7 Assoc N-↑, S-↓ N/S = 0.6		**silver maple** Median N=12,S=8 Assoc N-↑, S-↓ N/S = 0.27		**yellow birch** Median N=7 Assoc N-U N/S = 0.7		**sweet birch** Median S=12 Assoc S-↓ N/S = 0.58		**paper birch** Median S=4 Assoc S-↓ N/S = 0.42	
		count	**%**	**count**	**%**	**count**	**%**	**count**	**%**	**count**	**%**	**count**	**%**
5.2.1	Northern Lakes and Forests	85	1.4%	23972	23.6%	324	7.1%	3282	23.9%			9247	50.1%
5.2.2	Northern Minnesota Wetlands	53	0.9%	93	0.1%			1	0.0%			547	3.0%
5.3.1	Northeastern Highlands	17	0.3%	13245	13.1%	27	0.6%	6357	46.3%	1322	14.8%	4824	26.1%
5.3.3	North Central Appalachians			4883	4.8%			363	2.6%	1299	14.6%	93	0.5%
6.2.3	Northern Rockies											129	0.7%
6.2.4	Canadian Rockies											6	0.0%
6.2.5	North Cascades											1	0.0%
6.2.7	Cascades												
6.2.8	Eastern Cascades Slopes and Foothills												
6.2.9	Blue Mountains											4	0.0%
6.2.10	Middle Rockies											16	0.1%
6.2.11	Klamath Mountains												
6.2.12	Sierra Nevada												
6.2.13	Wasatch and Uinta Mountains												
6.2.14	Southern Rockies												
6.2.15	Idaho Batholith											6	0.0%
7.1.7	Puget Lowland											18	0.1%
7.1.8	Coast Range												
7.1.9	Willamette Valley												
8.1.1	Eastern Great Lakes Lowlands	86	1.4%	1618	1.6%	293	6.4%	204	1.5%	49	0.6%	57	0.3%
8.1.3	Northern Allegheny Plateau	8	0.1%	3565	3.5%	9	0.2%	394	2.9%	543	6.1%	82	0.4%
8.1.4	North Central Hardwood Forests	594	9.8%	4062	4.0%	448	9.8%	418	3.0%			978	5.3%

NA_L3 CODE	US_L3NAME	**boxelder** Median S=6 Assoc S-↓ N/S = 0.14		**red maple** Median N=9, S=7 Assoc N-↑, S-↓ N/S = 0.6		**silver maple** Median N=12,S=8 Assoc N-↑, S-↓ N/S = 0.27		**yellow birch** Median N=7 Assoc N-U N/S = 0.7		**sweet birch** Median S=12 Assoc S-↓ N/S = 0.58		**paper birch** Median S=4 Assoc S-↓ N/S = 0.42	
		count	**%**	**count**	**%**	**count**	**%**	**count**	**%**	**count**	**%**	**count**	**%**
8.1.5	Driftless Area	934	15.4%	631	0.6%	387	8.5%	31	0.2%			651	3.5%
8.1.6	Southern Michigan/Northern Indiana Drift Plains	106	1.7%	1463	1.4%	516	11.3%	36	0.3%			29	0.2%
8.1.7	Northeastern Coastal Zone	6	0.1%	4309	4.2%	18	0.4%	256	1.9%	975	10.9%	134	0.7%
8.1.8	Acadian Plains and Hills			5025	5.0%	9	0.2%	1236	9.0%			1437	7.8%
8.1.10	Erie Drift Plain	8	0.1%	1811	1.8%	83	1.8%	112	0.8%	6	0.1%		
8.2.1	Southeastern Wisconsin Till Plains	338	5.6%	156	0.2%	124	2.7%	55	0.4%			55	0.3%
8.2.2	Huron/Erie Lake Plains	82	1.4%	1123	1.1%	195	4.3%	6	0.0%			123	0.7%
8.2.3	Central Corn Belt Plains	76	1.3%	13	0.0%	101	2.2%						
8.2.4	Eastern Corn Belt Plains	202	3.3%	333	0.3%	197	4.3%	1	0.0%				
8.3.1	Northern Piedmont	71	1.2%	566	0.6%	38	0.8%	4	0.0%	109	1.2%		
8.3.2	Interior River Valleys and Hills	296	4.9%	423	0.4%	625	13.7%						
8.3.3	Interior Plateau	469	7.7%	1061	1.0%	82	1.8%						
8.3.4	Piedmont	135	2.2%	3119	3.1%					26	0.3%		
8.3.5	Southeastern Plains	154	2.5%	4363	4.3%	11	0.2%						
8.3.6	Mississippi Valley Loess Plains	192	3.2%	195	0.2%	35	0.8%						
8.3.7	South Central Plains	89	1.5%	742	0.7%	10	0.2%						
8.3.8	East Central Texas Plains	5	0.1%	4	0.0%								
8.4.1	Ridge and Valley	100	1.6%	4942	4.9%	18	0.4%	166	1.2%	1866	21.0%	12	0.1%
8.4.2	Central Appalachians	17	0.3%	4912	4.8%	3	0.1%	495	3.6%	1170	13.1%		
8.4.3	Western Allegheny Plateau	193	3.2%	3926	3.9%	103	2.3%	17	0.1%	230	2.6%		
8.4.4	Blue Ridge	18	0.3%	3707	3.7%	3	0.1%	283	2.1%	1280	14.4%		
8.4.5	Ozark Highlands	105	1.7%	185	0.2%	47	1.0%						

NA_L3 CODE	US_L3NAME	**boxelder** Median S=6 Assoc S-↓ N/S = 0.14		**red maple** Median N=9, S=7 Assoc N-↑, S-↓ N/S = 0.6		**silver maple** Median N=12,S=8 Assoc N-↑, S-↓ N/S = 0.27		**yellow birch** Median N=7 Assoc N-U N/S = 0.7		**sweet birch** Median S=12 Assoc S-↓ N/S = 0.58		**paper birch** Median S=4 Assoc S-↓ N/S = 0.42	
		count	**%**	**count**	**%**	**count**	**%**	**count**	**%**	**count**	**%**	**count**	**%**
8.4.6	Boston Mountains			174	0.2%								
8.4.7	Arkansas Valley	28	0.5%	56	0.1%	43	0.9%						
8.4.8	Ouachita Mountains	2	0.0%	156	0.2%	3	0.1%						
8.4.9	Southwestern Appalachians	25	0.4%	1401	1.4%			1	0.0%	24	0.3%		
8.5.1	Middle Atlantic Coastal Plain	19	0.3%	2982	2.9%	15	0.3%						
8.5.2	Mississippi Alluvial Plain	396	6.5%	471	0.5%	83	1.8%						
8.5.3	Southern Coastal Plain	4	0.1%	1425	1.4%								
8.5.4	Atlantic Coastal Pine Barrens	1	0.0%	256	0.3%					6	0.1%		
9.2.1	Northern Glaciated Plains	140	2.3%									4	0.0%
9.2.2	Lake Agassiz Plain	200	3.3%	3	0.0%							6	0.0%
9.2.3	Western Corn Belt Plains	555	9.1%	21	0.0%	420	9.2%	3	0.0%			6	0.0%
9.2.4	Central Irregular Plains	157	2.6%	0	0.0%	273	6.0%						
9.3.1	Northwestern Glaciated Plains	17	0.3%			2	0.0%						
9.3.3	Northwestern Great Plains	13	0.2%										
9.3.4	Nebraska Sand Hills	1	0.0%										
9.4.1	High Plains	3	0.0%										
9.4.2	Central Great Plains	48	0.8%			14	0.3%						
9.4.3	Southwestern Tablelands	4	0.1%										
9.4.4	Flint Hills	6	0.1%			1	0.0%						
9.4.5	Cross Timbers	3	0.0%			1	0.0%						
9.4.6	Edwards Plateau												
9.4.7	Texas Blackland Prairies												

NA_L3 CODE	US_L3NAME	**boxelder** Median S=6 Assoc S-↓ N/S = 0.14		**red maple** Median N=9, S=7 Assoc N-↑, S-↓ N/S = 0.6		**silver maple** Median N=12,S=8 Assoc N-↑, S-↓ N/S = 0.27		**yellow birch** Median N=7 Assoc N-U N/S = 0.7		**sweet birch** Median S=12 Assoc S-↓ N/S = 0.58		**paper birch** Median S=4 Assoc S-↓ N/S = 0.42	
		count	**%**	**count**	**%**	**count**	**%**	**count**	**%**	**count**	**%**	**count**	**%**
9.5.1	Western Gulf Coastal Plain			8	0.0%								
9.6.1	Southern Texas Plains												
10.1.2	Columbia Plateau												
10.1.3	Northern Basin and Range												
10.1.4	Wyoming Basin												
10.1.5	Central Basin and Range												
10.1.6	Colorado Plateaus												
10.1.7	Arizona/New Mexico Plateau												
10.1.8	Snake River Plain												
10.2.1	Mojave Basin and Range												
10.2.2	Sonoran Basin and Range												
10.2.10	Chihuahuan Deserts												
11.1.1	Southern and Central California Chaparral and Oak Woodlands												
11.1.2	Central California Valley	8	0.1%										
11.1.3	Southern California Mountains												
12.1.1	Madrean Archipelago												
13.1.1	Arizona/New Mexico Mountains	1	0.0%										
15.4.1	Southern Florida Coastal Plain			34	0.0%								
	Total Tree Counts	6070		101434		4561		13721		8905		18465	

NA_L3 CODE	US_L3NAME	**American hornbeam** Median S=7 Assoc S-↓ N/S = 0.23		**black hickory** Median N=10 Assoc N-↑ N/S = 0.17		**hackberry** Median N=11 Assoc N-U N/S = 0.17		**American beech** Median N=8,S=7 Assoc N-U,S-↓ N/S = 0.76		**white ash** Median N=10 Assoc N-↑ N/S = 0.54		**green ash** Median N=10, =6 Assoc N-U, S-↓ N/S = 0.45	
		count	**%**	**count**	**%**	**count**	**%**	**count**	**%**	**count**	**%**	**count**	**%**
5.2.1	Northern Lakes and Forests	9	0.4%					1378	6.6%	1273	7.4%	1807	11.6%
5.2.2	Northern Minnesota Wetlands					1	0.0%					200	1.3%
5.3.1	Northeastern Highlands	9	0.4%					8502	40.7%	2438	14.1%	39	0.3%
5.3.3	North Central Appalachians	45	2.1%					1520	7.3%	455	2.6%		
6.2.3	Northern Rockies												
6.2.4	Canadian Rockies												
6.2.5	North Cascades												
6.2.7	Cascades												
6.2.8	Eastern Cascades Slopes and Foothills												
6.2.9	Blue Mountains												
6.2.10	Middle Rockies					1	0.0%					24	0.2%
6.2.11	Klamath Mountains												
6.2.12	Sierra Nevada												
6.2.13	Wasatch and Uinta Mountains												
6.2.14	Southern Rockies												
6.2.15	Idaho Batholith												
7.1.7	Puget Lowland												
7.1.8	Coast Range												
7.1.9	Willamette Valley												
8.1.1	Eastern Great Lakes Lowlands	7	0.3%			3	0.1%	282	1.3%	652	3.8%	479	3.1%
8.1.3	Northern Allegheny Plateau	28	1.3%					1192	5.7%	1721	10.0%	76	0.5%

NA_L3 CODE	US_L3NAME	American hornbeam Median S=7 Assoc S-↓ N/S = 0.23		black hickory Median N=10 Assoc N-↑ N/S = 0.17		hackberry Median N=11 Assoc N-U N/S = 0.17		American beech Median N=8,S=7 Assoc N-U,S-↓ N/S = 0.76		white ash Median N=10 Assoc N-↑ N/S = 0.54		green ash Median N=10, =6 Assoc N-U, S-↓ N/S = 0.45	
		count	%	count	%	count	%	count	%	count	%	count	%
8.1.4	North Central Hardwood Forests	4	0.2%			46	0.9%	155	0.7%	595	3.4%	1429	9.2%
8.1.5	Driftless Area	2	0.1%			235	4.8%	0	0.0%	354	2.1%	187	1.2%
8.1.6	Southern Michigan/Northern Indiana Drift Plains	5	0.2%			35	0.7%	143	0.7%	265	1.5%	913	5.9%
8.1.7	Northeastern Coastal Zone	2	0.1%			5	0.1%	327	1.6%	399	2.3%	58	0.4%
8.1.8	Acadian Plains and Hills							1470	7.0%	758	4.4%	31	0.2%
8.1.10	Erie Drift Plain	10	0.5%			3	0.1%	290	1.4%	465	2.7%	128	0.8%
8.2.1	Southeastern Wisconsin Till Plains					23	0.5%	43	0.2%	177	1.0%	675	4.3%
8.2.2	Huron/Erie Lake Plains	1	0.0%			13	0.3%	29	0.1%	84	0.5%	667	4.3%
8.2.3	Central Corn Belt Plains	3	0.1%			75	1.5%			45	0.3%	102	0.7%
8.2.4	Eastern Corn Belt Plains	9	0.4%			271	5.5%	110	0.5%	708	4.1%	286	1.8%
8.3.1	Northern Piedmont	8	0.4%			29	0.6%	76	0.4%	256	1.5%	50	0.3%
8.3.2	Interior River Valleys and Hills	13	0.6%	79	2.0%	591	12.1%	120	0.6%	477	2.8%	547	3.5%
8.3.3	Interior Plateau	72	3.4%	24	0.6%	1031	21.0%	735	3.5%	1408	8.2%	714	4.6%
8.3.4	Piedmont	252	11.8%			65	1.3%	521	2.5%	291	1.7%	481	3.1%
8.3.5	Southeastern Plains	595	27.8%	7	0.2%	44	0.9%	609	2.9%	110	0.6%	1054	6.8%
8.3.6	Mississippi Valley Loess Plains	175	8.2%	26	0.7%	8	0.2%	102	0.5%	82	0.5%	254	1.6%
8.3.7	South Central Plains	469	21.9%	190	4.8%	9	0.2%	152	0.7%	140	0.8%	561	3.6%
8.3.8	East Central Texas Plains	2	0.1%	87	2.2%	2	0.0%			36	0.2%	168	1.1%
8.4.1	Ridge and Valley	19	0.9%			138	2.8%	434	2.1%	909	5.3%	196	1.3%
8.4.2	Central Appalachians	36	1.7%			5	0.1%	1403	6.7%	408	2.4%	44	0.3%
8.4.3	Western Allegheny Plateau	30	1.4%			70	1.4%	678	3.2%	1138	6.6%	106	0.7%
8.4.4	Blue Ridge	15	0.7%			5	0.1%	294	1.4%	317	1.8%	43	0.3%

NA_L3 CODE	US_L3NAME	**American hornbeam** Median S=7 Assoc S-↓ N/S = 0.23		**black hickory** Median N=10 Assoc N-↑ N/S = 0.17		**hackberry** Median N=11 Assoc N-U N/S = 0.17		**American beech** Median N=8,S=7 Assoc N-U,S-↓ N/S = 0.76		**white ash** Median N=10 Assoc N-↑ N/S = 0.54		**green ash** Median N=10, =6 Assoc N-U, S-↓ N/S = 0.45	
		count	**%**	**count**	**%**	**count**	**%**	**count**	**%**	**count**	**%**	**count**	**%**
8.4.5	Ozark Highlands	5	0.2%	1863	46.6%	262	5.3%			558	3.2%	155	1.0%
8.4.6	Boston Mountains	4	0.2%	681	17.0%	20	0.4%	55	0.3%	79	0.5%	18	0.1%
8.4.7	Arkansas Valley	10	0.5%	576	14.4%	32	0.7%			65	0.4%	137	0.9%
8.4.8	Ouachita Mountains	24	1.1%	385	9.6%	2	0.0%	8	0.0%	23	0.1%	80	0.5%
8.4.9	Southwestern Appalachians	27	1.3%	1	0.0%	19	0.4%	152	0.7%	214	1.2%	119	0.8%
8.5.1	Middle Atlantic Coastal Plain	101	4.7%			21	0.4%	89	0.4%	34	0.2%	369	2.4%
8.5.2	Mississippi Alluvial Plain	28	1.3%	21	0.5%	55	1.1%	9	0.0%	9	0.1%	717	4.6%
8.5.3	Southern Coastal Plain	108	5.1%			9	0.2%	2	0.0%	6	0.0%	440	2.8%
8.5.4	Atlantic Coastal Pine Barrens	2	0.1%			0	0.0%	14	0.1%	7	0.0%		
9.2.1	Northern Glaciated Plains					14	0.3%					337	2.2%
9.2.2	Lake Agassiz Plain											254	1.6%
9.2.3	Western Corn Belt Plains					571	11.6%			82	0.5%	416	2.7%
9.2.4	Central Irregular Plains			45	1.1%	779	15.9%			216	1.3%	354	2.3%
9.3.1	Northwestern Glaciated Plains					14	0.3%					81	0.5%
9.3.3	Northwestern Great Plains					3	0.1%					360	2.3%
9.3.4	Nebraska Sand Hills					6	0.1%					25	0.2%
9.4.1	High Plains					2	0.0%					18	0.1%
9.4.2	Central Great Plains					235	4.8%			1	0.0%	268	1.7%
9.4.3	Southwestern Tablelands					10	0.2%					9	0.1%
9.4.4	Flint Hills					131	2.7%			1	0.0%	42	0.3%
9.4.5	Cross Timbers			12	0.3%	9	0.2%			7	0.0%	24	0.2%
9.4.6	Edwards Plateau												

NA_L3 CODE	US_L3NAME	**American hornbeam** Median S=7 Assoc S-↓ N/S = 0.23		**black hickory** Median N=10 Assoc N-↑ N/S = 0.17		**hackberry** Median N=11 Assoc N-U N/S = 0.17		**American beech** Median N=8,S=7 Assoc N-U,S-↓ N/S = 0.76		**white ash** Median N=10 Assoc N-↑ N/S = 0.54		**green ash** Median N=10, =6 Assoc N-U, S-↓ N/S = 0.45	
		count	**%**	**count**	**%**	**count**	**%**	**count**	**%**	**count**	**%**	**count**	**%**
9.4.7	Texas Blackland Prairies									3	0.0%		
9.5.1	Western Gulf Coastal Plain	8	0.4%									28	0.2%
9.6.1	Southern Texas Plains												
10.1.2	Columbia Plateau												
10.1.3	Northern Basin and Range												
10.1.4	Wyoming Basin												
10.1.5	Central Basin and Range												
10.1.6	Colorado Plateaus												
10.1.7	Arizona/New Mexico Plateau												
10.1.8	Snake River Plain												
10.2.1	Mojave Basin and Range												
10.2.2	Sonoran Basin and Range												
10.2.10	Chihuahuan Deserts												
11.1.1	Southern and Central California Chaparral and Oak Woodlands												
11.1.2	Central California Valley												
11.1.3	Southern California Mountains												
12.1.1	Madrean Archipelago												
13.1.1	Arizona/New Mexico Mountains												
15.4.1	Southern Florida Coastal Plain											3	0.0%
Total Tree Counts		2137		3997		4902		20894		17266		15573	

NA_L3 CODE	US_L3NAME	honeylocust Median S=6 Assoc S-↓ N/S = 0.27		black walnut Median N=12,S=9 Assoc N-↑, S-↓ N/S = 0.08		Utah juniper Median N=3, S=1 Assoc N-↑, S-↓ N/S = 0.71		eastern redcedar Median S=7 Assoc S-↓ N/S = 0.3		sweetgum Median N=9, S=7 Assoc N-↑, S-↓ N/S = 0.37		yellow-poplar Median N=10 Assoc N-↑ N/S = 0.41	
		count	%	count	%	count	%	count	%	count	%	count	%
5.2.1	Northern Lakes and Forests							2	0.0%				
5.2.2	Northern Minnesota Wetlands												
5.3.1	Northeastern Highlands			6	0.1%			16	0.1%			91	0.4%
5.3.3	North Central Appalachians							4	0.0%			47	0.2%
6.2.3	Northern Rockies												
6.2.4	Canadian Rockies												
6.2.5	North Cascades												
6.2.7	Cascades												
6.2.8	Eastern Cascades Slopes and Foothills												
6.2.9	Blue Mountains												
6.2.10	Middle Rockies					33	0.3%						
6.2.11	Klamath Mountains												
6.2.12	Sierra Nevada												
6.2.13	Wasatch and Uinta Mountains					698	6.3%						
6.2.14	Southern Rockies					110	1.0%						
6.2.15	Idaho Batholith												
7.1.7	Puget Lowland												
7.1.8	Coast Range												
7.1.9	Willamette Valley												
8.1.1	Eastern Great Lakes Lowlands			30	0.5%			29	0.2%				
8.1.3	Northern Allegheny Plateau			34	0.6%			11	0.1%				

NA_L3 CODE	US_L3NAME	honeylocust Median S=6 Assoc S-↓ N/S = 0.27		black walnut Median N=12,S=9 Assoc N-↑, S-↓ N/S = 0.08		Utah juniper Median N=3, S=1 Assoc N-↑, S-↓ N/S = 0.71		eastern redcedar Median S=7 Assoc S-↓ N/S = 0.3		sweetgum Median N=9, S=7 Assoc N-↑, S-↓ N/S = 0.37		yellow-poplar Median N=10 Assoc N-↑ N/S = 0.41	
		count	%	count	%	count	%	count	%	count	%	count	%
8.1.4	North Central Hardwood Forests			13	0.2%			109	0.8%				
8.1.5	Driftless Area	6	0.3%	404	7.1%			276	1.9%				
8.1.6	Southern Michigan/Northern Indiana Drift Plains	2	0.1%	119	2.1%			24	0.2%			58	0.2%
8.1.7	Northeastern Coastal Zone			14	0.2%			84	0.6%	2	0.0%	46	0.2%
8.1.8	Acadian Plains and Hills												
8.1.10	Erie Drift Plain	1	0.0%	50	0.9%					1	0.0%	160	0.7%
8.2.1	Southeastern Wisconsin Till Plains	1	0.0%	80	1.4%			88	0.6%				
8.2.2	Huron/Erie Lake Plains	7	0.3%	35	0.6%			1	0.0%			4	0.0%
8.2.3	Central Corn Belt Plains	72	3.6%	130	2.3%			8	0.1%			3	0.0%
8.2.4	Eastern Corn Belt Plains	130	6.5%	417	7.4%			142	1.0%	62	0.2%	183	0.8%
8.3.1	Northern Piedmont	1	0.0%	142	2.5%			221	1.5%	37	0.1%	659	2.7%
8.3.2	Interior River Valleys and Hills	203	10.1%	457	8.1%			622	4.3%	444	1.5%	377	1.6%
8.3.3	Interior Plateau	166	8.3%	796	14.0%			3325	23.1%	791	2.7%	2259	9.3%
8.3.4	Piedmont	13	0.6%	153	2.7%			1031	7.2%	5544	19.0%	5178	21.4%
8.3.5	Southeastern Plains	11	0.5%	54	1.0%			645	4.5%	9331	32.0%	3421	14.2%
8.3.6	Mississippi Valley Loess Plains	32	1.6%	24	0.4%			163	1.1%	1538	5.3%	272	1.1%
8.3.7	South Central Plains	58	2.9%	14	0.2%			167	1.2%	4762	16.3%	13	0.1%
8.3.8	East Central Texas Plains	17	0.8%	4	0.1%			122	0.8%	209	0.7%		
8.4.1	Ridge and Valley	10	0.5%	353	6.2%			722	5.0%	546	1.9%	1657	6.9%
8.4.2	Central Appalachians	4	0.2%	50	0.9%			41	0.3%	94	0.3%	2997	12.4%
8.4.3	Western Allegheny Plateau	15	0.7%	386	6.8%			44	0.3%	14	0.0%	2390	9.9%
8.4.4	Blue Ridge	6	0.3%	65	1.1%			32	0.2%	65	0.2%	2779	11.5%

NA_L3 CODE	US_L3NAME	honeylocust Median S=6 Assoc S-↓ N/S = 0.27		black walnut Median N=12,S=9 Assoc N-↑, S-↓ N/S = 0.08		Utah juniper Median N=3, S=1 Assoc N-↑, S-↓ N/S = 0.71		eastern redcedar Median S=7 Assoc S-↓ N/S = 0.3		sweetgum Median N=9, S=7 Assoc N-↑, S-↓ N/S = 0.37		yellow-poplar Median N=10 Assoc N-↑ N/S = 0.41	
		count	%	count	%	count	%	count	%	count	%	count	%
8.4.5	Ozark Highlands	180	9.0%	710	12.5%			3519	24.5%	146	0.5%	4	0.0%
8.4.6	Boston Mountains	6	0.3%	28	0.5%			285	2.0%	172	0.6%		
8.4.7	Arkansas Valley	15	0.7%	9	0.2%			606	4.2%	226	0.8%		
8.4.8	Ouachita Mountains	17	0.8%	4	0.1%			210	1.5%	311	1.1%		
8.4.9	Southwestern Appalachians	4	0.2%	37	0.7%			370	2.6%	620	2.1%	1035	4.3%
8.5.1	Middle Atlantic Coastal Plain	1	0.0%	12	0.2%			13	0.1%	2428	8.3%	467	1.9%
8.5.2	Mississippi Alluvial Plain	100	5.0%	7	0.1%			19	0.1%	574	2.0%	10	0.0%
8.5.3	Southern Coastal Plain	2	0.1%					26	0.2%	1111	3.8%	43	0.2%
8.5.4	Atlantic Coastal Pine Barrens			2	0.0%			10	0.1%	60	0.2%	16	0.1%
9.2.1	Northern Glaciated Plains	9	0.4%										
9.2.2	Lake Agassiz Plain												
9.2.3	Western Corn Belt Plains	345	17.2%	325	5.7%			318	2.2%				
9.2.4	Central Irregular Plains	496	24.7%	617	10.9%			381	2.6%				
9.3.1	Northwestern Glaciated Plains							118	0.8%				
9.3.3	Northwestern Great Plains			1	0.0%			82	0.6%				
9.3.4	Nebraska Sand Hills							92	0.6%				
9.4.1	High Plains			1	0.0%			23	0.2%				
9.4.2	Central Great Plains	46	2.3%	22	0.4%			275	1.9%				
9.4.3	Southwestern Tablelands			6	0.1%			8	0.1%				
9.4.4	Flint Hills	30	1.5%	45	0.8%			55	0.4%				
9.4.5	Cross Timbers	3	0.1%	10	0.2%			17	0.1%				
9.4.6	Edwards Plateau												

NA_L3 CODE	US_L3NAME	honeylocust Median S=6 Assoc S-↓ N/S = 0.27		black walnut Median N=12,S=9 Assoc N-↑, S-↓ N/S = 0.08		Utah juniper Median N=3, S=1 Assoc N-↑, S-↓ N/S = 0.71		eastern redcedar Median S=7 Assoc S-↓ N/S = 0.3		sweetgum Median N=9, S=7 Assoc N-↑, S-↓ N/S = 0.37		yellow-poplar Median N=10 Assoc N-↑ N/S = 0.41	
		count	%	count	%	count	%	count	%	count	%	count	%
9.4.7	Texas Blackland Prairies							12	0.1%				
9.5.1	Western Gulf Coastal Plain							11	0.1%	92	0.3%		
9.6.1	Southern Texas Plains												
10.1.2	Columbia Plateau												
10.1.3	Northern Basin and Range					405	3.7%						
10.1.4	Wyoming Basin					66	0.6%						
10.1.5	Central Basin and Range					3112	28.1%						
10.1.6	Colorado Plateaus					3935	35.5%						
10.1.7	Arizona/New Mexico Plateau					1601	14.4%						
10.1.8	Snake River Plain												
10.2.1	Mojave Basin and Range					115	1.0%						
10.2.2	Sonoran Basin and Range												
10.2.10	Chihuahuan Deserts												
11.1.1	Southern and Central California Chaparral and Oak Woodlands												
11.1.2	Central California Valley												
11.1.3	Southern California Mountains												
12.1.1	Madrean Archipelago					1	0.0%						
13.1.1	Arizona/New Mexico Mountains					1008	9.1%						
15.4.1	Southern Florida Coastal Plain												
	Total Tree Count	2009		5666		11084		14379		29180		24169	

NA_L3 CODE	US_L3NAME	tanoak Median N=4 Assoc N-U N/S = 0.57		Osage-orange Median S=5 Assoc S-↓ N/S = 0.36		sweetbay Median N=7 Assoc N-U N/S = 0.34		water tupelo Median S=8 Assoc S-↓ N/S = 0.5		swamp tupelo Median N=7 Assoc N-U N/S = 0.47		white spruce Median S=4 Assoc S-↓ N/S =	
		count	%	count	%	count	%	count	%	count	%	count	%
5.2.1	Northern Lakes and Forests											3739	63.0%
5.2.2	Northern Minnesota Wetlands											245	4.1%
5.3.1	Northeastern Highlands											716	12.1%
5.3.3	North Central Appalachians											28	0.5%
6.2.3	Northern Rockies												
6.2.4	Canadian Rockies												
6.2.5	North Cascades												
6.2.7	Cascades	1	0.0%										
6.2.8	Eastern Cascades Slopes and Foothills												
6.2.9	Blue Mountains												
6.2.10	Middle Rockies											194	3.3%
6.2.11	Klamath Mountains	1561	51.9%										
6.2.12	Sierra Nevada	116	3.9%										
6.2.13	Wasatch and Uinta Mountains												
6.2.14	Southern Rockies												
6.2.15	Idaho Batholith												
7.1.7	Puget Lowland												
7.1.8	Coast Range	1276	42.4%										
7.1.9	Willamette Valley												
8.1.1	Eastern Great Lakes Lowlands											2	0.0%
8.1.3	Northern Allegheny Plateau											7	0.1%

NA_L3 CODE	US_L3NAME	tanoak Median N=4 Assoc N-U N/S = 0.57		Osage-orange Median S=5 Assoc S-↓ N/S = 0.36		sweetbay Median N=7 Assoc N-U N/S = 0.34		water tupelo Median S=8 Assoc S-↓ N/S = 0.5		swamp tupelo Median N=7 Assoc N-U N/S = 0.47		white spruce Median S=4 Assoc S-↓ N/S =	
		count	%	count	%	count	%	count	%	count	%	count	%
8.1.4	North Central Hardwood Forests											127	2.1%
8.1.5	Driftless Area											43	0.7%
8.1.6	Southern Michigan/Northern Indiana Drift Plains			3	0.1%							30	0.5%
8.1.7	Northeastern Coastal Zone											2	0.0%
8.1.8	Acadian Plains and Hills											754	12.7%
8.1.10	Erie Drift Plain			17	0.7%								
8.2.1	Southeastern Wisconsin Till Plains											25	0.4%
8.2.2	Huron/Erie Lake Plains											2	0.0%
8.2.3	Central Corn Belt Plains			73	3.1%								
8.2.4	Eastern Corn Belt Plains			139	5.8%								
8.3.1	Northern Piedmont			8	0.3%							1	0.0%
8.3.2	Interior River Valleys and Hills			231	9.7%			33	1.3%				
8.3.3	Interior Plateau			281	11.8%			2	0.1%	1	0.0%		
8.3.4	Piedmont			2	0.1%	34	1.0%			56	0.7%		
8.3.5	Southeastern Plains			72	3.0%	1848	56.6%	686	26.3%	3615	45.6%		
8.3.6	Mississippi Valley Loess Plains			5	0.2%	11	0.3%	59	2.3%	5	0.1%		
8.3.7	South Central Plains			81	3.4%	188	5.8%	147	5.6%	43	0.5%		
8.3.8	East Central Texas Plains			47	2.0%								
8.4.1	Ridge and Valley			49	2.1%	2	0.1%			1	0.0%		
8.4.2	Central Appalachians												
8.4.3	Western Allegheny Plateau			57	2.4%							4	0.1%
8.4.4	Blue Ridge												

NA_L3 CODE	US_L3NAME	tanoak Median N=4 Assoc N-U N/S = 0.57		Osage-orange Median S=5 Assoc S-↓ N/S = 0.36		sweetbay Median N=7 Assoc N-U N/S = 0.34		water tupelo Median S=8 Assoc S-↓ N/S = 0.5		swamp tupelo Median N=7 Assoc N-U N/S = 0.47		white spruce Median S=4 Assoc S-↓ N/S =	
		count	%	count	%	count	%	count	%	count	%	count	%
8.4.5	Ozark Highlands			105	4.4%								
8.4.6	Boston Mountains			1	0.0%								
8.4.7	Arkansas Valley			16	0.7%					2	0.0%		
8.4.8	Ouachita Mountains			24	1.0%								
8.4.9	Southwestern Appalachians							2	0.1%				
8.5.1	Middle Atlantic Coastal Plain					170	5.2%	540	20.7%	1491	18.8%		
8.5.2	Mississippi Alluvial Plain							682	26.2%	24	0.3%		
8.5.3	Southern Coastal Plain					993	30.4%	391	15.0%	2697	34.0%		
8.5.4	Atlantic Coastal Pine Barrens					10	0.3%						
9.2.1	Northern Glaciated Plains												
9.2.2	Lake Agassiz Plain											4	0.1%
9.2.3	Western Corn Belt Plains			141	5.9%							12	0.2%
9.2.4	Central Irregular Plains			758	31.8%								
9.3.1	Northwestern Glaciated Plains												
9.3.3	Northwestern Great Plains												
9.3.4	Nebraska Sand Hills												
9.4.1	High Plains												
9.4.2	Central Great Plains			88	3.7%								
9.4.3	Southwestern Tablelands			7	0.3%								
9.4.4	Flint Hills			154	6.5%								
9.4.5	Cross Timbers			22	0.9%								
9.4.6	Edwards Plateau												

NA_L3 CODE	US_L3NAME	tanoak Median N=4 Assoc N-U N/S = 0.57		Osage-orange Median S=5 Assoc S-↓ N/S = 0.36		sweetbay Median N=7 Assoc N-U N/S = 0.34		water tupelo Median S=8 Assoc S-↓ N/S = 0.5		swamp tupelo Median N=7 Assoc N-U N/S = 0.47		white spruce Median S=4 Assoc S-↓ N/S =	
		count	%	count	%	count	%	count	%	count	%	count	%
9.4.7	Texas Blackland Prairies			1	0.0%								
9.5.1	Western Gulf Coastal Plain			2	0.1%	1	0.0%	65	2.5%				
9.6.1	Southern Texas Plains												
10.1.2	Columbia Plateau												
10.1.3	Northern Basin and Range												
10.1.4	Wyoming Basin												
10.1.5	Central Basin and Range												
10.1.6	Colorado Plateaus												
10.1.7	Arizona/New Mexico Plateau												
10.1.8	Snake River Plain												
10.2.1	Mojave Basin and Range												
10.2.2	Sonoran Basin and Range												
10.2.10	Chihuahuan Deserts												
11.1.1	Southern and Central California Chaparral and Oak Woodlands	55	1.8%										
11.1.2	Central California Valley												
11.1.3	Southern California Mountains												
12.1.1	Madrean Archipelago												
13.1.1	Arizona/New Mexico Mountains												
15.4.1	Southern Florida Coastal Plain					6	0.2%			1	0.0%		
	Total Tree Count	3009		2384		3263		2607		7936		5935	

NA_L3 CODE	US_L3NAME	**shortleaf pine** Median S=6 Assoc S-↓ N/S = 0.16		**slash pine** Median S=5 Assoc S-↓ N/S = 0.46		**singleleaf pinyon** Median N=3 Assoc N-↑ N/S = 0.58		**longleaf pine** Median N=8 Assoc N-U N/S = 0.45		**red pine** Median N=8, S=5 Assoc N-↑, S-↓ N/S = 0.53		**pitch pine** Median N=10 Assoc N-U N/S = 0.66	
		count	**%**	**count**	**%**	**count**	**%**	**count**	**%**	**count**	**%**	**count**	**%**
5.2.1	Northern Lakes and Forests									5823	65.3%		
5.2.2	Northern Minnesota Wetlands									192	2.2%		
5.3.1	Northeastern Highlands									151	1.7%	81	3.1%
5.3.3	North Central Appalachians									35	0.4%	37	1.4%
6.2.3	Northern Rockies												
6.2.4	Canadian Rockies												
6.2.5	North Cascades												
6.2.7	Cascades												
6.2.8	Eastern Cascades Slopes and Foothills												
6.2.9	Blue Mountains												
6.2.10	Middle Rockies												
6.2.11	Klamath Mountains												
6.2.12	Sierra Nevada					109	3.0%						
6.2.13	Wasatch and Uinta Mountains					11	0.3%						
6.2.14	Southern Rockies												
6.2.15	Idaho Batholith												
7.1.7	Puget Lowland												
7.1.8	Coast Range												
7.1.9	Willamette Valley												
8.1.1	Eastern Great Lakes Lowlands									66	0.7%	6	0.2%
8.1.3	Northern Allegheny Plateau									120	1.3%	2	0.1%

NA_L3 CODE	US_L3NAME	shortleaf pine Median S=6 Assoc S-↓ N/S = 0.16		slash pine Median S=5 Assoc S-↓ N/S = 0.46		singleleaf pinyon Median N=3 Assoc N-↑ N/S = 0.58		longleaf pine Median N=8 Assoc N-U N/S = 0.45		red pine Median N=8, S=5 Assoc N-↑, S-↓ N/S = 0.53		pitch pine Median N=10 Assoc N-U N/S = 0.66	
		count	%	count	%	count	%	count	%	count	%	count	%
8.1.4	North Central Hardwood Forests									1456	16.3%		
8.1.5	Driftless Area									327	3.7%		
8.1.6	Southern Michigan/Northern Indiana Drift Plains									367	4.1%	1	0.0%
8.1.7	Northeastern Coastal Zone									7	0.1%	88	3.4%
8.1.8	Acadian Plains and Hills									159	1.8%	3	0.1%
8.1.10	Erie Drift Plain									12	0.1%		
8.2.1	Southeastern Wisconsin Till Plains									80	0.9%		
8.2.2	Huron/Erie Lake Plains									51	0.6%		
8.2.3	Central Corn Belt Plains									5	0.1%		
8.2.4	Eastern Corn Belt Plains									6	0.1%		
8.3.1	Northern Piedmont	7	0.1%										
8.3.2	Interior River Valleys and Hills	19	0.1%							21	0.2%	2	0.1%
8.3.3	Interior Plateau	191	1.4%							2	0.0%	5	0.2%
8.3.4	Piedmont	2037	15.3%	16	0.2%			203	4.4%			17	0.7%
8.3.5	Southeastern Plains	1451	10.9%	3418	34.4%			2729	58.9%			1	0.0%
8.3.6	Mississippi Valley Loess Plains	94	0.7%	2	0.0%			1	0.0%				
8.3.7	South Central Plains	1336	10.1%	190	1.9%			244	5.3%				
8.3.8	East Central Texas Plains	23	0.2%	6	0.1%								
8.4.1	Ridge and Valley	291	2.2%					87	1.9%	6	0.1%	363	14.1%
8.4.2	Central Appalachians	23	0.2%							11	0.1%	50	1.9%
8.4.3	Western Allegheny Plateau	39	0.3%							18	0.2%	81	3.1%
8.4.4	Blue Ridge	275	2.1%									210	8.1%

NA_L3 CODE	US_L3NAME	shortleaf pine Median S=6 Assoc S-↓ N/S = 0.16		slash pine Median S=5 Assoc S-↓ N/S = 0.46		singleleaf pinyon Median N=3 Assoc N-↑ N/S = 0.58		longleaf pine Median N=8 Assoc N-U N/S = 0.45		red pine Median N=8, S=5 Assoc N-↑, S-↓ N/S = 0.53		pitch pine Median N=10 Assoc N-U N/S = 0.66	
		count	%	count	%	count	%	count	%	count	%	count	%
8.4.5	Ozark Highlands	2400	18.1%										
8.4.6	Boston Mountains	567	4.3%										
8.4.7	Arkansas Valley	1059	8.0%										
8.4.8	Ouachita Mountains	3137	23.6%										
8.4.9	Southwestern Appalachians	251	1.9%					9	0.2%			9	0.3%
8.5.1	Middle Atlantic Coastal Plain	25	0.2%	105	1.1%			301	6.5%			33	1.3%
8.5.2	Mississippi Alluvial Plain	11	0.1%										
8.5.3	Southern Coastal Plain	2	0.0%	6030	60.6%			1054	22.7%				
8.5.4	Atlantic Coastal Pine Barrens	38	0.3%									1589	61.6%
9.2.1	Northern Glaciated Plains												
9.2.2	Lake Agassiz Plain												
9.2.3	Western Corn Belt Plains												
9.2.4	Central Irregular Plains									2	0.0%		
9.3.1	Northwestern Glaciated Plains												
9.3.3	Northwestern Great Plains												
9.3.4	Nebraska Sand Hills												
9.4.1	High Plains												
9.4.2	Central Great Plains												
9.4.3	Southwestern Tablelands												
9.4.4	Flint Hills												
9.4.5	Cross Timbers	2	0.0%										
9.4.6	Edwards Plateau												

NA_L3 CODE	US_L3NAME	shortleaf pine Median S=6 Assoc S-↓ N/S = 0.16		slash pine Median S=5 Assoc S-↓ N/S = 0.46		singleleaf pinyon Median N=3 Assoc N-↑ N/S = 0.58		longleaf pine Median N=8 Assoc N-U N/S = 0.45		red pine Median N=8, S=5 Assoc N-↑, S-↓ N/S = 0.53		pitch pine Median N=10 Assoc N-U N/S = 0.66	
		count	%	count	%	count	%	count	%	count	%	count	%
9.4.7	Texas Blackland Prairies												
9.5.1	Western Gulf Coastal Plain							7	0.2%				
9.6.1	Southern Texas Plains												
10.1.2	Columbia Plateau												
10.1.3	Northern Basin and Range					42	1.2%						
10.1.4	Wyoming Basin												
10.1.5	Central Basin and Range					2988	83.5%						
10.1.6	Colorado Plateaus					46	1.3%						
10.1.7	Arizona/New Mexico Plateau					86	2.4%						
10.1.8	Snake River Plain												
10.2.1	Mojave Basin and Range					153	4.3%						
10.2.2	Sonoran Basin and Range												
10.2.10	Chihuahuan Deserts												
11.1.1	Southern and Central California Chaparral and Oak Woodlands												
11.1.2	Central California Valley												
11.1.3	Southern California Mountains					127	3.5%						
12.1.1	Madrean Archipelago					3	0.1%						
13.1.1	Arizona/New Mexico Mountains					14	0.4%						
15.4.1	Southern Florida Coastal Plain			178	1.8%								
	Total Tree Count	13278		9945		3579		4635		8917		2578	

NA_L3 CODE	US_L3NAME	eastern white pine Median N=8, S=6 Assoc N-↑, S-↓ N/S = 0.59		loblolly pine Median S=7 Assoc S-↓ N/S = 0.32		bigtooth aspen Median S=6 Assoc S-↓ N/S = 0.57		quaking aspen Median N=7 Assoc N-U N/S = 0.6		black cherry Median N=11 Assoc N-U N/S = 0.33		Douglas-fir Median N=3, S=1 Assoc N-↑, S-↓ N/S = 0.65	
		count	%	count	%	count	%	count	%	count	%	count	%
5.2.1	Northern Lakes and Forests	3921	19.2%			6123	61.0%	23006	55.1%	1726	8.4%		
5.2.2	Northern Minnesota Wetlands	34	0.2%			13	0.1%	2488	6.0%				
5.3.1	Northeastern Highlands	3744	18.3%			397	4.0%	961	2.3%	1284	6.3%		
5.3.3	North Central Appalachians	525	2.6%			89	0.9%	136	0.3%	1260	6.2%		
6.2.3	Northern Rockies							44	0.1%			4096	10.4%
6.2.4	Canadian Rockies							74	0.2%			627	1.6%
6.2.5	North Cascades											2101	5.3%
6.2.7	Cascades											8882	22.6%
6.2.8	Eastern Cascades Slopes and Foothills							20	0.0%			1394	3.5%
6.2.9	Blue Mountains							6	0.0%			2946	7.5%
6.2.10	Middle Rockies							264	0.6%			3404	8.6%
6.2.11	Klamath Mountains							3	0.0%			5771	14.7%
6.2.12	Sierra Nevada							21	0.1%			880	2.2%
6.2.13	Wasatch and Uinta Mountains							2195	5.3%			653	1.7%
6.2.14	Southern Rockies							3606	8.6%			1841	4.7%
6.2.15	Idaho Batholith							3	0.0%			1294	3.3%
7.1.7	Puget Lowland											335	0.9%
7.1.8	Coast Range											3526	9.0%
7.1.9	Willamette Valley											155	0.4%
8.1.1	Eastern Great Lakes Lowlands	515	2.5%			71	0.7%	266	0.6%	344	1.7%		
8.1.3	Northern Allegheny Plateau	769	3.8%			138	1.4%	340	0.8%	697	3.4%	8	0.0%

NA_L3 CODE	US_L3NAME	eastern white pine Median N=8, S=6 Assoc N-↑, S-↓ N/S = 0.59		loblolly pine Median S=7 Assoc S-↓ N/S = 0.32		bigtooth aspen Median S=6 Assoc S-↓ N/S = 0.57		quaking aspen Median N=7 Assoc N-U N/S = 0.6		black cherry Median N=11 Assoc N-U N/S = 0.33		Douglas-fir Median N=3, S=1 Assoc N-↑, S-↓ N/S = 0.65	
		count	%	count	%	count	%	count	%	count	%	count	%
8.1.4	North Central Hardwood Forests	1587	7.8%			935	9.3%	3258	7.8%	608	3.0%		
8.1.5	Driftless Area	379	1.9%			425	4.2%	383	0.9%	774	3.8%		
8.1.6	Southern Michigan/Northern Indiana Drift Plains	320	1.6%	4	0.0%	308	3.1%	250	0.6%	1233	6.0%	1	0.0%
8.1.7	Northeastern Coastal Zone	2299	11.2%			124	1.2%	152	0.4%	244	1.2%		
8.1.8	Acadian Plains and Hills	2113	10.3%			513	5.1%	970	2.3%	136	0.7%		
8.1.10	Erie Drift Plain	25	0.1%			77	0.8%	143	0.3%	727	3.6%		
8.2.1	Southeastern Wisconsin Till Plains	129	0.6%			22	0.2%	233	0.6%	442	2.2%		
8.2.2	Huron/Erie Lake Plains	103	0.5%			177	1.8%	308	0.7%	133	0.7%		
8.2.3	Central Corn Belt Plains	23	0.1%					16	0.0%	308	1.5%		
8.2.4	Eastern Corn Belt Plains	45	0.2%			15	0.1%	9	0.0%	510	2.5%		
8.3.1	Northern Piedmont	55	0.3%	30	0.0%	14	0.1%	6	0.0%	208	1.0%	2	0.0%
8.3.2	Interior River Valleys and Hills	14	0.1%	34	0.1%	6	0.1%			485	2.4%		
8.3.3	Interior Plateau	91	0.4%	514	0.9%	21	0.2%			803	3.9%		
8.3.4	Piedmont	353	1.7%	13499	22.4%	7	0.1%			781	3.8%		
8.3.5	Southeastern Plains	1	0.0%	20564	34.1%	5	0.0%			929	4.5%		
8.3.6	Mississippi Valley Loess Plains			1302	2.2%					217	1.1%		
8.3.7	South Central Plains			12048	20.0%					120	0.6%		
8.3.8	East Central Texas Plains			120	0.2%					4	0.0%		
8.4.1	Ridge and Valley	1155	5.6%	1343	2.2%	81	0.8%	17	0.0%	1179	5.8%	1	0.0%
8.4.2	Central Appalachians	198	1.0%	4	0.0%	77	0.8%	19	0.0%	1189	5.8%		
8.4.3	Western Allegheny Plateau	340	1.7%	98	0.2%	398	4.0%	90	0.2%	2285	11.2%		
8.4.4	Blue Ridge	1531	7.5%	217	0.4%	1	0.0%			334	1.6%		

NA_L3 CODE	US_L3NAME	eastern white pine Median N=8, S=6 Assoc N-↑, S-↓ N/S = 0.59		loblolly pine Median S=7 Assoc S-↓ N/S = 0.32		bigtooth aspen Median S=6 Assoc S-↓ N/S = 0.57		quaking aspen Median N=7 Assoc N-U N/S = 0.6		black cherry Median N=11 Assoc N-U N/S = 0.33		Douglas-fir Median N=3, S=1 Assoc N-↑, S-↓ N/S = 0.65	
		count	%	count	%	count	%	count	%	count	%	count	%
8.4.5	Ozark Highlands			20	0.0%					392	1.9%		
8.4.6	Boston Mountains			48	0.1%					80	0.4%		
8.4.7	Arkansas Valley			170	0.3%					62	0.3%		
8.4.8	Ouachita Mountains			666	1.1%					83	0.4%		
8.4.9	Southwestern Appalachians	112	0.5%	1288	2.1%					211	1.0%		
8.5.1	Middle Atlantic Coastal Plain			6065	10.0%	1	0.0%			192	0.9%		
8.5.2	Mississippi Alluvial Plain			87	0.1%					29	0.1%		
8.5.3	Southern Coastal Plain			1960	3.2%					64	0.3%		
8.5.4	Atlantic Coastal Pine Barrens	93	0.5%	2	0.0%	2	0.0%			26	0.1%		
9.2.1	Northern Glaciated Plains							301	0.7%				
9.2.2	Lake Agassiz Plain							1548	3.7%	2	0.0%		
9.2.3	Western Corn Belt Plains					1	0.0%	71	0.2%	180	0.9%		
9.2.4	Central Irregular Plains									159	0.8%		
9.3.1	Northwestern Glaciated Plains							9	0.0%				
9.3.3	Northwestern Great Plains							24	0.1%	1	0.0%	190	0.5%
9.3.4	Nebraska Sand Hills												
9.4.1	High Plains												
9.4.2	Central Great Plains												
9.4.3	Southwestern Tablelands												
9.4.4	Flint Hills												
9.4.5	Cross Timbers									2	0.0%		
9.4.6	Edwards Plateau												

NA_L3 CODE	US_L3NAME	eastern white pine Median N=8, S=6 Assoc N-↑, S-↓ N/S = 0.59		loblolly pine Median S=7 Assoc S-↓ N/S = 0.32		bigtooth aspen Median S=6 Assoc S-↓ N/S = 0.57		quaking aspen Median N=7 Assoc N-U N/S = 0.6		black cherry Median N=11 Assoc N-U N/S = 0.33		Douglas-fir Median N=3, S=1 Assoc N-↑, S-↓ N/S = 0.65	
		count	%	count	%	count	%	count	%	count	%	count	%
9.4.7	Texas Blackland Prairies			54	0.1%								
9.5.1	Western Gulf Coastal Plain			237	0.4%					3	0.0%		
9.6.1	Southern Texas Plains												
10.1.2	Columbia Plateau							29	0.1%			102	0.3%
10.1.3	Northern Basin and Range							80	0.2%			102	0.3%
10.1.4	Wyoming Basin											25	0.1%
10.1.5	Central Basin and Range							44	0.1%			21	0.1%
10.1.6	Colorado Plateaus							187	0.4%			314	0.8%
10.1.7	Arizona/New Mexico Plateau											14	0.0%
10.1.8	Snake River Plain							24	0.1%				
10.2.1	Mojave Basin and Range												
10.2.2	Sonoran Basin and Range												
10.2.10	Chihuahuan Deserts												
11.1.1	Southern and Central California Chaparral and Oak Woodlands											173	0.4%
11.1.2	Central California Valley												
11.1.3	Southern California Mountains												
12.1.1	Madrean Archipelago											20	0.1%
13.1.1	Arizona/New Mexico Mountains							144	0.3%			486	1.2%
15.4.1	Southern Florida Coastal Plain												
	Total Tree Count	20474		60374		10041		41748		20446		39364	

NA_L3 CODE	US_L3NAME	scarlet oak Median N=10 Assoc N-U N/S = 0.37		northern pin oak Median N=10,S=5 Assoc N-↑, S-↓ N/S = 0.41		southern red oak Median N=9 Assoc N-U N/S = 0.36		bur oak Median N=9 Assoc N-↓ N/S = 0.59		water oak Median N=8, S=7 Assoc N-U, S-↓ N/S = 0.26		chestnut oak Median N=9 Assoc N-U N/S = 0.45	
		count	%	count	%	count	%	count	%	count	%	count	%
5.2.1	Northern Lakes and Forests			1942	53.7%			2075	28.9%				
5.2.2	Northern Minnesota Wetlands			1	0.0%			128	1.8%				
5.3.1	Northeastern Highlands	104	1.1%					2	0.0%			349	1.7%
5.3.3	North Central Appalachians	228	2.5%									1067	5.2%
6.2.3	Northern Rockies												
6.2.4	Canadian Rockies												
6.2.5	North Cascades												
6.2.7	Cascades												
6.2.8	Eastern Cascades Slopes and Foothills												
6.2.9	Blue Mountains												
6.2.10	Middle Rockies							128	1.8%				
6.2.11	Klamath Mountains												
6.2.12	Sierra Nevada												
6.2.13	Wasatch and Uinta Mountains												
6.2.14	Southern Rockies												
6.2.15	Idaho Batholith												
7.1.7	Puget Lowland												
7.1.8	Coast Range												
7.1.9	Willamette Valley												
8.1.1	Eastern Great Lakes Lowlands			1	0.0%			31	0.4%			3	0.0%

NA_L3 CODE	US_L3NAME	scarlet oak Median N=10 Assoc N-U N/S = 0.37		northern pin oak Median N=10,S=5 Assoc N-↑, S-↓ N/S = 0.41		southern red oak Median N=9 Assoc N-U N/S = 0.36		bur oak Median N=9 Assoc N-↓ N/S = 0.59		water oak Median N=8, S=7 Assoc N-U, S-↓ N/S = 0.26		chestnut oak Median N=9 Assoc N-U N/S = 0.45	
		count	%	count	%	count	%	count	%	count	%	count	%
8.1.3	Northern Allegheny Plateau	20	0.2%					3	0.0%			216	1.0%
8.1.4	North Central Hardwood Forests			1158	32.0%			1503	20.9%				
8.1.5	Driftless Area			220	6.1%			713	9.9%				
8.1.6	Southern Michigan/Northern Indiana Drift Plains	6	0.1%	124	3.4%			41	0.6%				
8.1.7	Northeastern Coastal Zone	567	6.2%					8	0.1%			174	0.8%
8.1.8	Acadian Plains and Hills												
8.1.10	Erie Drift Plain	4	0.0%					8	0.1%			5	0.0%
8.2.1	Southeastern Wisconsin Till Plains			65	1.8%			129	1.8%				
8.2.2	Huron/Erie Lake Plains	3	0.0%	65	1.8%			42	0.6%				
8.2.3	Central Corn Belt Plains			2	0.1%			38	0.5%				
8.2.4	Eastern Corn Belt Plains	4	0.0%					23	0.3%			4	0.0%
8.3.1	Northern Piedmont	56	0.6%			75	1.0%					331	1.6%
8.3.2	Interior River Valleys and Hills	64	0.7%			50	0.7%	43	0.6%			80	0.4%
8.3.3	Interior Plateau	476	5.2%			404	5.4%	8	0.1%	59	0.5%	1010	4.9%
8.3.4	Piedmont	786	8.6%			1431	19.1%			1374	11.1%	1180	5.7%
8.3.5	Southeastern Plains	263	2.9%			1906	25.5%			5531	44.8%	231	1.1%
8.3.6	Mississippi Valley Loess Plains	8	0.1%			217	2.9%	1	0.0%	454	3.7%		
8.3.7	South Central Plains	1	0.0%			1292	17.3%			2058	16.7%		
8.3.8	East Central Texas Plains					152	2.0%	1	0.0%	145	1.2%		
8.4.1	Ridge and Valley	1507	16.4%	1	0.0%	320	4.3%	1	0.0%	123	1.0%	7106	34.3%
8.4.2	Central Appalachians	745	8.1%			31	0.4%					2161	10.4%
8.4.3	Western Allegheny Plateau	479	5.2%			11	0.1%					1127	5.4%

NA_L3 CODE	US_L3NAME	scarlet oak Median N=10 Assoc N-U N/S = 0.37		northern pin oak Median N=10,S=5 Assoc N-↑, S-↓ N/S = 0.41		southern red oak Median N=9 Assoc N-U N/S = 0.36		bur oak Median N=9 Assoc N-↓ N/S = 0.59		water oak Median N=8, S=7 Assoc N-U, S-↓ N/S = 0.26		chestnut oak Median N=9 Assoc N-U N/S = 0.45	
		count	%	count	%	count	%	count	%	count	%	count	%
8.4.4	Blue Ridge	1507	16.4%			225	3.0%			20	0.2%	4206	20.3%
8.4.5	Ozark Highlands	1365	14.9%			494	6.6%	12	0.2%	1	0.0%		
8.4.6	Boston Mountains					70	0.9%	6	0.1%				
8.4.7	Arkansas Valley					126	1.7%	1	0.0%	109	0.9%		
8.4.8	Ouachita Mountains					107	1.4%			62	0.5%		
8.4.9	Southwestern Appalachians	502	5.5%			266	3.6%			72	0.6%	1358	6.6%
8.5.1	Middle Atlantic Coastal Plain	75	0.8%			190	2.5%			991	8.0%	24	0.1%
8.5.2	Mississippi Alluvial Plain	1	0.0%			58	0.8%			209	1.7%		
8.5.3	Southern Coastal Plain	1	0.0%			17	0.2%			1039	8.4%		
8.5.4	Atlantic Coastal Pine Barrens	394	4.3%			21	0.3%					79	0.4%
9.2.1	Northern Glaciated Plains							340	4.7%				
9.2.2	Lake Agassiz Plain							527	7.3%				
9.2.3	Western Corn Belt Plains			29	0.8%			486	6.8%				
9.2.4	Central Irregular Plains	1	0.0%	8	0.2%	1	0.0%	151	2.1%				
9.3.1	Northwestern Glaciated Plains							212	3.0%				
9.3.3	Northwestern Great Plains							421	5.9%				
9.3.4	Nebraska Sand Hills							16	0.2%				
9.4.1	High Plains							3	0.0%				
9.4.2	Central Great Plains							35	0.5%				
9.4.3	Southwestern Tablelands												
9.4.4	Flint Hills							43	0.6%				
9.4.5	Cross Timbers							1	0.0%				

NA_L3 CODE	US_L3NAME	scarlet oak Median N=10 Assoc N-U N/S = 0.37		northern pin oak Median N=10,S=5 Assoc N-↑, S-↓ N/S = 0.41		southern red oak Median N=9 Assoc N-U N/S = 0.36		bur oak Median N=9 Assoc N-↓ N/S = 0.59		water oak Median N=8, S=7 Assoc N-U, S-↓ N/S = 0.26		chestnut oak Median N=9 Assoc N-U N/S = 0.45	
		count	%	count	%	count	%	count	%	count	%	count	%
9.4.6	Edwards Plateau												
9.4.7	Texas Blackland Prairies					1	0.0%			3	0.0%		
9.5.1	Western Gulf Coastal Plain					14	0.2%			102	0.8%		
9.6.1	Southern Texas Plains												
10.1.2	Columbia Plateau												
10.1.3	Northern Basin and Range												
10.1.4	Wyoming Basin												
10.1.5	Central Basin and Range												
10.1.6	Colorado Plateaus												
10.1.7	Arizona/New Mexico Plateau												
10.1.8	Snake River Plain												
10.2.1	Mojave Basin and Range												
10.2.2	Sonoran Basin and Range												
10.2.10	Chihuahuan Deserts												
11.1.1	Southern and Central California Chaparral and Oak Woodlands												
11.1.2	Central California Valley												
11.1.3	Southern California Mountains												
12.1.1	Madrean Archipelago												
13.1.1	Arizona/New Mexico Mountains												
15.4.1	Southern Florida Coastal Plain												
	Total Tree Count	9167		3616		7479		7180		12352		20711	

NA_L3 CODE	US_L3NAME	**northern red oak** Median N=10 Assoc N-↑ N/S = 0.42		**black oak** Median N=11 Assoc N-↑ N/S = 0.13		**black locust** Median N=11, S=11 Assoc N-↑, S-↓ N/S = 0.18		**black willow** Median N=10,S=7 Assoc N-U,S-↓ N/S = 0.29		**sassafras** Median N=11 Assoc N-↑ N/S = 0.28		**pondcypress** Median N=7 Assoc N-↑ N/S = 0.71	
		count	**%**	**count**	**%**	**count**	**%**	**count**	**%**	**count**	**%**	**count**	**%**
5.2.1	Northern Lakes and Forests	5778	20.2%	690	3.7%	28	0.7%	28	1.4%	22	0.4%		
5.2.2	Northern Minnesota Wetlands	2	0.0%										
5.3.1	Northeastern Highlands	2993	10.5%	152	0.8%	28	0.7%	3	0.1%	25	0.5%		
5.3.3	North Central Appalachians	1020	3.6%	158	0.9%	6	0.2%	3	0.1%	221	4.4%		
6.2.3	Northern Rockies												
6.2.4	Canadian Rockies												
6.2.5	North Cascades												
6.2.7	Cascades												
6.2.8	Eastern Cascades Slopes and Foothills												
6.2.9	Blue Mountains												
6.2.10	Middle Rockies												
6.2.11	Klamath Mountains												
6.2.12	Sierra Nevada												
6.2.13	Wasatch and Uinta Mountains												
6.2.14	Southern Rockies												
6.2.15	Idaho Batholith												
7.1.7	Puget Lowland												
7.1.8	Coast Range												
7.1.9	Willamette Valley												
8.1.1	Eastern Great Lakes Lowlands	223	0.8%	23	0.1%	26	0.7%	40	2.0%	2	0.0%		
8.1.3	Northern Allegheny Plateau	822	2.9%	111	0.6%	51	1.3%	28	1.4%	8	0.2%		

NA_L3 CODE	US_L3NAME	**northern red oak** Median N=10 Assoc N-↑ N/S = 0.42		**black oak** Median N=11 Assoc N-↑ N/S = 0.13		**black locust** Median N=11, S=11 Assoc N-↑, S-↓ N/S = 0.18		**black willow** Median N=10,S=7 Assoc N-U,S-↓ N/S = 0.29		**sassafras** Median N=11 Assoc N-↑ N/S = 0.28		**pondcypress** Median N=7 Assoc N-↑ N/S = 0.71	
		count	**%**	**count**	**%**	**count**	**%**	**count**	**%**	**count**	**%**	**count**	**%**
8.1.4	North Central Hardwood Forests	1459	5.1%	827	4.5%	118	3.1%	74	3.6%				
8.1.5	Driftless Area	1474	5.2%	636	3.4%	110	2.9%	25	1.2%				
8.1.6	Southern Michigan/Northern Indiana Drift Plains	630	2.2%	684	3.7%	108	2.8%	68	3.3%	412	8.3%		
8.1.7	Northeastern Coastal Zone	1657	5.8%	794	4.3%	52	1.4%	6	0.3%	41	0.8%		
8.1.8	Acadian Plains and Hills	846	3.0%	8	0.0%								
8.1.10	Erie Drift Plain	180	0.6%	46	0.2%	62	1.6%	29	1.4%	49	1.0%		
8.2.1	Southeastern Wisconsin Till Plains	141	0.5%	48	0.3%	88	2.3%	71	3.5%				
8.2.2	Huron/Erie Lake Plains	175	0.6%	22	0.1%	6	0.2%	18	0.9%	58	1.2%		
8.2.3	Central Corn Belt Plains	60	0.2%	81	0.4%	74	1.9%	20	1.0%	39	0.8%		
8.2.4	Eastern Corn Belt Plains	200	0.7%	84	0.5%	148	3.9%	44	2.1%	115	2.3%		
8.3.1	Northern Piedmont	150	0.5%	122	0.7%	68	1.8%	12	0.6%	76	1.5%		
8.3.2	Interior River Valleys and Hills	461	1.6%	686	3.7%	149	3.9%	170	8.3%	496	10.0%		
8.3.3	Interior Plateau	664	2.3%	823	4.4%	338	8.8%	34	1.7%	785	15.8%		
8.3.4	Piedmont	773	2.7%	635	3.4%	68	1.8%	61	3.0%	52	1.0%		
8.3.5	Southeastern Plains	165	0.6%	350	1.9%	24	0.6%	274	13.4%	111	2.2%	487	14.1%
8.3.6	Mississippi Valley Loess Plains	39	0.1%	65	0.4%	36	0.9%	105	5.1%	88	1.8%		
8.3.7	South Central Plains	6	0.0%	60	0.3%	7	0.2%	114	5.6%	85	1.7%	4	0.1%
8.3.8	East Central Texas Plains			4	0.0%	2	0.1%	16	0.8%	15	0.3%		
8.4.1	Ridge and Valley	2335	8.2%	1371	7.4%	548	14.3%	8	0.4%	527	10.6%		
8.4.2	Central Appalachians	1114	3.9%	573	3.1%	397	10.4%	9	0.4%	381	7.7%		
8.4.3	Western Allegheny Plateau	793	2.8%	843	4.5%	437	11.4%	20	1.0%	668	13.4%		
8.4.4	Blue Ridge	1240	4.3%	501	2.7%	428	11.2%	7	0.3%	161	3.2%		

NA_L3 CODE	US_L3NAME	**northern red oak** Median N=10 Assoc N-↑ N/S = 0.42		**black oak** Median N=11 Assoc N-↑ N/S = 0.13		**black locust** Median N=11, S=11 Assoc N-↑, S-↓ N/S = 0.18		**black willow** Median N=10,S=7 Assoc N-U,S-↓ N/S = 0.29		**sassafras** Median N=11 Assoc N-↑ N/S = 0.28		**pondcypress** Median N=7 Assoc N-↑ N/S = 0.71	
		count	**%**	**count**	**%**	**count**	**%**	**count**	**%**	**count**	**%**	**count**	**%**
8.4.5	Ozark Highlands	1331	4.7%	6319	34.0%	20	0.5%	9	0.4%	269	5.4%		
8.4.6	Boston Mountains	631	2.2%	418	2.3%	51	1.3%			29	0.6%		
8.4.7	Arkansas Valley	147	0.5%	112	0.6%	4	0.1%	22	1.1%	4	0.1%		
8.4.8	Ouachita Mountains	364	1.3%	153	0.8%	4	0.1%	0	0.0%	1	0.0%		
8.4.9	Southwestern Appalachians	273	1.0%	442	2.4%	38	1.0%	5	0.2%	110	2.2%		
8.5.1	Middle Atlantic Coastal Plain	18	0.1%	67	0.4%	24	0.6%	71	3.5%	45	0.9%	109	3.2%
8.5.2	Mississippi Alluvial Plain	7	0.0%	11	0.1%	9	0.2%	412	20.1%	12	0.2%	37	1.1%
8.5.3	Southern Coastal Plain			2	0.0%			34	1.7%			2281	65.9%
8.5.4	Atlantic Coastal Pine Barrens	13	0.0%	236	1.3%	7	0.2%	10	0.5%	59	1.2%		
9.2.1	Northern Glaciated Plains												
9.2.2	Lake Agassiz Plain	2	0.0%					1	0.0%				
9.2.3	Western Corn Belt Plains	168	0.6%	67	0.4%	53	1.4%	72	3.5%				
9.2.4	Central Irregular Plains	198	0.7%	293	1.6%	160	4.2%	72	3.5%	3	0.1%		
9.3.1	Northwestern Glaciated Plains												
9.3.3	Northwestern Great Plains												
9.3.4	Nebraska Sand Hills							2	0.1%				
9.4.1	High Plains							1	0.0%				
9.4.2	Central Great Plains					42	1.1%	22	1.1%				
9.4.3	Southwestern Tablelands					3	0.1%	0	0.0%				
9.4.4	Flint Hills	3	0.0%					2	0.1%				
9.4.5	Cross Timbers	2	0.0%	42	0.2%			7	0.3%	2	0.0%		
9.4.6	Edwards Plateau												

NA_L3 CODE	US_L3NAME	**northern red oak** Median N=10 Assoc N-↑ N/S = 0.42		**black oak** Median N=11 Assoc N-↑ N/S = 0.13		**black locust** Median N=11, S=11 Assoc N-↑, S-↓ N/S = 0.18		**black willow** Median N=10,S=7 Assoc N-U,S-↓ N/S = 0.29		**sassafras** Median N=11 Assoc N-↑ N/S = 0.28		**pondcypress** Median N=7 Assoc N-↑ N/S = 0.71	
		count	**%**	**count**	**%**	**count**	**%**	**count**	**%**	**count**	**%**	**count**	**%**
9.4.7	Texas Blackland Prairies												
9.5.1	Western Gulf Coastal Plain							20	1.0%				
9.6.1	Southern Texas Plains												
10.1.2	Columbia Plateau												
10.1.3	Northern Basin and Range												
10.1.4	Wyoming Basin												
10.1.5	Central Basin and Range												
10.1.6	Colorado Plateaus												
10.1.7	Arizona/New Mexico Plateau												
10.1.8	Snake River Plain												
10.2.1	Mojave Basin and Range												
10.2.2	Sonoran Basin and Range												
10.2.10	Chihuahuan Deserts												
11.1.1	Southern and Central California Chaparral and Oak Woodlands												
11.1.2	Central California Valley												
11.1.3	Southern California Mountains												
12.1.1	Madrean Archipelago												
13.1.1	Arizona/New Mexico Mountains												
15.4.1	Southern Florida Coastal Plain											541	15.6%
	Total Tree Count	28557		18559		3822		2049		4971		3459	

NA_L3 CODE	US_L3NAME	**baldcypress** Median S=6 Assoc S-↓ N/S = 0.54		**American basswood** Median N=9, S=5 Assoc N-↑, S-↓ N/S = 0.4		**eastern hemlock** Median N=8 Assoc N-↓ N/S = 0.78		**western hemlock** Median N=3 Assoc N-U N/S = 0.34		**American elm** Median N=11, S=6 Assoc N-↑, S-↓ N/S = 0.25		**slippery elm** Median S=8 Assoc S-↓ N/S = 0.09	
		count	**%**	**count**	**%**	**count**	**%**	**count**	**%**	**count**	**%**	**count**	**%**
5.2.1	Northern Lakes and Forests			5591	44.4%	3098	13.5%			1082	7.6%	26	0.6%
5.2.2	Northern Minnesota Wetlands			139	1.1%					97	0.7%		
5.3.1	Northeastern Highlands			200	1.6%	7154	31.3%			162	1.1%	3	0.1%
5.3.3	North Central Appalachians			148	1.2%	1271	5.6%			19	0.1%	2	0.0%
6.2.3	Northern Rockies							705	7.5%				
6.2.4	Canadian Rockies							1	0.0%				
6.2.5	North Cascades							1747	18.6%				
6.2.7	Cascades							4379	46.5%				
6.2.8	Eastern Cascades Slopes and Foothills							59	0.6%				
6.2.9	Blue Mountains												
6.2.10	Middle Rockies									2	0.0%		
6.2.11	Klamath Mountains							28	0.3%				
6.2.12	Sierra Nevada												
6.2.13	Wasatch and Uinta Mountains												
6.2.14	Southern Rockies												
6.2.15	Idaho Batholith												
7.1.7	Puget Lowland							179	1.9%				
7.1.8	Coast Range							2307	24.5%				
7.1.9	Willamette Valley							10	0.1%				
8.1.1	Eastern Great Lakes Lowlands			276	2.2%	923	4.0%			358	2.5%	13	0.3%
8.1.3	Northern Allegheny Plateau			325	2.6%	2010	8.8%			80	0.6%	1	0.0%

NA_L3 CODE	US_L3NAME	baldcypress Median S=6 Assoc S-↓ N/S = 0.54		American basswood Median N=9, S=5 Assoc N-↑, S-↓ N/S = 0.4		eastern hemlock Median N=8 Assoc N-↓ N/S = 0.78		western hemlock Median N=3 Assoc N-U N/S = 0.34		American elm Median N=11, S=6 Assoc N-↑, S-↓ N/S = 0.25		slippery elm Median S=8 Assoc S-↓ N/S = 0.09	
		count	%	count	%	count	%	count	%	count	%	count	%
8.1.4	North Central Hardwood Forests			1698	13.5%	548	2.4%			1033	7.3%	131	3.2%
8.1.5	Driftless Area			969	7.7%					1512	10.6%	473	11.6%
8.1.6	Southern Michigan/Northern Indiana Drift Plains			352	2.8%	38	0.2%			566	4.0%	56	1.4%
8.1.7	Northeastern Coastal Zone			44	0.3%	1195	5.2%			119	0.8%	1	0.0%
8.1.8	Acadian Plains and Hills			44	0.3%	2657	11.6%			53	0.4%		
8.1.10	Erie Drift Plain			107	0.9%	383	1.7%			234	1.6%	45	1.1%
8.2.1	Southeastern Wisconsin Till Plains			288	2.3%	14	0.1%			396	2.8%	54	1.3%
8.2.2	Huron/Erie Lake Plains			183	1.5%	7	0.0%			305	2.1%	28	0.7%
8.2.3	Central Corn Belt Plains			40	0.3%					125	0.9%	39	1.0%
8.2.4	Eastern Corn Belt Plains			232	1.8%					474	3.3%	140	3.4%
8.3.1	Northern Piedmont			13	0.1%	9	0.0%			78	0.5%	20	0.5%
8.3.2	Interior River Valleys and Hills	7	0.2%	77	0.6%					907	6.4%	286	7.0%
8.3.3	Interior Plateau	2	0.1%	100	0.8%	1	0.0%			674	4.7%	437	10.7%
8.3.4	Piedmont	4	0.1%	10	0.1%	26	0.1%			273	1.9%	142	3.5%
8.3.5	Southeastern Plains	549	19.0%	23	0.2%	9	0.0%			371	2.6%	168	4.1%
8.3.6	Mississippi Valley Loess Plains	56	1.9%	1	0.0%					233	1.6%	127	3.1%
8.3.7	South Central Plains	458	15.8%	9	0.1%					287	2.0%	75	1.8%
8.3.8	East Central Texas Plains			4	0.0%					43	0.3%		
8.4.1	Ridge and Valley			295	2.3%	966	4.2%			166	1.2%	141	3.4%
8.4.2	Central Appalachians			556	4.4%	908	4.0%			87	0.6%	92	2.3%
8.4.3	Western Allegheny Plateau			154	1.2%	347	1.5%			659	4.6%	498	12.2%
8.4.4	Blue Ridge			209	1.7%	1018	4.5%			16	0.1%	13	0.3%

NA_L3 CODE	US_L3NAME	baldcypress Median S=6 Assoc S-↓ N/S = 0.54		American basswood Median N=9, S=5 Assoc N-↑, S-↓ N/S = 0.4		eastern hemlock Median N=8 Assoc N-↓ N/S = 0.78		western hemlock Median N=3 Assoc N-U N/S = 0.34		American elm Median N=11, S=6 Assoc N-↑, S-↓ N/S = 0.25		slippery elm Median S=8 Assoc S-↓ N/S = 0.09	
		count	%	count	%	count	%	count	%	count	%	count	%
8.4.5	Ozark Highlands			21	0.2%					687	4.8%	387	9.5%
8.4.6	Boston Mountains			20	0.2%					27	0.2%	35	0.9%
8.4.7	Arkansas Valley									37	0.3%	16	0.4%
8.4.8	Ouachita Mountains			2	0.0%					20	0.1%	13	0.3%
8.4.9	Southwestern Appalachians			54	0.4%	282	1.2%			35	0.2%	21	0.5%
8.5.1	Middle Atlantic Coastal Plain	344	11.9%	0	0.0%					107	0.8%	35	0.9%
8.5.2	Mississippi Alluvial Plain	525	18.2%	2	0.0%					449	3.2%	203	5.0%
8.5.3	Southern Coastal Plain	793	27.4%	1	0.0%					205	1.4%	14	0.3%
8.5.4	Atlantic Coastal Pine Barrens			0	0.0%							1	0.0%
9.2.1	Northern Glaciated Plains			9	0.1%					18	0.1%		
9.2.2	Lake Agassiz Plain			102	0.8%					103	0.7%	1	0.0%
9.2.3	Western Corn Belt Plains			232	1.8%					654	4.6%	190	4.6%
9.2.4	Central Irregular Plains			48	0.4%					1001	7.0%	126	3.1%
9.3.1	Northwestern Glaciated Plains			4	0.0%					47	0.3%	2	0.0%
9.3.3	Northwestern Great Plains									63	0.4%		
9.3.4	Nebraska Sand Hills									1	0.0%		
9.4.1	High Plains									7	0.0%		
9.4.2	Central Great Plains									174	1.2%	14	0.3%
9.4.3	Southwestern Tablelands									7	0.0%		
9.4.4	Flint Hills			5	0.0%					103	0.7%	10	0.2%
9.4.5	Cross Timbers									32	0.2%	3	0.1%
9.4.6	Edwards Plateau												

NA_L3 CODE	US_L3NAME	**baldcypress** Median S=6 Assoc S-↓ N/S = 0.54		**American basswood** Median N=9, S=5 Assoc N-↑, S-↓ N/S = 0.4		**eastern hemlock** Median N=8 Assoc N-↓ N/S = 0.78		**western hemlock** Median N=3 Assoc N-U N/S = 0.34		**American elm** Median N=11, S=6 Assoc N-↑, S-↓ N/S = 0.25		**slippery elm** Median S=8 Assoc S-↓ N/S = 0.09	
		count	**%**	**count**	**%**	**count**	**%**	**count**	**%**	**count**	**%**	**count**	**%**
9.4.7	Texas Blackland Prairies									2	0.0%		
9.5.1	Western Gulf Coastal Plain	16	0.6%							20	0.1%	5	0.1%
9.6.1	Southern Texas Plains												
10.1.2	Columbia Plateau												
10.1.3	Northern Basin and Range												
10.1.4	Wyoming Basin												
10.1.5	Central Basin and Range												
10.1.6	Colorado Plateaus												
10.1.7	Arizona/New Mexico Plateau												
10.1.8	Snake River Plain												
10.2.1	Mojave Basin and Range												
10.2.2	Sonoran Basin and Range												
10.2.10	Chihuahuan Deserts												
11.1.1	Southern and Central California Chaparral and Oak Woodlands												
11.1.2	Central California Valley												
11.1.3	Southern California Mountains												
12.1.1	Madrean Archipelago												
13.1.1	Arizona/New Mexico Mountains												
15.4.1	Southern Florida Coastal Plain	138	4.8%										
	Total Tree Count	2892		12587		22864		9415		14210		4087	

Attachment 2B
Species-specific Sample Distribution across Ecoregions for Species with Statistically Significant Associations of Survival with N/S from Horn et al. (2018) Supplemental Information Dataset

Key:

NA_L3 = North American Ecoregion, code for level III

US_L3NAME = Name of Ecoregion at level III

See: https://www.epa.gov/eco-research/ecoregions

Median = Tree-specific median S and/or N deposition for the species samples

Assoc = U= unimodal, ↑=positive, ↓=negative

N/S = correlation coefficient for N and S deposition values for the species samples

Count = number of species' tree samples assessed in all plots in that ecoregion

% = percent of species' tree samples in that ecoregion

NA_L3 CODE	US_L3NAME	boxelder Median S = 6 Assoc S-↓ N/S = 0.13		red maple Median N=9, S=7 Assoc N-U, S-↓ N/S = 0.59		sugar maple Median S=8 Assoc S-↓ N/S = 0.67		yellow birch Median S = 5 Assoc S-↓ N/S = 0.71		sweet birch Median N=10, S =13 Assoc N-U, S-↓ N/S = 0.57		paper birch Median N=7, S=4 Assoc N-U, S-↓ NS = 0.42	
		count	%	count	%	count	%	count	%	count	%	count	%
5.2.1	Northern Lakes and Forests	97	1.3%	26666	22.0%	31512	42.2%	3912	24.0%			12403	50.0%
5.2.2	Northern Minnesota Wetlands	61	0.8%	98	0.1%	77	0.1%	2	0.0%			657	2.6%
5.3.1	Northeastern Highlands	24	0.3%	15529	12.8%	12843	17.2%	7577	46.6%	1471	14.4%	6728	27.1%
5.3.3	North Central Appalachians			5908	4.9%	2186	2.9%	452	2.8%	1477	14.5%	122	0.5%
6.2.3	Northern Rockies											219	0.9%
6.2.4	Canadian Rockies											10	0.0%
6.2.5	North Cascades											2	0.0%
6.2.7	Cascades												
6.2.8	Eastern Cascades Slopes and Foothills												
6.2.9	Blue Mountains											4	0.0%
6.2.10	Middle Rockies											28	0.1%
6.2.11	Klamath Mountains												
6.2.12	Sierra Nevada												
6.2.13	Wasatch and Uinta Mountains												
6.2.14	Southern Rockies												
6.2.15	Idaho Batholith											12	0.0%
7.1.7	Puget Lowland											24	0.1%
7.1.8	Coast Range												
7.1.9	Willamette Valley												
8.1.1	Eastern Great Lakes Lowlands	111	1.5%	1943	1.6%	1122	1.5%	240	1.5%	56	0.5%	80	0.3%
8.1.3	Northern Allegheny Plateau	10	0.1%	4347	3.6%	3421	4.6%	466	2.9%	626	6.1%	110	0.4%
8.1.4	North Central Hardwood Forests	684	9.1%	4556	3.8%	3346	4.5%	488	3.0%			1249	5.0%

NA_L3 CODE	US_L3NAME	**boxelder** Median S = 6 Assoc S-↓ N/S = 0.13		**red maple** Median N=9, S=7 Assoc N-U, S-↓ N/S = 0.59		**sugar maple** Median S=8 Assoc S-↓ N/S = 0.67		**yellow birch** Median S = 5 Assoc S-↓ N/S = 0.71		**sweet birch** Median N=10, S =13 Assoc N-U, S-↓ N/S = 0.57		**paper birch** Median N=7, S=4 Assoc N-U, S-↓ NS = 0.42	
		count	**%**	**count**	**%**	**count**	**%**	**count**	**%**	**count**	**%**	**count**	**%**
8.1.5	Driftless Area	1095	14.6%	750	0.6%	916	1.2%	32	0.2%			891	3.6%
8.1.6	Southern Michigan/Northern Indiana Drift Plains	142	1.9%	1923	1.6%	631	0.8%	42	0.3%			39	0.2%
8.1.7	Northeastern Coastal Zone	7	0.1%	5239	4.3%	510	0.7%	300	1.8%	1095	10.7%	181	0.7%
8.1.8	Acadian Plains and Hills			5482	4.5%	1267	1.7%	1347	8.3%			1784	7.2%
8.1.10	Erie Drift Plain	8	0.1%	2203	1.8%	984	1.3%	124	0.8%	10	0.1%		
8.2.1	Southeastern Wisconsin Till Plains	404	5.4%	198	0.2%	229	0.3%	61	0.4%			72	0.3%
8.2.2	Huron/Erie Lake Plains	103	1.4%	1283	1.1%	117	0.2%	10	0.1%			157	0.6%
8.2.3	Central Corn Belt Plains	103	1.4%	17	0.0%	63	0.1%						
8.2.4	Eastern Corn Belt Plains	251	3.3%	462	0.4%	1233	1.6%	2	0.0%				
8.3.1	Northern Piedmont	92	1.2%	759	0.6%	99	0.1%	4	0.0%	144	1.4%		
8.3.2	Interior River Valleys and Hills	370	4.9%	588	0.5%	1377	1.8%						
8.3.3	Interior Plateau	600	8.0%	1367	1.1%	4029	5.4%						
8.3.4	Piedmont	195	2.6%	4099	3.4%	14	0.0%	2	0.0%	29	0.3%		
8.3.5	Southeastern Plains	207	2.8%	5825	4.8%	47	0.1%						
8.3.6	Mississippi Valley Loess Plains	252	3.4%	261	0.2%	74	0.1%						
8.3.7	South Central Plains	120	1.6%	1001	0.8%	2	0.0%						
8.3.8	East Central Texas Plains	7	0.1%	5	0.0%								
8.4.1	Ridge and Valley	131	1.7%	6002	4.9%	1707	2.3%	201	1.2%	2133	20.9%	20	0.1%
8.4.2	Central Appalachians	25	0.3%	5895	4.9%	2534	3.4%	594	3.7%	1347	13.2%		
8.4.3	Western Allegheny Plateau	249	3.3%	4725	3.9%	2779	3.7%	17	0.1%	265	2.6%		
8.4.4	Blue Ridge	23	0.3%	4545	3.7%	397	0.5%	395	2.4%	1524	14.9%		
8.4.5	Ozark Highlands	121	1.6%	219	0.2%	531	0.7%						

NA_L3 CODE	US_L3NAME	boxelder Median S = 6 Assoc S-↓ N/S = 0.13		red maple Median N=9, S=7 Assoc N-U, S-↓ N/S = 0.59		sugar maple Median S=8 Assoc S-↓ N/S = 0.67		yellow birch Median S = 5 Assoc S-↓ N/S = 0.71		sweet birch Median N=10, S =13 Assoc N-U, S-↓ N/S = 0.57		paper birch Median N=7, S=4 Assoc N-U, S-↓ NS = 0.42	
		count	%	count	%	count	%	count	%	count	%	count	%
8.4.6	Boston Mountains			223	0.2%	26	0.0%						
8.4.7	Arkansas Valley	36	0.5%	68	0.1%	1	0.0%						
8.4.8	Ouachita Mountains	5	0.1%	197	0.2%								
8.4.9	Southwestern Appalachians	27	0.4%	1760	1.5%	527	0.7%	2	0.0%	30	0.3%		
8.5.1	Middle Atlantic Coastal Plain	23	0.3%	4009	3.3%								
8.5.2	Mississippi Alluvial Plain	524	7.0%	716	0.6%	8	0.0%						
8.5.3	Southern Coastal Plain	4	0.1%	1984	1.6%								
8.5.4	Atlantic Coastal Pine Barrens	1	0.0%	325	0.3%					8	0.1%		
9.2.1	Northern Glaciated Plains	157	2.1%									6	0.0%
9.2.2	Lake Agassiz Plain	221	2.9%	3	0.0%	39						8	0.0%
9.2.3	Western Corn Belt Plains	692	9.2%	30	0.0%	78	0.1%	3	0.0%			9	0.0%
9.2.4	Central Irregular Plains	202	2.7%	2	0.0%	34	0.0%						
9.3.1	Northwestern Glaciated Plains	21	0.3%										
9.3.3	Northwestern Great Plains	14	0.2%										
9.3.4	Nebraska Sand Hills	1	0.0%										
9.4.1	High Plains	4	0.1%										
9.4.2	Central Great Plains	62	0.8%										
9.4.3	Southwestern Tablelands	4	0.1%										
9.4.4	Flint Hills	8	0.1%										
9.4.5	Cross Timbers	4	0.1%										
9.4.6	Edwards Plateau												
9.4.7	Texas Blackland Prairies												

NA_L3 CODE	US_L3NAME	**boxelder** Median S = 6 Assoc S-↓ N/S = 0.13		**red maple** Median N=9, S=7 Assoc N-U, S-↓ N/S = 0.59		**sugar maple** Median S=8 Assoc S-↓ N/S = 0.67		**yellow birch** Median S = 5 Assoc S-↓ N/S = 0.71		**sweet birch** Median N=10, S =13 Assoc N-U, S-↓ N/S = 0.57		**paper birch** Median N=7, S=4 Assoc N-U, S-↓ NS = 0.42	
		count	**%**	**count**	**%**	**count**	**%**	**count**	**%**	**count**	**%**	**count**	**%**
9.5.1	Western Gulf Coastal Plain			11	0.0%								
9.6.1	Southern Texas Plains												
10.1.2	Columbia Plateau												
10.1.3	Northern Basin and Range												
10.1.4	Wyoming Basin												
10.1.5	Central Basin and Range												
10.1.6	Colorado Plateaus												
10.1.7	Arizona/New Mexico Plateau												
10.1.8	Snake River Plain												
10.2.1	Mojave Basin and Range												
10.2.2	Sonoran Basin and Range												
10.2.10	Chihuahuan Deserts												
11.1.1	Southern and Central California Chaparral and Oak Woodlands												
11.1.2	Central California Valley	10	0.1%										
11.1.3	Southern California Mountains												
12.1.1	Madrean Archipelago												
13.1.1	Arizona/New Mexico Mountains	1	0.0%										
15.4.1	Southern Florida Coastal Plain			65	0.1%								
	Total Tree Count	7513		121288		74760		16273		10215		24815	

NA_L3 CODE	US_L3NAME	American hornbeam Median S=7 Assoc S-↓ N/S = 0.26		mockernut hickory Median N=10 Assoc N-↓ N/S = 0.15		pignut hickory Median S=10 Assoc S-↓ N/S = 0.4		hackberry Median S=7 Assoc S-↓ N/S = 0.18		American beech Median N=8, S=7 Assoc N-U, S-↓ N/S = 0.76		white ash Median N=10 Assoc N-U N/S = 0.53	
		count	%	count	%	count	%	count	%	count	%	count	%
5.2.1	Northern Lakes and Forests	14	0.4%							1565	6.4%	1415	7.0%
5.2.2	Northern Minnesota Wetlands							1	0.0%				
5.3.1	Northeastern Highlands	14	0.4%	30	0.3%	171	1.4%			9630	39.5%	2787	13.8%
5.3.3	North Central Appalachians	51	1.6%	13	0.1%	97	0.8%			1747	7.2%	541	2.7%
6.2.3	Northern Rockies												
6.2.4	Canadian Rockies												
6.2.5	North Cascades												
6.2.7	Cascades												
6.2.8	Eastern Cascades Slopes and Foothills												
6.2.9	Blue Mountains												
6.2.10	Middle Rockies							1	0.0%				
6.2.11	Klamath Mountains												
6.2.12	Sierra Nevada												
6.2.13	Wasatch and Uinta Mountains												
6.2.14	Southern Rockies												
6.2.15	Idaho Batholith												
7.1.7	Puget Lowland												
7.1.8	Coast Range												
7.1.9	Willamette Valley												
8.1.1	Eastern Great Lakes Lowlands	9	0.3%			20	0.2%	4	0.1%	324	1.3%	757	3.7%
8.1.3	Northern Allegheny Plateau	35	1.1%	9	0.1%	97	0.8%			1415	5.8%	1958	9.7%
8.1.4	North Central Hardwood Forests	6	0.2%					49	0.9%	198	0.8%	657	3.2%

NA_L3 CODE	US_L3NAME	**American hornbeam** Median S=7 Assoc S-↓ N/S = 0.26		**mockernut hickory** Median N=10 Assoc N-↓ N/S = 0.15		**pignut hickory** Median S=10 Assoc S-↓ N/S = 0.4		**hackberry** Median S=7 Assoc S-↓ N/S = 0.18		**American beech** Median N=8, S=7 Assoc N-U, S-↓ N/S = 0.76		**white ash** Median N=10 Assoc N-U N/S = 0.53	
		count	**%**	**count**	**%**	**count**	**%**	**count**	**%**	**count**	**%**	**count**	**%**
8.1.5	Driftless Area	2	0.1%					251	4.5%			399	2.0%
8.1.6	Southern Michigan/Northern Indiana Drift Plains	12	0.4%	20	0.2%	138	1.1%	39	0.7%	177	0.7%	370	1.8%
8.1.7	Northeastern Coastal Zone	6	0.2%	60	0.5%	253	2.1%	5	0.1%	375	1.5%	503	2.5%
8.1.8	Acadian Plains and Hills	0	0.0%							1672	6.9%	788	3.9%
8.1.10	Erie Drift Plain	14	0.4%	5	0.0%	23	0.2%	4	0.1%	358	1.5%	558	2.8%
8.2.1	Southeastern Wisconsin Till Plains	0	0.0%					26	0.5%	47	0.2%	196	1.0%
8.2.2	Huron/Erie Lake Plains	2	0.1%	3	0.0%	13	0.1%	14	0.3%	33	0.1%	135	0.7%
8.2.3	Central Corn Belt Plains	3	0.1%	8	0.1%	3	0.0%	89	1.6%			55	0.3%
8.2.4	Eastern Corn Belt Plains	12	0.4%	47	0.4%	115	0.9%	309	5.6%	135	0.6%	885	4.4%
8.3.1	Northern Piedmont	13	0.4%	197	1.7%	230	1.9%	32	0.6%	97	0.4%	321	1.6%
8.3.2	Interior River Valleys and Hills	17	0.5%	296	2.6%	673	5.5%	637	11.4%	155	0.6%	591	2.9%
8.3.3	Interior Plateau	95	3.0%	782	6.9%	2090	17.2%	1210	21.7%	914	3.7%	1695	8.4%
8.3.4	Piedmont	406	12.6%	1261	11.1%	1301	10.7%	80	1.4%	626	2.6%	347	1.7%
8.3.5	Southeastern Plains	879	27.3%	1163	10.2%	982	8.1%	63	1.1%	758	3.1%	129	0.6%
8.3.6	Mississippi Valley Loess Plains	253	7.9%	211	1.9%	263	2.2%	10	0.2%	134	0.5%	96	0.5%
8.3.7	South Central Plains	708	22.0%	497	4.4%	68	0.6%	9	0.2%	207	0.8%	163	0.8%
8.3.8	East Central Texas Plains	6	0.2%	29	0.3%	6	0.0%	2	0.0%			37	0.2%
8.4.1	Ridge and Valley	23	0.7%	1025	9.0%	1455	11.9%	151	2.7%	508	2.1%	1076	5.3%
8.4.2	Central Appalachians	53	1.6%	580	5.1%	786	6.5%	6	0.1%	1724	7.1%	511	2.5%
8.4.3	Western Allegheny Plateau	44	1.4%	815	7.2%	952	7.8%	79	1.4%	829	3.4%	1367	6.7%
8.4.4	Blue Ridge	23	0.7%	511	4.5%	804	6.6%	5	0.1%	353	1.4%	395	1.9%
8.4.5	Ozark Highlands	9	0.3%	1530	13.4%	664	5.4%	294	5.3%	1	0.0%	649	3.2%

NA_L3 CODE	US_L3NAME	**American hornbeam** Median S=7 Assoc S-↓ N/S = 0.26		**mockernut hickory** Median N=10 Assoc N-↓ N/S = 0.15		**pignut hickory** Median S=10 Assoc S-↓ N/S = 0.4		**hackberry** Median S=7 Assoc S-↓ N/S = 0.18		**American beech** Median N=8, S=7 Assoc N-U, S-↓ N/S = 0.76		**white ash** Median N=10 Assoc N-U N/S = 0.53	
		count	**%**	**count**	**%**	**count**	**%**	**count**	**%**	**count**	**%**	**count**	**%**
8.4.6	Boston Mountains	5	0.2%	619	5.4%	24	0.2%	24	0.4%	65	0.3%	102	0.5%
8.4.7	Arkansas Valley	14	0.4%	288	2.5%	1	0.0%	40	0.7%			79	0.4%
8.4.8	Ouachita Mountains	37	1.2%	565	5.0%	2	0.0%	6	0.1%	9	0.0%	29	0.1%
8.4.9	Southwestern Appalachians	38	1.2%	536	4.7%	754	6.2%	35	0.6%	199	0.8%	240	1.2%
8.5.1	Middle Atlantic Coastal Plain	170	5.3%	110	1.0%	58	0.5%	26	0.5%	109	0.4%	41	0.2%
8.5.2	Mississippi Alluvial Plain	39	1.2%	85	0.7%	29	0.2%	71	1.3%	11	0.0%	9	0.0%
8.5.3	Southern Coastal Plain	186	5.8%	14	0.1%	96	0.8%	10	0.2%	3	0.0%	16	0.1%
8.5.4	Atlantic Coastal Pine Barrens	3	0.1%	4	0.0%	6	0.0%	0	0.0%	19	0.1%	7	0.0%
9.2.1	Northern Glaciated Plains							17	0.3%				
9.2.2	Lake Agassiz Plain												
9.2.3	Western Corn Belt Plains			2	0.0%	2	0.0%	651	11.7%			104	0.5%
9.2.4	Central Irregular Plains			69	0.6%	7	0.1%	855	15.4%			243	1.2%
9.3.1	Northwestern Glaciated Plains							16	0.3%				
9.3.3	Northwestern Great Plains							3	0.1%				
9.3.4	Nebraska Sand Hills							7	0.1%				
9.4.1	High Plains							2	0.0%				
9.4.2	Central Great Plains							260	4.7%			2	0.0%
9.4.3	Southwestern Tablelands							11	0.2%				
9.4.4	Flint Hills							151	2.7%			2	0.0%
9.4.5	Cross Timbers			5	0.0%	1	0.0%	10	0.2%			8	0.0%
9.4.6	Edwards Plateau												
9.4.7	Texas Blackland Prairies			1	0.0%							3	0.0%

NA_L3 CODE	US_L3NAME	**American hornbeam** Median S=7 Assoc S-↓ N/S = 0.26		**mockernut hickory** Median N=10 Assoc N-↓ N/S = 0.15		**pignut hickory** Median S=10 Assoc S-↓ N/S = 0.4		**hackberry** Median S=7 Assoc S-↓ N/S = 0.18		**American beech** Median N=8, S=7 Assoc N-U, S-↓ N/S = 0.76		**white ash** Median N=10 Assoc N-U N/S = 0.53	
		count	**%**	**count**	**%**	**count**	**%**	**count**	**%**	**count**	**%**	**count**	**%**
9.5.1	Western Gulf Coastal Plain	13	0.4%	2	0.0%	1	0.0%						
9.6.1	Southern Texas Plains												
10.1.2	Columbia Plateau												
10.1.3	Northern Basin and Range												
10.1.4	Wyoming Basin												
10.1.5	Central Basin and Range												
10.1.6	Colorado Plateaus												
10.1.7	Arizona/New Mexico Plateau												
10.1.8	Snake River Plain												
10.2.1	Mojave Basin and Range												
10.2.2	Sonoran Basin and Range												
10.2.10	Chihuahuan Deserts												
11.1.1	Southern and Central California Chaparral and Oak Woodlands												
11.1.2	Central California Valley												
11.1.3	Southern California Mountains												
12.1.1	Madrean Archipelago												
13.1.1	Arizona/New Mexico Mountains												
15.4.1	Southern Florida Coastal Plain												
	Total Tree Count	3214		11392		12185		5565		24397		20266	

NA_L3 CODE	US_L3NAME	green ash Median N=10,S=6 Assoc N-U, S-↓ N/S = 0.45		black walnut Median N=12 Assoc N-U N/S = 0.13		Utah juniper Median N=3 Assoc N-U N/S = 0.72		eastern redcedar Median N=11,S=7 Assoc N-U,S-↓ N/S Cor r= 0.3		sweetgum Median N=9, S=7 Assoc N-U, S-↓ N/S = 0.37		yellow poplar Median S=11 Assoc S-↓ N/S = 0.4	
		count	%	count	%	count	%	count	%	count	%	count	%
5.2.1	Northern Lakes and Forests	1941	10.3%	4	0.1%			2	0.0%				
5.2.2	Northern Minnesota Wetlands	216	1.1%										
5.3.1	Northeastern Highlands	45	0.2%	6	0.1%			28	0.2%			112	0.4%
5.3.3	North Central Appalachians			1	0.0%			4	0.0%			57	0.2%
6.2.3	Northern Rockies												
6.2.4	Canadian Rockies												
6.2.5	North Cascades												
6.2.7	Cascades												
6.2.8	Eastern Cascades Slopes and Foothills												
6.2.9	Blue Mountains												
6.2.10	Middle Rockies	25	0.1%			60	0.3%						
6.2.11	Klamath Mountains												
6.2.12	Sierra Nevada												
6.2.13	Wasatch and Uinta Mountains					1089	5.8%						
6.2.14	Southern Rockies					219	1.2%						
6.2.15	Idaho Batholith												
7.1.7	Puget Lowland												
7.1.8	Coast Range												
7.1.9	Willamette Valley												
8.1.1	Eastern Great Lakes Lowlands	546	2.9%	40	0.6%			48	0.3%			9	0.0%

NA_L3 CODE	US_L3NAME	green ash Median N=10,S=6 Assoc N-U, S-↓ N/S = 0.45		black walnut Median N=12 Assoc N-U N/S = 0.13		Utah juniper Median N=3 Assoc N-U N/S = 0.72		eastern redcedar Median N=11,S=7 Assoc N-U,S-↓ N/S Cor r= 0.3		sweetgum Median N=9, S=7 Assoc N-U, S-↓ N/S = 0.37		yellow poplar Median S=11 Assoc S-↓ N/S = 0.4	
		count	%	count	%	count	%	count	%	count	%	count	%
8.1.3	Northern Allegheny Plateau	99	0.5%	40	0.6%			21	0.1%			7	0.0%
8.1.4	North Central Hardwood Forests	1561	8.3%	13	0.2%			123	0.7%				
8.1.5	Driftless Area	217	1.2%	448	6.8%			313	1.8%				
8.1.6	Southern Michigan/Northern Indiana Drift Plains	1128	6.0%	145	2.2%			27	0.2%			71	0.3%
8.1.7	Northeastern Coastal Zone	62	0.3%	23	0.3%			172	1.0%	2	0.0%	58	0.2%
8.1.8	Acadian Plains and Hills	31	0.2%										
8.1.10	Erie Drift Plain	162	0.9%	63	1.0%					1	0.0%	184	0.7%
8.2.1	Southeastern Wisconsin Till Plains	735	3.9%	88	1.3%			102	0.6%				
8.2.2	Huron/Erie Lake Plains	813	4.3%	42	0.6%			1	0.0%			4	0.0%
8.2.3	Central Corn Belt Plains	126	0.7%	148	2.2%			8	0.0%			6	0.0%
8.2.4	Eastern Corn Belt Plains	359	1.9%	469	7.1%			161	0.9%	84	0.2%	206	0.7%
8.3.1	Northern Piedmont	61	0.3%	171	2.6%			277	1.6%	63	0.2%	804	2.9%
8.3.2	Interior River Valleys and Hills	666	3.5%	514	7.8%			738	4.3%	569	1.5%	436	1.6%
8.3.3	Interior Plateau	904	4.8%	931	14.1%			3994	23.2%	1003	2.7%	2609	9.4%
8.3.4	Piedmont	621	3.3%	192	2.9%			1329	7.7%	6822	18.3%	5776	20.9%
8.3.5	Southeastern Plains	1302	6.9%	66	1.0%			873	5.1%	11773	31.6%	4012	14.5%
8.3.6	Mississippi Valley Loess Plains	322	1.7%	29	0.4%			208	1.2%	2090	5.6%	327	1.2%
8.3.7	South Central Plains	711	3.8%	24	0.4%			249	1.4%	6197	16.7%	13	0.0%
8.3.8	East Central Texas Plains	191	1.0%	4	0.1%			173	1.0%	275	0.7%		
8.4.1	Ridge and Valley	262	1.4%	414	6.3%			883	5.1%	690	1.9%	1880	6.8%
8.4.2	Central Appalachians	52	0.3%	64	1.0%			47	0.3%	118	0.3%	3411	12.3%
8.4.3	Western Allegheny Plateau	134	0.7%	454	6.9%			63	0.4%	15	0.0%	2714	9.8%

NA_L3 CODE	US_L3NAME	green ash Median N=10,S=6 Assoc N-U, S-↓ N/S = 0.45		black walnut Median N=12 Assoc N-U N/S = 0.13		Utah juniper Median N=3 Assoc N-U N/S = 0.72		eastern redcedar Median N=11,S=7 Assoc N-U,S-↓ N/S Cor r= 0.3		sweetgum Median N=9, S=7 Assoc N-U, S-↓ N/S = 0.37		yellow poplar Median S=11 Assoc S-↓ N/S = 0.4	
		count	%	count	%	count	%	count	%	count	%	count	%
8.4.4	Blue Ridge	61	0.3%	82	1.2%			40	0.2%	85	0.2%	3203	11.6%
8.4.5	Ozark Highlands	176	0.9%	810	12.3%			3903	22.6%	168	0.5%	4	0.0%
8.4.6	Boston Mountains	21	0.1%	35	0.5%			333	1.9%	218	0.6%		
8.4.7	Arkansas Valley	166	0.9%	10	0.2%			727	4.2%	268	0.7%		
8.4.8	Ouachita Mountains	101	0.5%	5	0.1%			260	1.5%	386	1.0%		
8.4.9	Southwestern Appalachians	154	0.8%	49	0.7%			415	2.4%	752	2.0%	1154	4.2%
8.5.1	Middle Atlantic Coastal Plain	530	2.8%	14	0.2%			21	0.1%	3130	8.4%	551	2.0%
8.5.2	Mississippi Alluvial Plain	1109	5.9%	9	0.1%			22	0.1%	877	2.4%	19	0.1%
8.5.3	Southern Coastal Plain	630	3.3%					43	0.2%	1422	3.8%	55	0.2%
8.5.4	Atlantic Coastal Pine Barrens	1	0.0%	3	0.0%			16	0.1%	85	0.2%	19	0.1%
9.2.1	Northern Glaciated Plains	383	2.0%					1	0.0%				
9.2.2	Lake Agassiz Plain	280	1.5%										
9.2.3	Western Corn Belt Plains	502	2.7%	381	5.8%			384	2.2%				
9.2.4	Central Irregular Plains	408	2.2%	701	10.6%			435	2.5%				
9.3.1	Northwestern Glaciated Plains	96	0.5%					130	0.8%				
9.3.3	Northwestern Great Plains	417	2.2%	1	0.0%			95	0.6%				
9.3.4	Nebraska Sand Hills	29	0.2%					104	0.6%				
9.4.1	High Plains	27	0.1%	1	0.0%			33	0.2%				
9.4.2	Central Great Plains	337	1.8%	27	0.4%			319	1.8%				
9.4.3	Southwestern Tablelands	12	0.1%	7	0.1%			8	0.0%				
9.4.4	Flint Hills	50	0.3%	50	0.8%			73	0.4%				
9.4.5	Cross Timbers	25	0.1%	13	0.2%			19	0.1%				

NA_L3 CODE	US_L3NAME	green ash Median N=10,S=6 Assoc N-U, S-↓ N/S = 0.45		black walnut Median N=12 Assoc N-U N/S = 0.13		Utah juniper Median N=3 Assoc N-U N/S = 0.72		eastern redcedar Median N=11,S=7 Assoc N-U,S-↓ N/S Cor r= 0.3		sweetgum Median N=9, S=7 Assoc N-U, S-↓ N/S = 0.37		yellow poplar Median S=11 Assoc S-↓ N/S = 0.4	
		count	%	count	%	count	%	count	%	count	%	count	%
9.4.6	Edwards Plateau												
9.4.7	Texas Blackland Prairies	1	0.0%					13	0.1%				
9.5.1	Western Gulf Coastal Plain	39	0.2%					11	0.1%	118	0.3%		
9.6.1	Southern Texas Plains												
10.1.2	Columbia Plateau												
10.1.3	Northern Basin and Range					605	3.2%						
10.1.4	Wyoming Basin					189	1.0%						
10.1.5	Central Basin and Range					4944	26.5%						
10.1.6	Colorado Plateaus					7696	41.2%						
10.1.7	Arizona/New Mexico Plateau					2211	11.8%						
10.1.8	Snake River Plain												
10.2.1	Mojave Basin and Range					207	1.1%						
10.2.2	Sonoran Basin and Range					2	0.0%						
10.2.10	Chihuahuan Deserts												
11.1.1	Southern and Central California Chaparral and Oak Woodlands												
11.1.2	Central California Valley												
11.1.3	Southern California Mountains												
12.1.1	Madrean Archipelago					2	0.0%						
13.1.1	Arizona/New Mexico Mountains					1457	7.8%						
15.4.1	Southern Florida Coastal Plain	7	0.0%										
	Total Tree Count	18854		6591		18681		17249		37211		27701	

NA_L3 CODE	US_L3NAME	**sweetbay** Median S=7 Assoc S-↓ N/S = 0.35		**swamp tupelo** Median S=6 Assoc S-↓ N/S = 0.46		**blackgum** Median N=10 Assoc N-U N/S = 0.42		**eastern hophornbeam** Median S=6 Assoc S-↓ N/S = 0.36		**sourwood** Median N=9, S=10 Assoc N-U, S-↓ N/S = 0.3		**shortleaf pine** Median S=6 Assoc S-↓ N/S = 0.18	
		count	**%**	**count**	**%**	**count**	**%**	**count**	**%**	**count**	**%**	**count**	**%**
5.2.1	Northern Lakes and Forests					2	0.0%	1051	17.8%				
5.2.2	Northern Minnesota Wetlands							5	0.1%				
5.3.1	Northeastern Highlands					29	0.2%	703	11.9%				
5.3.3	North Central Appalachians					171	1.3%	116	2.0%				
6.2.3	Northern Rockies												
6.2.4	Canadian Rockies												
6.2.5	North Cascades												
6.2.7	Cascades												
6.2.8	Eastern Cascades Slopes and Foothills												
6.2.9	Blue Mountains												
6.2.10	Middle Rockies							3	0.1%				
6.2.11	Klamath Mountains												
6.2.12	Sierra Nevada												
6.2.13	Wasatch and Uinta Mountains												
6.2.14	Southern Rockies												
6.2.15	Idaho Batholith												
7.1.7	Puget Lowland												
7.1.8	Coast Range												
7.1.9	Willamette Valley												
8.1.1	Eastern Great Lakes Lowlands					32	0.2%	241	4.1%				
8.1.3	Northern Allegheny Plateau					16	0.1%	432	7.3%				

NA_L3 CODE	US_L3NAME	sweetbay Median S=7 Assoc S-↓ N/S = 0.35		swamp tupelo Median S=6 Assoc S-↓ N/S = 0.46		blackgum Median N=10 Assoc N-U N/S = 0.42		eastern hophornbeam Median S=6 Assoc S-↓ N/S = 0.36		sourwood Median N=9, S=10 Assoc N-U, S-↓ N/S = 0.3		shortleaf pine Median S=6 Assoc S-↓ N/S = 0.18	
		count	%	count	%	count	%	count	%	count	%	count	%
8.1.4	North Central Hardwood Forests							560	9.5%				
8.1.5	Driftless Area							454	7.7%				
8.1.6	Southern Michigan/Northern Indiana Drift Plains					43	0.3%	68	1.2%				
8.1.7	Northeastern Coastal Zone					121	0.9%	34	0.6%				
8.1.8	Acadian Plains and Hills							231	3.9%				
8.1.10	Erie Drift Plain					72	0.5%	84	1.4%	1	0.0%		
8.2.1	Southeastern Wisconsin Till Plains							34	0.6%				
8.2.2	Huron/Erie Lake Plains					37	0.3%	18	0.3%				
8.2.3	Central Corn Belt Plains					40	0.3%	13	0.2%				
8.2.4	Eastern Corn Belt Plains					47	0.3%	78	1.3%				
8.3.1	Northern Piedmont					179	1.3%	4	0.1%	6	0.1%	8	0.0%
8.3.2	Interior River Valleys and Hills					214	1.6%	83	1.4%	26	0.3%	27	0.2%
8.3.3	Interior Plateau			3	0.0%	885	6.6%	201	3.4%	673	7.5%	276	1.6%
8.3.4	Piedmont	42	1.0%	79	0.7%	957	7.1%	85	1.4%	1953	21.8%	2761	16.2%
8.3.5	Southeastern Plains	2345	55.8%	4949	44.5%	2415	17.9%	239	4.0%	678	7.6%	1941	11.4%
8.3.6	Mississippi Valley Loess Plains	12	0.3%	10	0.1%	164	1.2%	176	3.0%	51	0.6%	141	0.8%
8.3.7	South Central Plains	262	6.2%	78	0.7%	1232	9.2%	309	5.2%	6	0.1%	1891	11.1%
8.3.8	East Central Texas Plains					42	0.3%					30	0.2%
8.4.1	Ridge and Valley	3	0.1%	1	0.0%	1668	12.4%	121	2.0%	804	9.0%	383	2.2%
8.4.2	Central Appalachians					606	4.5%	44	0.7%	845	9.4%	47	0.3%
8.4.3	Western Allegheny Plateau					586	4.4%	100	1.7%	504	5.6%	65	0.4%
8.4.4	Blue Ridge					1029	7.6%	50	0.8%	2363	26.4%	376	2.2%

NA_L3 CODE	US_L3NAME	**sweetbay** Median S=7 Assoc S-↓ N/S = 0.35		**swamp tupelo** Median S=6 Assoc S-↓ N/S = 0.46		**blackgum** Median N=10 Assoc N-U N/S = 0.42		**eastern hophornbeam** Median S=6 Assoc S-↓ N/S = 0.36		**sourwood** Median N=9, S=10 Assoc N-U, S-↓ N/S = 0.3		**shortleaf pine** Median S=6 Assoc S-↓ N/S = 0.18	
		count	**%**	**count**	**%**	**count**	**%**	**count**	**%**	**count**	**%**	**count**	**%**
8.4.5	Ozark Highlands					660	4.9%	12	0.2%			2867	16.8%
8.4.6	Boston Mountains					333	2.5%	10	0.2%			679	4.0%
8.4.7	Arkansas Valley			5	0.0%	112	0.8%	10	0.2%			1259	7.4%
8.4.8	Ouachita Mountains					288	2.1%	43	0.7%			3804	22.3%
8.4.9	Southwestern Appalachians					547	4.1%	36	0.6%	989	11.1%	381	2.2%
8.5.1	Middle Atlantic Coastal Plain	225	5.4%	2181	19.6%	506	3.8%	11	0.2%	43	0.5%	31	0.2%
8.5.2	Mississippi Alluvial Plain			65	0.6%	33	0.2%	13	0.2%	3	0.0%	16	0.1%
8.5.3	Southern Coastal Plain	1293	30.8%	3738	33.6%	278	2.1%	17	0.3%	1	0.0%	2	0.0%
8.5.4	Atlantic Coastal Pine Barrens	10	0.2%			99	0.7%					40	0.2%
9.2.1	Northern Glaciated Plains							10	0.2%				
9.2.2	Lake Agassiz Plain							11	0.2%				
9.2.3	Western Corn Belt Plains							160	2.7%				
9.2.4	Central Irregular Plains							36	0.6%				
9.3.1	Northwestern Glaciated Plains							2	0.0%				
9.3.3	Northwestern Great Plains							2	0.0%				
9.3.4	Nebraska Sand Hills												
9.4.1	High Plains												
9.4.2	Central Great Plains												
9.4.3	Southwestern Tablelands												
9.4.4	Flint Hills							1	0.0%				
9.4.5	Cross Timbers											3	0.0%
9.4.6	Edwards Plateau												

NA_L3 CODE	US_L3NAME	**sweetbay** Median S=7 Assoc S-↓ N/S = 0.35		**swamp tupelo** Median S=6 Assoc S-↓ N/S = 0.46		**blackgum** Median N=10 Assoc N-U N/S = 0.42		**eastern hophornbeam** Median S=6 Assoc S-↓ N/S = 0.36		**sourwood** Median N=9, S=10 Assoc N-U, S-↓ N/S = 0.3		**shortleaf pine** Median S=6 Assoc S-↓ N/S = 0.18	
		count	**%**	**count**	**%**	**count**	**%**	**count**	**%**	**count**	**%**	**count**	**%**
9.4.7	Texas Blackland Prairies												
9.5.1	Western Gulf Coastal Plain	1	0.0%			21	0.2%	1	0.0%				
9.6.1	Southern Texas Plains												
10.1.2	Columbia Plateau												
10.1.3	Northern Basin and Range												
10.1.4	Wyoming Basin												
10.1.5	Central Basin and Range												
10.1.6	Colorado Plateaus												
10.1.7	Arizona/New Mexico Plateau												
10.1.8	Snake River Plain												
10.2.1	Mojave Basin and Range												
10.2.2	Sonoran Basin and Range												
10.2.10	Chihuahuan Deserts												
11.1.1	Southern and Central California Chaparral and Oak Woodlands												
11.1.2	Central California Valley												
11.1.3	Southern California Mountains												
12.1.1	Madrean Archipelago												
13.1.1	Arizona/New Mexico Mountains												
15.4.1	Southern Florida Coastal Plain	6	0.1%	1	0.0%								
	Total Tree Count	4199		11110		13464		5912		8946		17028	

NA_L3 CODE	US_L3NAME	slash pine Median N=7, S=5 Assoc N-U, S-↓ N/S = 0.44		longleaf pine Median S=7 Assoc S-↓ N/S = 0.44		red pine Median N=8, S=5 Assoc N-U, S-↓ N/S = 0.54		pitch pine Median N=10,S=12 Assoc N-U,S-↓ N/S = 0.65		eastern white pine Median S=6 Assoc S-↓ N/S = 0.6		loblolly pine Median S=7 Assoc S-↓ N/S = 0.32	
		count	%	Count	%	count	%	count	%	count	%	count	%
5.2.1	Northern Lakes and Forests					6593	65.0%			4340	18.4%		
5.2.2	Northern Minnesota Wetlands					208	2.1%			38	0.2%		
5.3.1	Northeastern Highlands					189	1.9%	88	2.8%	4501	19.1%		
5.3.3	North Central Appalachians					44	0.4%	47	1.5%	566	2.4%		
6.2.3	Northern Rockies												
6.2.4	Canadian Rockies												
6.2.5	North Cascades												
6.2.7	Cascades												
6.2.8	Eastern Cascades Slopes and Foothills												
6.2.9	Blue Mountains												
6.2.10	Middle Rockies												
6.2.11	Klamath Mountains												
6.2.12	Sierra Nevada												
6.2.13	Wasatch and Uinta Mountains												
6.2.14	Southern Rockies												
6.2.15	Idaho Batholith												
7.1.7	Puget Lowland												
7.1.8	Coast Range												
7.1.9	Willamette Valley												
8.1.1	Eastern Great Lakes Lowlands					79	0.8%	7	0.2%	602	2.6%		
8.1.3	Northern Allegheny Plateau					155	1.5%	3	0.1%	955	4.1%		

NA_L3 CODE	US_L3NAME	slash pine Median N=7, S=5 Assoc N-U, S-↓ N/S = 0.44		longleaf pine Median S=7 Assoc S-↓ N/S = 0.44		red pine Median N=8, S=5 Assoc N-U, S-↓ N/S = 0.54		pitch pine Median N=10,S=12 Assoc N-U,S-↓ N/S = 0.65		eastern white pine Median S=6 Assoc S-↓ N/S = 0.6		loblolly pine Median S=7 Assoc S-↓ N/S = 0.32	
		count	%	Count	%	count	%	count	%	count	%	count	%
8.1.4	North Central Hardwood Forests					1609	15.9%			1751	7.4%		
8.1.5	Driftless Area					346	3.4%			436	1.9%		
8.1.6	Southern Michigan/Northern Indiana Drift Plains					466	4.6%	2	0.1%	362	1.5%		
8.1.7	Northeastern Coastal Zone					13	0.1%	106	3.4%	2711	11.5%		
8.1.8	Acadian Plains and Hills					163	1.6%	5	0.2%	2353	10.0%		
8.1.10	Erie Drift Plain					20	0.2%			33	0.1%		
8.2.1	Southeastern Wisconsin Till Plains					85	0.8%			140	0.6%		
8.2.2	Huron/Erie Lake Plains					60	0.6%			128	0.5%		
8.2.3	Central Corn Belt Plains					19	0.2%			29	0.1%		
8.2.4	Eastern Corn Belt Plains					13	0.1%			47	0.2%		
8.3.1	Northern Piedmont									69	0.3%	33	0.0%
8.3.2	Interior River Valleys and Hills					29	0.3%	5	0.2%	16	0.1%	40	0.1%
8.3.3	Interior Plateau					2	0.0%	7	0.2%	106	0.4%	578	0.8%
8.3.4	Piedmont	29	0.2%	224	4.2%			25	0.8%	401	1.7%	15095	21.8%
8.3.5	Southeastern Plains	4035	33.9%	3108	57.8%			1	0.0%	1	0.0%	23675	34.2%
8.3.6	Mississippi Valley Loess Plains	3	0.0%	1	0.0%							1599	2.3%
8.3.7	South Central Plains	235	2.0%	311	5.8%							13971	20.2%
8.3.8	East Central Texas Plains	37	0.3%									151	0.2%
8.4.1	Ridge and Valley			100	1.9%	7	0.1%	472	14.9%	1308	5.6%	1494	2.2%
8.4.2	Central Appalachians					13	0.1%	80	2.5%	228	1.0%	4	0.0%
8.4.3	Western Allegheny Plateau					24	0.2%	107	3.4%	391	1.7%	100	0.1%
8.4.4	Blue Ridge							312	9.9%	1817	7.7%	304	0.4%

NA_L3 CODE	US_L3NAME	slash pine Median N=7, S=5 Assoc N-U, S-↓ N/S = 0.44		longleaf pine Median S=7 Assoc S-↓ N/S = 0.44		red pine Median N=8, S=5 Assoc N-U, S-↓ N/S = 0.54		pitch pine Median N=10,S=12 Assoc N-U,S-↓ N/S = 0.65		eastern white pine Median S=6 Assoc S-↓ N/S = 0.6		loblolly pine Median S=7 Assoc S-↓ N/S = 0.32	
		count	%	Count	%	count	%	count	%	count	%	count	%
8.4.5	Ozark Highlands											21	0.0%
8.4.6	Boston Mountains											50	0.1%
8.4.7	Arkansas Valley											205	0.3%
8.4.8	Ouachita Mountains											715	1.0%
8.4.9	Southwestern Appalachians			9	0.2%			27	0.9%	130	0.6%	1486	2.1%
8.5.1	Middle Atlantic Coastal Plain	120	1.0%	353	6.6%			42	1.3%			7035	10.1%
8.5.2	Mississippi Alluvial Plain											103	0.1%
8.5.3	Southern Coastal Plain	7160	60.2%	1258	23.4%							2279	3.3%
8.5.4	Atlantic Coastal Pine Barrens							1827	57.8%	103	0.4%	2	0.0%
9.2.1	Northern Glaciated Plains												
9.2.2	Lake Agassiz Plain												
9.2.3	Western Corn Belt Plains												
9.2.4	Central Irregular Plains					2	0.0%						
9.3.1	Northwestern Glaciated Plains												
9.3.3	Northwestern Great Plains												
9.3.4	Nebraska Sand Hills												
9.4.1	High Plains												
9.4.2	Central Great Plains												
9.4.3	Southwestern Tablelands												
9.4.4	Flint Hills												
9.4.5	Cross Timbers												
9.4.6	Edwards Plateau												

NA_L3 CODE	US_L3NAME	**slash pine** Median N=7, S=5 Assoc N-U, S-↓ N/S = 0.44		**longleaf pine** Median S=7 Assoc S-↓ N/S = 0.44		**red pine** Median N=8, S=5 Assoc N-U, S-↓ N/S = 0.54		**pitch pine** Median N=10,S=12 Assoc N-U,S-↓ N/S = 0.65		**eastern white pine** Median S=6 Assoc S-↓ N/S = 0.6		**loblolly pine** Median S=7 Assoc S-↓ N/S = 0.32	
		count	**%**	**Count**	**%**	**count**	**%**	**count**	**%**	**count**	**%**	**count**	**%**
9.4.7	Texas Blackland Prairies											75	0.1%
9.5.1	Western Gulf Coastal Plain			9	0.2%							306	0.4%
9.6.1	Southern Texas Plains												
10.1.2	Columbia Plateau												
10.1.3	Northern Basin and Range												
10.1.4	Wyoming Basin												
10.1.5	Central Basin and Range												
10.1.6	Colorado Plateaus												
10.1.7	Arizona/New Mexico Plateau												
10.1.8	Snake River Plain												
10.2.1	Mojave Basin and Range												
10.2.2	Sonoran Basin and Range												
10.2.10	Chihuahuan Deserts												
11.1.1	Southern and Central California Chaparral and Oak Woodlands												
11.1.2	Central California Valley												
11.1.3	Southern California Mountains												
12.1.1	Madrean Archipelago												
13.1.1	Arizona/New Mexico Mountains												
15.4.1	Southern Florida Coastal Plain	283	2.4%										
	Total Tree Count	11902		5373		10139		3163		23562		69321	

NA_L3 CODE	US_L3NAME	**Virginia pine** Median N=10,S=11 Assoc N-U, S-↓ N/S = 0.44		**bigtooth aspen** Median N=9, S=6 Assoc N-U, S-↓ N/S = 0.57		**quaking aspen** Median N=7, S=3 Assoc N-U, S-↓ N/S = 0.61		**black cherry** Median N=11 Assoc N-U N/S = 0.33		**Douglas-fir** Median N=3 Assoc N-U N/S = 0.65		**white oak** Median N=10, S=8 Assoc N-U, S-↓ N/S = 0.17	
		count	**%**	**count**	**%**	**count**	**%**	**count**	**%**	**count**	**%**	**count**	**%**
5.2.1	Northern Lakes and Forests			6784	58.8%	27494	52.9%	1989	8.1%			1615	3.4%
5.2.2	Northern Minnesota Wetlands			13	0.1%	2995	5.8%						
5.3.1	Northeastern Highlands			496	4.3%	1273	2.5%	1539	6.3%			243	0.5%
5.3.3	North Central Appalachians			115	1.0%	167	0.3%	1516	6.2%			1032	2.2%
6.2.3	Northern Rockies					67	0.1%			4463	9.4%		
6.2.4	Canadian Rockies					92	0.2%			800	1.7%		
6.2.5	North Cascades									2702	5.7%		
6.2.7	Cascades									10680	22.5%		
6.2.8	Eastern Cascades Slopes and Foothills					28	0.1%			1631	3.4%		
6.2.9	Blue Mountains					7	0.0%			3271	6.9%		
6.2.10	Middle Rockies					398	0.8%			3908	8.2%		
6.2.11	Klamath Mountains					3	0.0%			6885	14.5%		
6.2.12	Sierra Nevada					30	0.1%			1066	2.2%		
6.2.13	Wasatch and Uinta Mountains					3024	5.8%			774	1.6%		
6.2.14	Southern Rockies					5182	10.0%			2161	4.6%		
6.2.15	Idaho Batholith					15	0.0%			1699	3.6%		
7.1.7	Puget Lowland									409	0.9%		
7.1.8	Coast Range									4823	10.2%		
7.1.9	Willamette Valley									184	0.4%		
8.1.1	Eastern Great Lakes Lowlands			88	0.8%	384	0.7%	408	1.7%			27	0.1%
8.1.3	Northern Allegheny Plateau			166	1.4%	428	0.8%	867	3.5%	9	0.0%	180	0.4%

NA_L3 CODE	US_L3NAME	**Virginia pine** Median N=10,S=11 Assoc N-U, S-↓ N/S = 0.44		**bigtooth aspen** Median N=9, S=6 Assoc N-U, S-↓ N/S = 0.57		**quaking aspen** Median N=7, S=3 Assoc N-U, S-↓ N/S = 0.61		**black cherry** Median N=11 Assoc N-U N/S = 0.33		**Douglas-fir** Median N=3 Assoc N-U N/S = 0.65		**white oak** Median N=10, S=8 Assoc N-U, S-↓ N/S = 0.17	
		count	**%**	**count**	**%**	**count**	**%**	**count**	**%**	**count**	**%**	**count**	**%**
8.1.4	North Central Hardwood Forests			1026	8.9%	4024	7.7%	692	2.8%			1060	2.3%
8.1.5	Driftless Area			540	4.7%	488	0.9%	894	3.7%			1247	2.7%
8.1.6	Southern Michigan/Northern Indiana Drift Plains			396	3.4%	344	0.7%	1508	6.2%	1	0.0%	749	1.6%
8.1.7	Northeastern Coastal Zone			157	1.4%	184	0.4%	312	1.3%			817	1.7%
8.1.8	Acadian Plains and Hills			612	5.3%	1183	2.3%	169	0.7%			17	0.0%
8.1.10	Erie Drift Plain			105	0.9%	202	0.4%	879	3.6%			74	0.2%
8.2.1	Southeastern Wisconsin Till Plains			30	0.3%	296	0.6%	490	2.0%			164	0.3%
8.2.2	Huron/Erie Lake Plains			200	1.7%	421	0.8%	154	0.6%			97	0.2%
8.2.3	Central Corn Belt Plains	1	0.0%			21	0.0%	350	1.4%			128	0.3%
8.2.4	Eastern Corn Belt Plains	3	0.0%	21	0.2%	8	0.0%	603	2.5%			177	0.4%
8.3.1	Northern Piedmont	238	2.6%	18	0.2%	6	0.0%	283	1.2%	9	0.0%	328	0.7%
8.3.2	Interior River Valleys and Hills	78	0.8%	9	0.1%			578	2.4%			1594	3.4%
8.3.3	Interior Plateau	339	3.6%	30	0.3%			976	4.0%			3240	6.9%
8.3.4	Piedmont	3590	38.5%	9	0.1%			970	4.0%			4634	9.9%
8.3.5	Southeastern Plains	412	4.4%	7	0.1%			1163	4.7%			2853	6.1%
8.3.6	Mississippi Valley Loess Plains							258	1.1%			448	1.0%
8.3.7	South Central Plains	1	0.0%					157	0.6%			1221	2.6%
8.3.8	East Central Texas Plains							4	0.0%				
8.4.1	Ridge and Valley	1653	17.7%	111	1.0%	24	0.0%	1398	5.7%	1	0.0%	2923	6.2%
8.4.2	Central Appalachians	212	2.3%	101	0.9%	27	0.1%	1396	5.7%			1976	4.2%
8.4.3	Western Allegheny Plateau	574	6.2%	505	4.4%	109	0.2%	2712	11.1%			2442	5.2%
8.4.4	Blue Ridge	946	10.1%	1	0.0%			417	1.7%			1265	2.7%

NA_L3 CODE	US_L3NAME	Virginia pine Median N=10,S=11 Assoc N-U, S-↓ N/S = 0.44		bigtooth aspen Median N=9, S=6 Assoc N-U, S-↓ N/S = 0.57		quaking aspen Median N=7, S=3 Assoc N-U, S-↓ N/S = 0.61		black cherry Median N=11 Assoc N-U N/S = 0.33		Douglas-fir Median N=3 Assoc N-U N/S = 0.65		white oak Median N=10, S=8 Assoc N-U, S-↓ N/S = 0.17	
		count	%	count	%	count	%	count	%	count	%	count	%
8.4.5	Ozark Highlands	3	0.0%					448	1.8%			8989	19.2%
8.4.6	Boston Mountains							96	0.4%			1619	3.5%
8.4.7	Arkansas Valley							73	0.3%			487	1.0%
8.4.8	Ouachita Mountains							106	0.4%			1268	2.7%
8.4.9	Southwestern Appalachians	1155	12.4%					260	1.1%			2152	4.6%
8.5.1	Middle Atlantic Coastal Plain	83	0.9%	2	0.0%			250	1.0%			458	1.0%
8.5.2	Mississippi Alluvial Plain							40	0.2%			83	0.2%
8.5.3	Southern Coastal Plain	14	0.2%					85	0.3%			13	0.0%
8.5.4	Atlantic Coastal Pine Barrens	22	0.2%	3	0.0%			40	0.2%			519	1.1%
9.2.1	Northern Glaciated Plains					377	0.7%						
9.2.2	Lake Agassiz Plain			1	0.0%	1773	3.4%	3	0.0%				
9.2.3	Western Corn Belt Plains			1	0.0%	86	0.2%	210	0.9%			112	0.2%
9.2.4	Central Irregular Plains							192	0.8%			676	1.4%
9.3.1	Northwestern Glaciated Plains					10	0.0%			12	0.0%		
9.3.3	Northwestern Great Plains					30	0.1%	1	0.0%	260	0.5%		
9.3.4	Nebraska Sand Hills												
9.4.1	High Plains												
9.4.2	Central Great Plains												
9.4.3	Southwestern Tablelands												
9.4.4	Flint Hills												
9.4.5	Cross Timbers							2	0.0%				
9.4.6	Edwards Plateau												

NA_L3 CODE	US_L3NAME	**Virginia pine** Median N=10,S=11 Assoc N-U, S-↓ N/S = 0.44		**bigtooth aspen** Median N=9, S=6 Assoc N-U, S-↓ N/S = 0.57		**quaking aspen** Median N=7, S=3 Assoc N-U, S-↓ N/S = 0.61		**black cherry** Median N=11 Assoc N-U N/S = 0.33		**Douglas-fir** Median N=3 Assoc N-U N/S = 0.65		**white oak** Median N=10, S=8 Assoc N-U, S-↓ N/S = 0.17	
		count	**%**	**count**	**%**	**count**	**%**	**count**	**%**	**count**	**%**	**count**	**%**
9.4.7	Texas Blackland Prairies												
9.5.1	Western Gulf Coastal Plain							5	0.0%				
9.6.1	Southern Texas Plains												
10.1.2	Columbia Plateau					37	0.1%			174	0.4%		
10.1.3	Northern Basin and Range					112	0.2%			105	0.2%		
10.1.4	Wyoming Basin									27	0.1%		
10.1.5	Central Basin and Range					62	0.1%			26	0.1%		
10.1.6	Colorado Plateaus					282	0.5%			403	0.8%		
10.1.7	Arizona/New Mexico Plateau					3	0.0%			17	0.0%		
10.1.8	Snake River Plain					32	0.1%						
10.2.1	Mojave Basin and Range												
10.2.2	Sonoran Basin and Range												
10.2.10	Chihuahuan Deserts												
11.1.1	Southern and Central California Chaparral and Oak Woodlands									222	0.5%		
11.1.2	Central California Valley												
11.1.3	Southern California Mountains												
12.1.1	Madrean Archipelago									38	0.1%		
13.1.1	Arizona/New Mexico Mountains					218	0.4%			657	1.4%		
15.4.1	Southern Florida Coastal Plain												
	Total Tree Count	9324		11547		51946		24493		47417		46927	

NA_L3 CODE	US_L3NAME	**scarlet oak** Median N=10,S=10 Assoc N-↓, S-↓ N/S = 0.37		**southern red oak** Median N=9 Assoc N-↓ N/S = 0.36		**laurel oak** Median S=6 Assoc S-↓ N/S = 0.41		**chinkapin oak** Median N=11 Assoc N-U N/S = 0.31		**water oak** Median N=8, S=9 Assoc N-↓, S-↓ N/S = 0.26		**chestnut oak** Median S=12 Assoc S-↓ N/S = 0.44	
		count	**%**	**count**	**%**	**count**	**%**	**count**	**%**	**count**	**%**	**count**	**%**
5.2.1	Northern Lakes and Forests												
5.2.2	Northern Minnesota Wetlands												
5.3.1	Northeastern Highlands	112	1.1%									386	1.6%
5.3.3	North Central Appalachians	236	2.2%									1155	4.9%
6.2.3	Northern Rockies												
6.2.4	Canadian Rockies												
6.2.5	North Cascades												
6.2.7	Cascades												
6.2.8	Eastern Cascades Slopes and Foothills												
6.2.9	Blue Mountains												
6.2.10	Middle Rockies												
6.2.11	Klamath Mountains												
6.2.12	Sierra Nevada												
6.2.13	Wasatch and Uinta Mountains												
6.2.14	Southern Rockies												
6.2.15	Idaho Batholith												
7.1.7	Puget Lowland												
7.1.8	Coast Range							2	0.1%				
7.1.9	Willamette Valley												
8.1.1	Eastern Great Lakes Lowlands											3	0.0%
8.1.3	Northern Allegheny Plateau	21	0.2%									229	1.0%
8.1.4	North Central Hardwood Forests												

NA_L3 CODE	US_L3NAME	**scarlet oak** Median N=10,S=10 Assoc N-↓, S-↓ N/S = 0.37		**southern red oak** Median N=9 Assoc N-↓ N/S = 0.36		**laurel oak** Median S=6 Assoc S-↓ N/S = 0.41		**chinkapin oak** Median N=11 Assoc N-U N/S = 0.31		**water oak** Median N=8, S=9 Assoc N-↓, S-↓ N/S = 0.26		**chestnut oak** Median S=12 Assoc S-↓ N/S = 0.44	
		count	**%**	**count**	**%**	**count**	**%**	**count**	**%**	**count**	**%**	**count**	**%**
8.1.5	Driftless Area							17	0.6%				
8.1.6	Southern Michigan/Northern Indiana Drift Plains	7	0.1%					3	0.1%				
8.1.7	Northeastern Coastal Zone	636	6.0%					4	0.1%			187	0.8%
8.1.8	Acadian Plains and Hills												
8.1.10	Erie Drift Plain	5	0.0%									5	0.0%
8.2.1	Southeastern Wisconsin Till Plains												
8.2.2	Huron/Erie Lake Plains	5	0.0%					4	0.1%				
8.2.3	Central Corn Belt Plains							9	0.3%				
8.2.4	Eastern Corn Belt Plains	5	0.0%					104	3.4%			6	0.0%
8.3.1	Northern Piedmont	79	0.7%	90	1.0%							400	1.7%
8.3.2	Interior River Valleys and Hills	72	0.7%	57	0.6%			275	8.9%			86	0.4%
8.3.3	Interior Plateau	612	5.8%	498	5.6%	1	0.0%	1002	32.4%	70	0.5%	1183	5.1%
8.3.4	Piedmont	961	9.0%	1646	18.6%	52	0.9%	2	0.1%	1546	10.6%	1313	5.6%
8.3.5	Southeastern Plains	326	3.1%	2263	25.6%	2724	46.9%	28	0.9%	6454	44.3%	269	1.1%
8.3.6	Mississippi Valley Loess Plains	14	0.1%	278	3.1%	29	0.5%	22	0.7%	605	4.2%		
8.3.7	South Central Plains	1	0.0%	1597	18.0%	341	5.9%	11	0.4%	2446	16.8%		
8.3.8	East Central Texas Plains			181	2.0%	3	0.1%			203	1.4%		
8.4.1	Ridge and Valley	1707	16.0%	385	4.3%	17	0.3%	124	4.0%	149	1.0%	7903	33.7%
8.4.2	Central Appalachians	874	8.2%	34	0.4%			44	1.4%			2496	10.7%
8.4.3	Western Allegheny Plateau	586	5.5%	11	0.1%			53	1.7%			1305	5.6%
8.4.4	Blue Ridge	1749	16.4%	254	2.9%			5	0.2%	22	0.2%	4800	20.5%
8.4.5	Ozark Highlands	1508	14.2%	538	6.1%	2	0.0%	848	27.5%	1	0.0%		

NA_L3 CODE	US_L3NAME	scarlet oak Median N=10,S=10 Assoc N-↓, S-↓ N/S = 0.37		southern red oak Median N=9 Assoc N-↓ N/S = 0.36		laurel oak Median S=6 Assoc S-↓ N/S = 0.41		chinkapin oak Median N=11 Assoc N-U N/S = 0.31		water oak Median N=8, S=9 Assoc N-↓, S-↓ N/S = 0.26		chestnut oak Median S=12 Assoc S-↓ N/S = 0.44	
		count	%	count	%	count	%	count	%	count	%	count	%
8.4.6	Boston Mountains			81	0.9%			75	2.4%				
8.4.7	Arkansas Valley			156	1.8%	2	0.0%	3	0.1%	140	1.0%		
8.4.8	Ouachita Mountains			130	1.5%			5	0.2%	76	0.5%		
8.4.9	Southwestern Appalachians	586	5.5%	305	3.4%	2	0.0%	253	8.2%	82	0.6%	1590	6.8%
8.5.1	Middle Atlantic Coastal Plain	102	1.0%	211	2.4%	544	9.4%			1111	7.6%	25	0.1%
8.5.2	Mississippi Alluvial Plain	1	0.0%	75	0.8%	20	0.3%	8	0.3%	286	2.0%		
8.5.3	Southern Coastal Plain	1	0.0%	20	0.2%	2003	34.5%	2	0.1%	1233	8.5%		
8.5.4	Atlantic Coastal Pine Barrens	432	4.1%	22	0.2%							84	0.4%
9.2.1	Northern Glaciated Plains												
9.2.2	Lake Agassiz Plain												
9.2.3	Western Corn Belt Plains							47	1.5%				
9.2.4	Central Irregular Plains	2	0.0%	1	0.0%			96	3.1%				
9.3.1	Northwestern Glaciated Plains												
9.3.3	Northwestern Great Plains												
9.3.4	Nebraska Sand Hills												
9.4.1	High Plains												
9.4.2	Central Great Plains							1	0.0%				
9.4.3	Southwestern Tablelands												
9.4.4	Flint Hills							34	1.1%				
9.4.5	Cross Timbers							7	0.2%				
9.4.6	Edwards Plateau												
9.4.7	Texas Blackland Prairies			1	0.0%					3	0.0%		

NA_L3 CODE	US_L3NAME	**scarlet oak** Median N=10,S=10 Assoc N-↓, S-↓ N/S = 0.37		**southern red oak** Median N=9 Assoc N-↓ N/S = 0.36		**laurel oak** Median S=6 Assoc S-↓ N/S = 0.41		**chinkapin oak** Median N=11 Assoc N-U N/S = 0.31		**water oak** Median N=8, S=9 Assoc N-↓, S-↓ N/S = 0.26		**chestnut oak** Median S=12 Assoc S-↓ N/S = 0.44	
		count	**%**	**count**	**%**	**count**	**%**	**count**	**%**	**count**	**%**	**count**	**%**
9.5.1	Western Gulf Coastal Plain			21	0.2%	29	0.5%			139	1.0%		
9.6.1	Southern Texas Plains												
10.1.2	Columbia Plateau												
10.1.3	Northern Basin and Range												
10.1.4	Wyoming Basin												
10.1.5	Central Basin and Range												
10.1.6	Colorado Plateaus												
10.1.7	Arizona/New Mexico Plateau												
10.1.8	Snake River Plain												
10.2.1	Mojave Basin and Range												
10.2.2	Sonoran Basin and Range												
10.2.10	Chihuahuan Deserts												
11.1.1	Southern and Central California Chaparral and Oak Woodlands												
11.1.2	Central California Valley												
11.1.3	Southern California Mountains												
12.1.1	Madrean Archipelago												
13.1.1	Arizona/New Mexico Mountains							1	0.0%				
15.4.1	Southern Florida Coastal Plain					44	0.8%						
	Total Tree Count	10640		8855		5813		3089		14566		23425	

NA_L3 CODE	US_L3NAME	**northern red oak** Median N=10 Assoc N-U N/S = 0.41		**post oak** Median 10 Assoc N-U N/S = 0.14		**black oak** Median S=8 Assoc S-↓ N/S = 0.15		**black locust** Median N=11,S=12 Assoc N-↑,S-↓ N/S = 0.19		**sassafras** Median S=12 Assoc S-↓ N/S = 0.3		**baldcypress** Median S=6 Assoc S-↓ N/S = 0.55	
		count	**%**	**count**	**%**	**count**	**%**	**count**	**%**	**count**	**%**	**count**	**%**
5.2.1	Northern Lakes and Forests	6123	19.3%			743	3.4%	28	0.5%	25	0.4%		
5.2.2	Northern Minnesota Wetlands	2	0.0%										
5.3.1	Northeastern Highlands	3162	10.0%			173	0.8%	34	0.6%	31	0.5%		
5.3.3	North Central Appalachians	1104	3.5%			174	0.8%	12	0.2%	244	3.9%		
6.2.3	Northern Rockies												
6.2.4	Canadian Rockies												
6.2.5	North Cascades												
6.2.7	Cascades												
6.2.8	Eastern Cascades Slopes and Foothills												
6.2.9	Blue Mountains												
6.2.10	Middle Rockies												
6.2.11	Klamath Mountains												
6.2.12	Sierra Nevada												
6.2.13	Wasatch and Uinta Mountains												
6.2.14	Southern Rockies												
6.2.15	Idaho Batholith												
7.1.7	Puget Lowland												
7.1.8	Coast Range												
7.1.9	Willamette Valley												
8.1.1	Eastern Great Lakes Lowlands	258	0.8%			24	0.1%	30	0.5%	2	0.0%		
8.1.3	Northern Allegheny Plateau	886	2.8%			123	0.6%	65	1.2%	9	0.1%		
8.1.4	North Central Hardwood Forests	1604	5.1%			988	4.5%	143	2.6%				

NA_L3 CODE	US_L3NAME	northern red oak Median N=10 Assoc N-U N/S = 0.41		post oak Median 10 Assoc N-U N/S = 0.14		black oak Median S=8 Assoc S-↓ N/S = 0.15		black locust Median N=11,S=12 Assoc N-↑,S-↓ N/S = 0.19		sassafras Median S=12 Assoc S-↓ N/S = 0.3		baldcypress Median S=6 Assoc S-↓ N/S = 0.55	
		count	%	count	%	count	%	count	%	count	%	count	%
8.1.5	Driftless Area	1673	5.3%			748	3.4%	128	2.3%				
8.1.6	Southern Michigan/Northern Indiana Drift Plains	724	2.3%			820	3.7%	137	2.5%	491	7.8%		
8.1.7	Northeastern Coastal Zone	1761	5.6%			911	4.2%	80	1.4%	46	0.7%		
8.1.8	Acadian Plains and Hills	868	2.7%			9	0.0%						
8.1.10	Erie Drift Plain	233	0.7%			60	0.3%	88	1.6%	60	1.0%		
8.2.1	Southeastern Wisconsin Till Plains	157	0.5%			64	0.3%	108	2.0%				
8.2.2	Huron/Erie Lake Plains	194	0.6%			25	0.1%	7	0.1%	63	1.0%		
8.2.3	Central Corn Belt Plains	73	0.2%			104	0.5%	93	1.7%	43	0.7%		
8.2.4	Eastern Corn Belt Plains	250	0.8%	3	0.0%	107	0.5%	184	3.3%	148	2.4%		
8.3.1	Northern Piedmont	192	0.6%	10	0.0%	179	0.8%	103	1.9%	91	1.4%		
8.3.2	Interior River Valleys and Hills	536	1.7%	551	2.7%	819	3.7%	189	3.4%	600	9.6%	7	0.2%
8.3.3	Interior Plateau	793	2.5%	687	3.4%	1053	4.8%	520	9.4%	1078	17.2%	6	0.1%
8.3.4	Piedmont	901	2.8%	991	4.9%	773	3.5%	120	2.2%	68	1.1%	4	0.1%
8.3.5	Southeastern Plains	200	0.6%	1416	7.0%	445	2.0%	45	0.8%	141	2.2%	712	17.1%
8.3.6	Mississippi Valley Loess Plains	44	0.1%	172	0.8%	81	0.4%	51	0.9%	119	1.9%	88	2.1%
8.3.7	South Central Plains	7	0.0%	1673	8.3%	86	0.4%	14	0.3%	112	1.8%	643	15.4%
8.3.8	East Central Texas Plains			934	4.6%	4	0.0%	3	0.1%	19	0.3%		
8.4.1	Ridge and Valley	2603	8.2%	299	1.5%	1593	7.3%	826	14.9%	642	10.2%		
8.4.2	Central Appalachians	1336	4.2%	26	0.1%	678	3.1%	603	10.9%	524	8.3%		
8.4.3	Western Allegheny Plateau	952	3.0%	46	0.2%	1009	4.6%	605	10.9%	836	13.3%	1	0.0%
8.4.4	Blue Ridge	1439	4.5%	88	0.4%	593	2.7%	752	13.6%	218	3.5%		
8.4.5	Ozark Highlands	1437	4.5%	6909	34.1%	7233	33.0%	31	0.6%	334	5.3%		

NA_L3 CODE	US_L3NAME	**northern red oak** Median N=10 Assoc N-U N/S = 0.41		**post oak** Median 10 Assoc N-U N/S = 0.14		**black oak** Median S=8 Assoc S-↓ N/S = 0.15		**black locust** Median N=11,S=12 Assoc N-↑,S-↓ N/S = 0.19		**sassafras** Median S=12 Assoc S-↓ N/S = 0.3		**baldcypress** Median S=6 Assoc S-↓ N/S = 0.55	
		count	**%**	**count**	**%**	**count**	**%**	**count**	**%**	**count**	**%**	**count**	**%**
8.4.6	Boston Mountains	741	2.3%	996	4.9%	527	2.4%	78	1.4%	36	0.6%		
8.4.7	Arkansas Valley	188	0.6%	2263	11.2%	151	0.7%	6	0.1%	4	0.1%		
8.4.8	Ouachita Mountains	451	1.4%	1902	9.4%	225	1.0%	5	0.1%	2	0.0%		
8.4.9	Southwestern Appalachians	327	1.0%	276	1.4%	547	2.5%	70	1.3%	140	2.2%		
8.5.1	Middle Atlantic Coastal Plain	26	0.1%	63	0.3%	73	0.3%	29	0.5%	61	1.0%	456	11.0%
8.5.2	Mississippi Alluvial Plain	8	0.0%	153	0.8%	13	0.1%	18	0.3%	17	0.3%	985	23.7%
8.5.3	Southern Coastal Plain			39	0.2%	2	0.0%			1	0.0%	1044	25.1%
8.5.4	Atlantic Coastal Pine Barrens	16	0.1%	31	0.2%	280	1.3%	10	0.2%	67	1.1%		
9.2.1	Northern Glaciated Plains			0	0.0%								
9.2.2	Lake Agassiz Plain	2	0.0%	0	0.0%								
9.2.3	Western Corn Belt Plains	192	0.6%	7	0.0%	84	0.4%	69	1.2%				
9.2.4	Central Irregular Plains	220	0.7%	337	1.7%	336	1.5%	196	3.5%	4	0.1%		
9.3.1	Northwestern Glaciated Plains												
9.3.3	Northwestern Great Plains												
9.3.4	Nebraska Sand Hills												
9.4.1	High Plains												
9.4.2	Central Great Plains							49	0.9%				
9.4.3	Southwestern Tablelands							3	0.1%				
9.4.4	Flint Hills	4	0.0%	3	0.0%								
9.4.5	Cross Timbers	2	0.0%	372	1.8%	56	0.3%			2	0.0%		
9.4.6	Edwards Plateau			0	0.0%								
9.4.7	Texas Blackland Prairies			18	0.1%								

NA_L3 CODE	US_L3NAME	**northern red oak** Median N=10 Assoc N-U N/S = 0.41		**post oak** Median 10 Assoc N-U N/S = 0.14		**black oak** Median S=8 Assoc S-↓ N/S = 0.15		**black locust** Median N=11,S=12 Assoc N-↑,S-↓ N/S = 0.19		**sassafras** Median S=12 Assoc S-↓ N/S = 0.3		**baldcypress** Median S=6 Assoc S-↓ N/S = 0.55	
		count	**%**	**count**	**%**	**count**	**%**	**count**	**%**	**count**	**%**	**count**	**%**
9.5.1	Western Gulf Coastal Plain			12	0.1%	1	0.0%	1	0.0%			29	0.7%
9.6.1	Southern Texas Plains												
10.1.2	Columbia Plateau												
10.1.3	Northern Basin and Range												
10.1.4	Wyoming Basin												
10.1.5	Central Basin and Range												
10.1.6	Colorado Plateaus												
10.1.7	Arizona/New Mexico Plateau												
10.1.8	Snake River Plain												
10.2.1	Mojave Basin and Range												
10.2.2	Sonoran Basin and Range												
10.2.10	Chihuahuan Deserts												
11.1.1	Southern and Central California Chaparral and Oak Woodlands												
11.1.2	Central California Valley												
11.1.3	Southern California Mountains												
12.1.1	Madrean Archipelago												
13.1.1	Arizona/New Mexico Mountains												
15.4.1	Southern Florida Coastal Plain											187	4.5%
	Total Tree Count	31689		20277		21914		5533		6278		4162	

NA_L3 CODE	US_L3NAME	**American basswood** Median S=5 Assoc S-↓ N/S = 0.39		**eastern hemlock** Median N=8 Assoc N-U N/S = 0.78		**winged elm** Median N=10 Assoc N-↓ N/S = 0.37		**American elm** Median N=11,S=6 Assoc N-↓,S-↓ N/S = 0.24		**slippery elm** Median N=11, S=8 Assoc N-U, S-↓ N/S = 0.07			
		count	**%**	**count**	**%**	**count**	**%**	**count**	**%**	**count**	**%**		
5.2.1	Northern Lakes and Forests	6603	43.4%	3441	13.4%			1340	7.0%	29	0.5%		
5.2.2	Northern Minnesota Wetlands	155	1.0%					124	0.6%				
5.3.1	Northeastern Highlands	238	1.6%	7956	31.0%			233	1.2%	3	0.1%		
5.3.3	North Central Appalachians	192	1.3%	1416	5.5%			23	0.1%	4	0.1%		
6.2.3	Northern Rockies												
6.2.4	Canadian Rockies												
6.2.5	North Cascades												
6.2.7	Cascades												
6.2.8	Eastern Cascades Slopes and Foothills												
6.2.9	Blue Mountains												
6.2.10	Middle Rockies							2	0.0%				
6.2.11	Klamath Mountains												
6.2.12	Sierra Nevada												
6.2.13	Wasatch and Uinta Mountains												
6.2.14	Southern Rockies												
6.2.15	Idaho Batholith												
7.1.7	Puget Lowland												
7.1.8	Coast Range												
7.1.9	Willamette Valley												
8.1.1	Eastern Great Lakes Lowlands	343	2.3%	1010	3.9%			508	2.7%	16	0.3%		
8.1.3	Northern Allegheny Plateau	398	2.6%	2257	8.8%			136	0.7%	1	0.0%		
8.1.4	North Central Hardwood Forests	2072	13.6%	598	2.3%			1420	7.4%	171	3.1%		

NA_L3 CODE	US_L3NAME	American basswood Median S=5 Assoc S-↓ N/S = 0.39		eastern hemlock Median N=8 Assoc N-U N/S = 0.78		winged elm Median N=10 Assoc N-↓ N/S = 0.37		American elm Median N=11,S=6 Assoc N-↓,S-↓ N/S = 0.24		slippery elm Median N=11, S=8 Assoc N-U, S-↓ N/S = 0.07			
		count	%	count	%	count	%	count	%	count	%		
8.1.5	Driftless Area	1198	7.9%	0	0.0%			2130	11.1%	653	11.9%		
8.1.6	Southern Michigan/Northern Indiana Drift Plains	437	2.9%	39	0.2%			895	4.7%	107	1.9%		
8.1.7	Northeastern Coastal Zone	54	0.4%	1322	5.1%			169	0.9%	2	0.0%		
8.1.8	Acadian Plains and Hills	51	0.3%	2848	11.1%			67	0.4%				
8.1.10	Erie Drift Plain	129	0.8%	423	1.6%			355	1.9%	64	1.2%		
8.2.1	Southeastern Wisconsin Till Plains	333	2.2%	14	0.1%			591	3.1%	86	1.6%		
8.2.2	Huron/Erie Lake Plains	214	1.4%	7	0.0%	4	0.1%	444	2.3%	44	0.8%		
8.2.3	Central Corn Belt Plains	50	0.3%					167	0.9%	67	1.2%		
8.2.4	Eastern Corn Belt Plains	278	1.8%					612	3.2%	200	3.6%		
8.3.1	Northern Piedmont	15	0.1%	10	0.0%			117	0.6%	33	0.6%		
8.3.2	Interior River Valleys and Hills	100	0.7%			156	2.3%	1168	6.1%	393	7.1%		
8.3.3	Interior Plateau	126	0.8%	1	0.0%	718	10.6%	834	4.4%	572	10.4%		
8.3.4	Piedmont	13	0.1%	29	0.1%	1071	15.8%	344	1.8%	182	3.3%		
8.3.5	Southeastern Plains	32	0.2%	12	0.0%	763	11.3%	496	2.6%	211	3.8%		
8.3.6	Mississippi Valley Loess Plains	2	0.0%			524	7.8%	311	1.6%	166	3.0%		
8.3.7	South Central Plains	11	0.1%			1437	21.3%	391	2.0%	103	1.9%		
8.3.8	East Central Texas Plains	5	0.0%			258	3.8%	58	0.3%				
8.4.1	Ridge and Valley	367	2.4%	1269	4.9%	143	2.1%	235	1.2%	177	3.2%		
8.4.2	Central Appalachians	742	4.9%	1032	4.0%	8	0.1%	110	0.6%	128	2.3%		
8.4.3	Western Allegheny Plateau	189	1.2%	385	1.5%	2	0.0%	869	4.5%	655	11.9%		
8.4.4	Blue Ridge	251	1.6%	1305	5.1%	7	0.1%	24	0.1%	21	0.4%		
8.4.5	Ozark Highlands	33	0.2%			457	6.8%	821	4.3%	468	8.5%		

NA_L3 CODE	US_L3NAME	**American basswood** Median S=5 Assoc S-↓ N/S = 0.39		**eastern hemlock** Median N=8 Assoc N-U N/S = 0.78		**winged elm** Median N=10 Assoc N-↓ N/S = 0.37		**American elm** Median N=11,S=6 Assoc N-↓,S-↓ N/S = 0.24		**slippery elm** Median N=11, S=8 Assoc N-U, S-↓ N/S = 0.07			
		count	**%**	**count**	**%**	**count**	**%**	**count**	**%**	**count**	**%**		
8.4.6	Boston Mountains	21	0.1%			115	1.7%	36	0.2%	43	0.8%		
8.4.7	Arkansas Valley	2	0.0%			310	4.6%	62	0.3%	24	0.4%		
8.4.8	Ouachita Mountains	3	0.0%			327	4.8%	28	0.1%	15	0.3%		
8.4.9	Southwestern Appalachians	66	0.4%	302	1.2%	94	1.4%	43	0.2%	29	0.5%		
8.5.1	Middle Atlantic Coastal Plain					20	0.3%	139	0.7%	49	0.9%		
8.5.2	Mississippi Alluvial Plain	3	0.0%			223	3.3%	579	3.0%	255	4.6%		
8.5.3	Southern Coastal Plain	1	0.0%			24	0.4%	266	1.4%	20	0.4%		
8.5.4	Atlantic Coastal Pine Barrens									1	0.0%		
9.2.1	Northern Glaciated Plains	13	0.1%					30	0.2%				
9.2.2	Lake Agassiz Plain	122	0.8%					139	0.7%	1	0.0%		
9.2.3	Western Corn Belt Plains	300	2.0%					960	5.0%	290	5.3%		
9.2.4	Central Irregular Plains	54	0.4%			27	0.4%	1250	6.5%	172	3.1%		
9.3.1	Northwestern Glaciated Plains	4	0.0%					65	0.3%	2	0.0%		
9.3.3	Northwestern Great Plains							69	0.4%				
9.3.4	Nebraska Sand Hills							1	0.0%				
9.4.1	High Plains							9	0.0%				
9.4.2	Central Great Plains							230	1.2%	16	0.3%		
9.4.3	Southwestern Tablelands							10	0.1%	0	0.0%		
9.4.4	Flint Hills	5	0.0%					129	0.7%	13	0.2%		
9.4.5	Cross Timbers					47	0.7%	36	0.2%	5	0.1%		
9.4.6	Edwards Plateau												
9.4.7	Texas Blackland Prairies					5	0.1%	3	0.0%				

NA_L3 CODE	US_L3NAME	**American basswood** Median S=5 Assoc S-↓ N/S = 0.39		**eastern hemlock** Median N=8 Assoc N-U N/S = 0.78		**winged elm** Median N=10 Assoc N-↓ N/S = 0.37		**American elm** Median N=11,S=6 Assoc N-↓,S-↓ N/S = 0.24		**slippery elm** Median N=11, S=8 Assoc N-U, S-↓ N/S = 0.07			
		count	**%**	**count**	**%**	**count**	**%**	**count**	**%**	**count**	**%**		
9.5.1	Western Gulf Coastal Plain					20	0.3%	29	0.2%	6	0.1%		
9.6.1	Southern Texas Plains												
10.1.2	Columbia Plateau												
10.1.3	Northern Basin and Range												
10.1.4	Wyoming Basin												
10.1.5	Central Basin and Range												
10.1.6	Colorado Plateaus												
10.1.7	Arizona/New Mexico Plateau												
10.1.8	Snake River Plain												
10.2.1	Mojave Basin and Range												
10.2.2	Sonoran Basin and Range												
10.2.10	Chihuahuan Deserts												
11.1.1	Southern and Central California Chaparral and Oak Woodlands												
11.1.2	Central California Valley												
11.1.3	Southern California Mountains												
12.1.1	Madrean Archipelago												
13.1.1	Arizona/New Mexico Mountains												
15.4.1	Southern Florida Coastal Plain												
	Total Tree Count	15225		25676		6760		19107		5497			

APPENDIX 6A

DERIVATION OF THE ECOREGION AIR QUALITY METRICS

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Maps Showing Monitor Sites of Influence for NO_2, $PM_{2.5}$, and SO_2 (3-hour metric) Based on Different Inclusion Criteria for 16 Example Ecoregions

6A.1. INTRODUCTION

In order to better understand the relationship between air quality concentrations and downwind nitrogen (N) and sulfur (S) deposition, as described in Chapter 6, we conducted air parcel trajectory modeling, using the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model,[1] to help identify the meteorological patterns that determine the transport of pollutant material from source to receptor. Using ambient air quality monitoring sites as trajectory starting points, we estimated potential "sites of influence" for each of the 84 Level III ecoregions in the contiguous U.S. The "sites of influence" are used to identify upwind geographic areas from which emissions potentially contribute to N and S deposition in each ecoregion. The air quality design values (DV)[2] for each ecoregion's set of "sites of influence" were then used to estimate an Ecoregion Air Quality Metric (EAQM). The EAQM values were calculated for each ecoregion and for three separate pollutants: NO_2, SO_2, and $PM_{2.5}$. Further, we derived two sets of EAQM values for SO_2, one reflecting the DV for the current 3-hour secondary standard (averaged over three years) and a second for an annual average metric (averaged over three years). The EAQM values provide a perspective of air quality levels in the upwind regions that could potentially contribute to downwind deposition levels.

This Appendix describes the methodology used to calculate the air parcel trajectories that identified sites of influence (6A.2), the methodology used to estimate the EAQM values for each ecoregion-pollutant metric combination using DVs, or design value-like metrics, based on historical air quality data (6A.3), as well as the method by which EAQM values and TDep estimates were linked (6A.4). The appendix then briefly summarizes the results of a series of sensitivity analyses on several aspects of the EAQM methodology (6A.5). Finally, all of the plots and tables generated within this analysis are provided (6A.6).

6A.2. HYSPLIT TRAJECTORY METHODOLOGY

The HYSPLIT model is commonly used to compute simple air parcel trajectories using historical meteorological data. HYSPLIT can simulate the trajectory of air parcels as they are

[1] Stein, A.F., Draxler, R.R, Rolph, G.D., Stunder, B.J.B., Cohen, M.D., and Ngan, F., (2015). NOAA's HYSPLIT atmospheric transport and dispersion modeling system, Bull. Amer. Meteor. Soc., 96, 2059-2077, http://dx.doi.org/10.1175/BAMS-D-14-00110.1.

[2] A design value is a statistic that summarizes the air quality data for a given area in terms of the indicator, averaging time, and form of the standard. Design values can be compared to the level of the standard and are typically used to designate areas as meeting or not meeting the standard and assess progress towards meeting the NAAQS. Design values are computed and published annually by EPA (https://www.epa.gov/air-trends/air-quality-design-values). It should be noted that not all of the air quality metrics considered here are existing NAAQS. In those cases we are using the term "design value" as a proxy for design value-like metrics of the air quality data at a location.

transported through the atmosphere for a given set of meteorological conditions. One common application of HYSPLIT is to apply the model in a forward-trajectory mode to evaluate the transport of hypothetical emissions releases from a specific origin. When trajectories are calculated over a large number of time periods with representative meteorological conditions, one can develop a potential zone of influence, or "footprint," for any emissions source from a specific location. In this exercise, HYSPLIT was used to estimate the frequency at which simulated air transport trajectories from individual monitoring sites could plausibly have impacted a downwind ecoregion. In this way, the upwind monitor sites of influence for each of the 84 ecoregions in the contiguous U.S. were established, indicating the areas where emissions potentially contribute to deposition in a downwind ecoregion.

In a subsequent step, we investigated relationships between pollutant concentrations at the upwind sites of influence and N or S deposition estimates for each of the ecoregions. The air quality metrics utilized for each of the pollutants in this step included DVs for existing standards, or a DV-like metric for other considered pollutant/averaging time combinations. As explained in more detail below, multiple HYSPLIT trajectories were generated and analyzed to determine potential sites of influence for each region, and then all of the valid data from those monitors were assessed to generate an EAQM for multiple ecoregion-pollutant metric pairs.

Two sets of trajectories were generated, an original analysis and a final analysis. The basic configurations of the two sets of HYSPLIT simulations were as follows:

- Forward trajectories (i.e., where the air will go from its source)
- Trajectory origin: monitor sites with any valid DV (or DV-like metric) for the pollutant (in the 2000-2020 period)
- Trajectory length: 48-hour (original analysis), 120-hour (final analysis)
- Trajectory start time: 1800 GMT (i.e., intended to be mid-day)
- Trajectories per site-day: 1
- Trajectory start height: 500 meters
- Trajectory output tracking: every 10 minutes
- Meteorological year: 2016
- Meteorological data:
    - Original analysis: 32-km North American Regional Reanalysis (NARR-32)[3];

[3] National Centers for Environmental Prediction/National Weather Service/NOAA/U.S. Department of Commerce. 2005, updated monthly. NCEP North American Regional Reanalysis (NARR). Research Data Archive at the National Center for Atmospheric Research, Computational and Information Systems Laboratory. https://rda.ucar.edu/datasets/ds608.0/. Accessed 25 May 2017.

- Final analysis: 12-km North American Mesoscale Forecast System (NAM-12)[4]

In all, 568,398 individual trajectories were generated. There are several assumptions built into this HYSPLIT application that kept the exercise manageable, but that may also influence the outcome. We discuss the rationale for this particular configuration below and introduce sensitivity analyses conducted for several aspects of this application.

This analysis used a single year of meteorology in 2016. While no single year can be considered truly representative of all possible wind trajectories and their frequency at any given location, we note that 2016 marked the transition from a strongly positive Oceanic Niño Index (ONI) to a weakly negative one by the end of the year[5]. Using meteorology from a year that captures each phase of the ONI is presumed to be more likely to represent a broader variety of wind and transport patterns than a year which captures only a single phase. To compare 2016 to other meteorological years, Figure 6A-1 illustrates how the average maximum temperatures across the U.S. in 2016 ranked across the entire 1895-2016 climatological period. The 2016 annual maximum temperatures were higher than the climatological average in the longer record, due to the changing climate in recent decades, but the difference is generally consistent across the country (i.e., most areas are "much above average"). Figure 6A-2 shows how annual precipitation amounts in 2016 compared to the longer-term climatological average. There were parts of the northeast and southeastern U.S. where 2016 was an anomalously dry year, while other parts of the U.S. were much wetter than average (e.g., WI, MN, ND). However, a large part of the U.S. experienced relatively normal precipitation levels in 2016. Based on this cursory evaluation of the meteorological conditions in 2016, we see no evidence that suggests this year of meteorology would yield unrepresentative trajectory patterns. We also note that a single meteorological year, and specifically the year 2016, has also been used in EPA regulatory actions where transport patterns are evaluated to assess how upwind emissions may impact downwind areas (e.g., the 2015 Ozone NAAQS Good Neighbor Plan final rulemaking[6]). We recognize that the use of a single meteorological year may add uncertainty to the identification of monitoring

[4] National Centers for Environmental Prediction, National Weather Service, NOAA, U.S. Department of Commerce. 2015, updated daily. North American Mesoscale Forecast System (NAM). Research Data Archive at the National Center for Atmospheric Research, Computational and Information Systems Laboratory. https://www.ncei.noaa.gov/data/north-american-mesoscale-model/access. Accessed 3 July 2023

[5] National Weather Service, Climate Prediction Center. https://origin.cpc.ncep.noaa.gov/products/analysis_monitoring/ensostuff/ONI_v5.php. Accessed 06 October 2023.

[6] USEPA, Air Quality Modeling Final Rule Technical Support Document - 2015 Ozone NAAQS Good Neighbor Plan. https://www.epa.gov/system/files/documents/2023-03/AQ%20Modeling%20Final%20Rule%20TSD.pdf. Accessed 10/06/2023.

sites with ambient air concentrations that may contribute to downwind deposition, but we expect that uncertainty is relatively small. And, as noted in Chapter 6, the purpose of these trajectory-based assessments of upwind concentrations and downwind deposition is to identify the pollutant and metrics for which such relationships are most evident. This analysis was not designed to be predictive of any such association.

This analysis evaluated one trajectory per day from each of the monitoring site locations with a valid DV (or DV-like metric) for a given pollutant across the 2000-2020 period. The locations of the monitoring sites for each pollutant are shown in Figure 6A-3. The daily trajectories were initiated at 1800 GMT which is generally midday over the U.S. This methodological decision ensures that the trajectories start in a period in which the planetary boundary layer is generally well-mixed and therefore representative of all emissions within the boundary layer (i.e., ground-level sources and sources with elevated stacks). Additionally, the decision was made to initiate all of the daily trajectories at 500 meters. Again, this is designed to ensure that trajectories are generated that are representative of the entire mixed layer where the most significant transport takes place (as opposed to the shallow surface layer that may exist at night or during temperature inversion conditions). We note that this choice of trajectory height is consistent with past EPA practice (e.g., designations guidance[7]) and consistent with the recommendations of HYSPLIT developers ("if only starting at one height, then a good choice might be one half of the planetary boundary layer"[8]). Again, these methodological choices, while sensible and consistent with past practice, do have the potential to affect the ultimate identification of the sites of influence. That said, we think the uncertainty associated with these choices is relatively low and entirely consistent with the intended use of these data (i.e., inform illustrative relationships).

[7] Memorandum from Gina McCarthy to USEPA Regional Administrators, https://www3.epa.gov/pmdesignations/2012standards/docs/april2013guidance.pdf. Accessed 10/06/2023.

[8] NOAA, HYSPLIT Cheat Sheet. https://www.ready.noaa.gov/documents/ppts/Cheat_Sheet_2020.pdf. Accessed 10/06/2023.

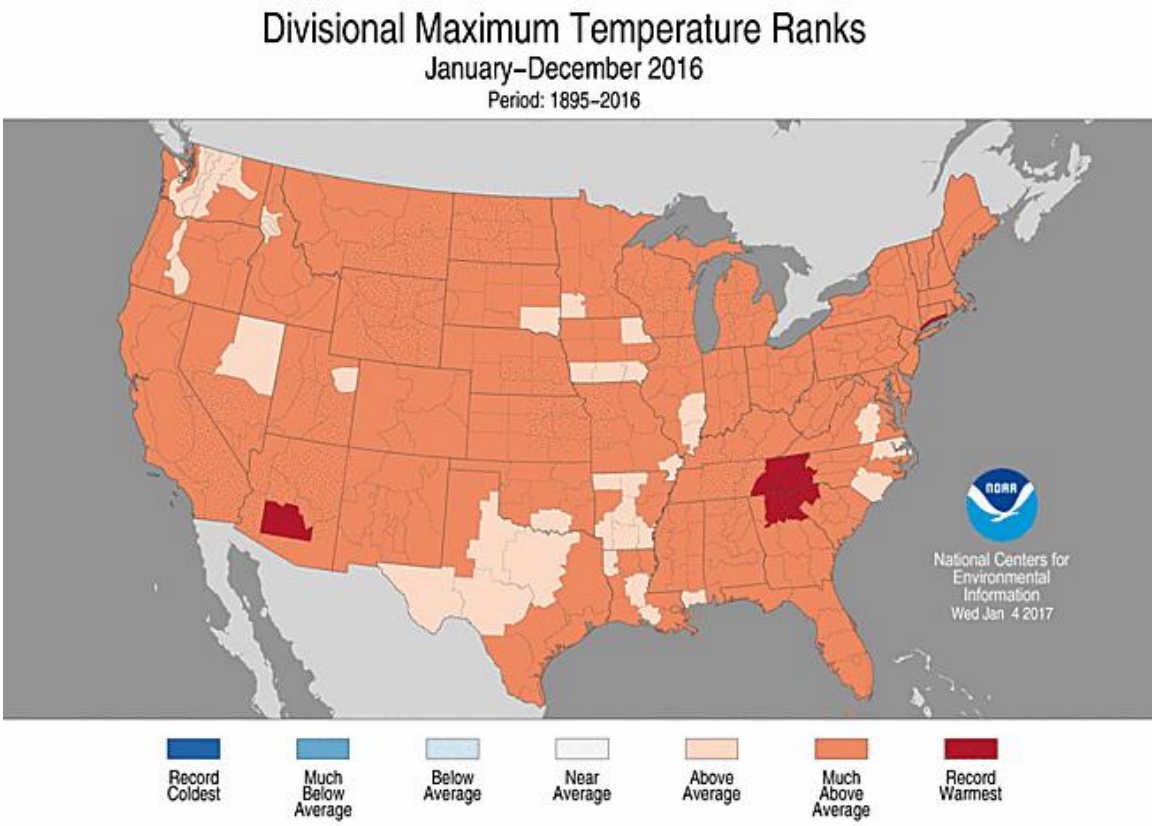

Figure 6A-1. 2016 annual average maximum temperatures by U.S. divisions binned across seven categories based on how 2016 differed from the 1895-2016 climatological average. (Source: NOAA/NCEI)

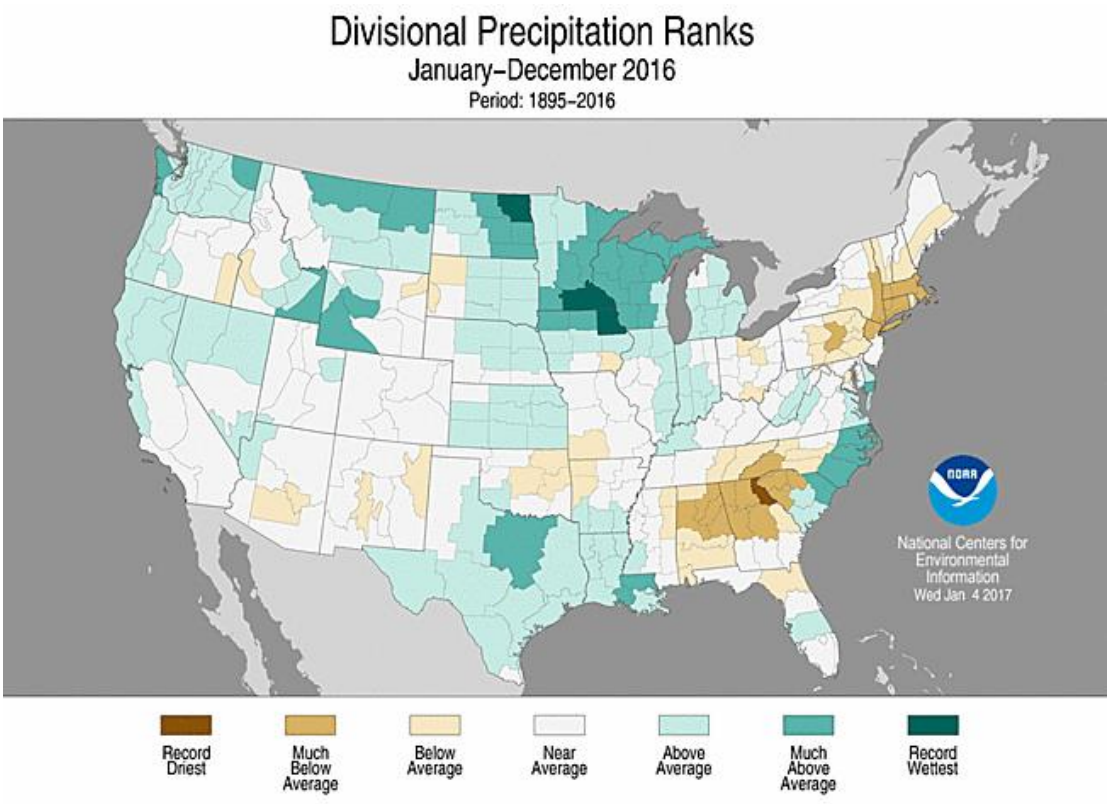

Figure 6A-2. 2016 annual average precipitation amounts by U.S. divisions binned across seven categories based on how 2016 differed from the 1895-2016 climatological average. (Source: NOAA/NCEI)

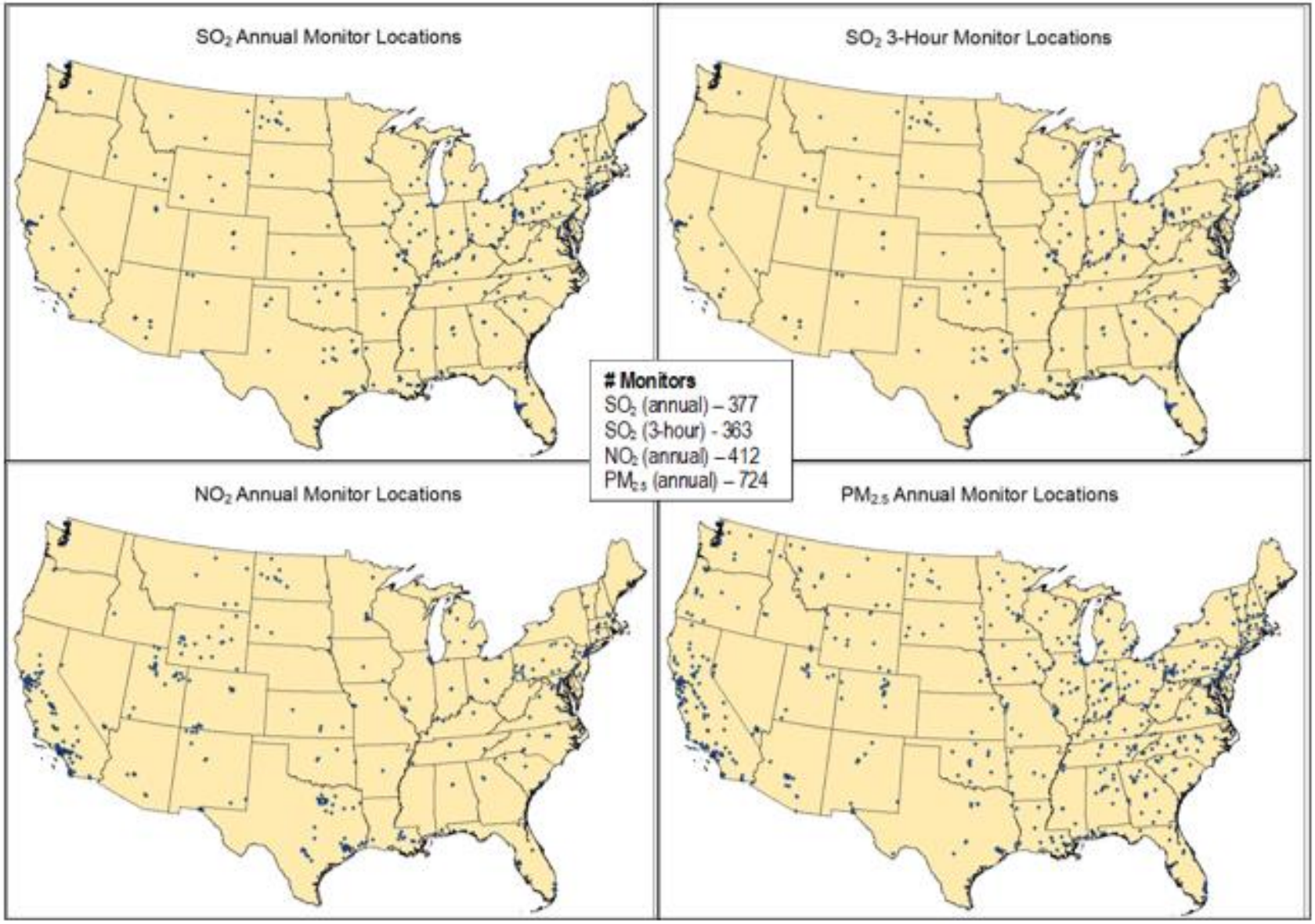

Figure 6A-3. Monitoring site locations for which daily HYSPLIT trajectories were generated for four air quality metrics.

As noted above, two sets of analyses were generated: the original analyses and the final analyses. Forward trajectories were calculated of a specified duration (48- or 120-hours) with an initial plume height of 500 m and a single year (2016) of meteorological data. For the original analyses, the meteorological input were from the 32-km resolution North American Regional Reanalysis (NARR-32). The NARR-32 dataset is one of several meteorological input options for HYSPLIT. Because the resolution of the meteorological data governing the forward trajectories was a relatively coarse 32 km for the original 48-hr modeling, there can be some uncertainty in cases where a trajectory only interacts with the periphery of an ecoregion, as to whether or not the upwind site should be considered as a site of influence. For the final analyses, we used meteorological data from the finer-resolution 12-km North American Mesoscale System (NAM-12) for the 120-hr HYSPLIT modeling. While the finer resolution meteorological data is expected to allow for more precision, this choice is not expected to significantly affect the results, and the same considerations with respect to the periphery of an ecoregion would likely still apply. The meteorological data source and the trajectory length were the only inputs that differed between the original and final analyses.

Each trajectory was divided into sequential segments corresponding to 10 minutes of the trajectory length (i.e., 288 segments for a 48-hour trajectory) to trace the trajectory at a relatively fine temporal frequency. Using geospatial tools, we assessed the number of forward trajectory segments for a day's trajectory from an individual monitoring site that fell into each of ecoregions. If at any point, the trajectory crossed into the boundary of the ecoregion, this trajectory site-day was counted as a "hit." The analysis evaluated the frequency of trajectory "hits" for each monitoring site / ecoregion pair. In the initial analysis, if more than 1% of the total hits for an ecoregion could be tracked back to a monitoring site, then that site was considered to be potentially representative of the air quality concentrations that influence deposition in that ecoregion. Figure 6A-4 depicts the outcome of this analysis using this "monitor inclusion criterion" of 1% and a 48-hour trajectory duration for one ecoregion-pollutant metric pair. For this ecoregion in central Kentucky (8.3.3), given the prevailing winds, the original trajectory analysis indicates that $PM_{2.5}$ from sites within the ecoregion itself, along with some sites in surrounding upwind areas (e.g., Southwest IN, Central TN) may contribute to N and S deposition within the ecoregion, given the analysis parameters.

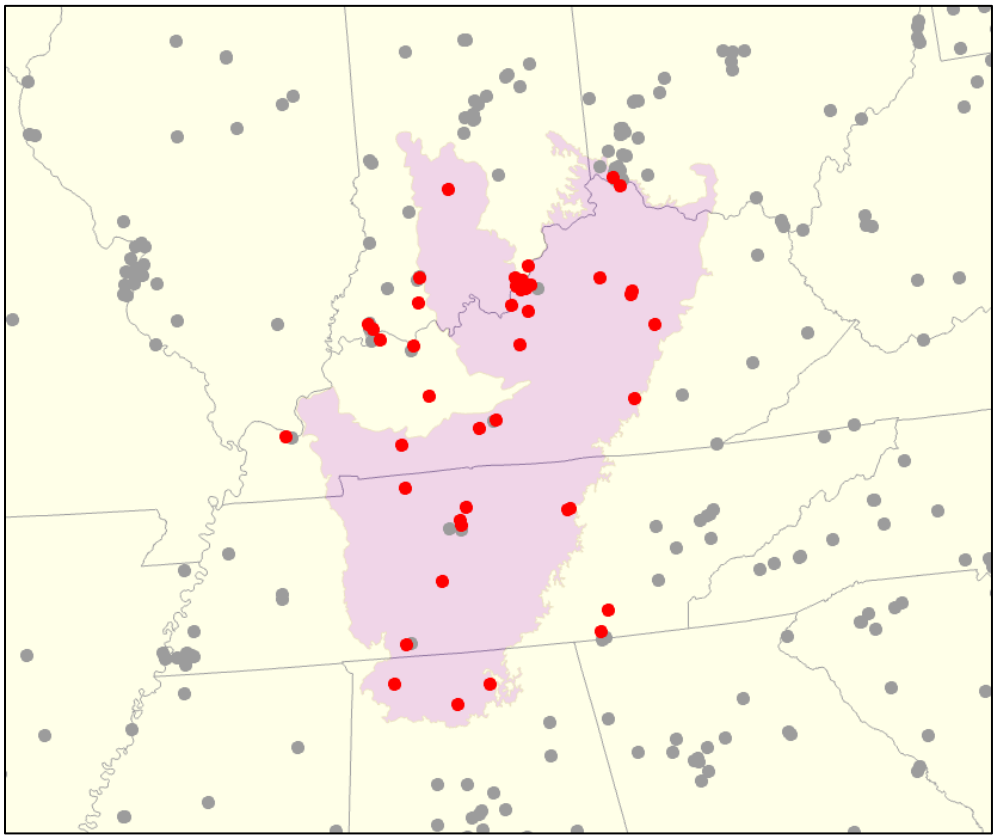

Figure 6A-4. Map of $PM_{2.5}$ monitoring sites of potential influence (red circles) for ecoregion 8.3.3 (purple shaded region) based on the original trajectories and a 1% hit rate as criterion for monitoring site inclusion. Other PM monitoring sites that did not meet the criterion are shown as gray circles.

We then considered whether a longer duration trajectory might be more appropriate for evaluating the totality of transport paths of S and N emissions that can contribute to downwind deposition. The final trajectory analysis was borne out of sensitivity testing that considered 5-day (120-hour) trajectories. Additionally, we conducted sensitivity tests to assess how different values for the monitoring site inclusion criterion could affect the determination of potential sites of influence. These sensitivity analysis were designed to enable consideration of more distant monitoring locations that could also be considered as part of an ecoregion's set of sites of influence given the relatively long atmospheric lifetimes of some pollutants and potential long transport distances that can contribute to deposition. We considered three different hit rates as criteria for monitoring site inclusion (1.0%, 0.5%, and 0.1%).

A set of sample sensitivity results are shown in Figures 6A-5 through 6A-20. These maps illustrate the impact, for 16 example ecoregions, of using different trajectory hit rates as criteria for monitoring site inclusion using the monitoring sites for the annual SO_2 metric. Sample results

for these 16 ecoregions for the other three metrics are included in Attachment 1 to Appendix 6A. In each figure, the black circles represent sites contributing at least 1.0% of the total trajectory "hits" to the ecoregion. Dark blue circles are sites contributing 0.5% to 1% of the ecoregion's trajectory hits. Light blue circles are sites contributing 0.1 to 0.5% of the total trajectory hits. Other monitoring sites contributing less than 0.1% of an ecoregion's total trajectory hits are shown as gray circles.

Looking at the Northern Lakes and Forests Ecoregion (5.2.1), Figure 6A-5 shows that the upwind sites contributing at least 1.0 percent of the total trajectory hits are located in the ecoregion itself or in an area in close proximity to the ecoregion. These locations are depicted by the black circles and suggest that transport of air pollution into this ecoregion is generally from the south and the west. Reducing the hit rate inclusion criterion to include sites contributing as low as 0.5% of the ecoregion's total hits increases the number of sites of influence. Specifically, including sites with hit rates at or above 0.5% (dark blue and black circles) results in sites of influence in an area extending from North Dakota to northern Oklahoma, as well as many more sites in the northern Mississippi River Valley (Figure 6A-5). Finally, we examined the sites of influence associated with a hit rate at or above 0.1% (light blue, dark blue and black circles). For this inclusion criterion, the analysis indicates trajectories reaching the Northern Lakes and Forests ecoregion from as far away as California and southern Texas (Figure 6A-5). The remainder of the figures show the sensitivity results for 15 other ecoregions.

It is not possible to determine with certainty which monitor inclusion criterion is most appropriate for identifying the possible sites of influence (over which the EAQM is calculated). Based on the results of our sensitivity analyses, using 120-hour trajectories, the 0.5% threshold appears to be a better match with pollutant deposition lifetimes and therefore appropriate for considering how transport from one upwind area may affect a downwind area. As a result, the final analysis has used this threshold value (0.5%).

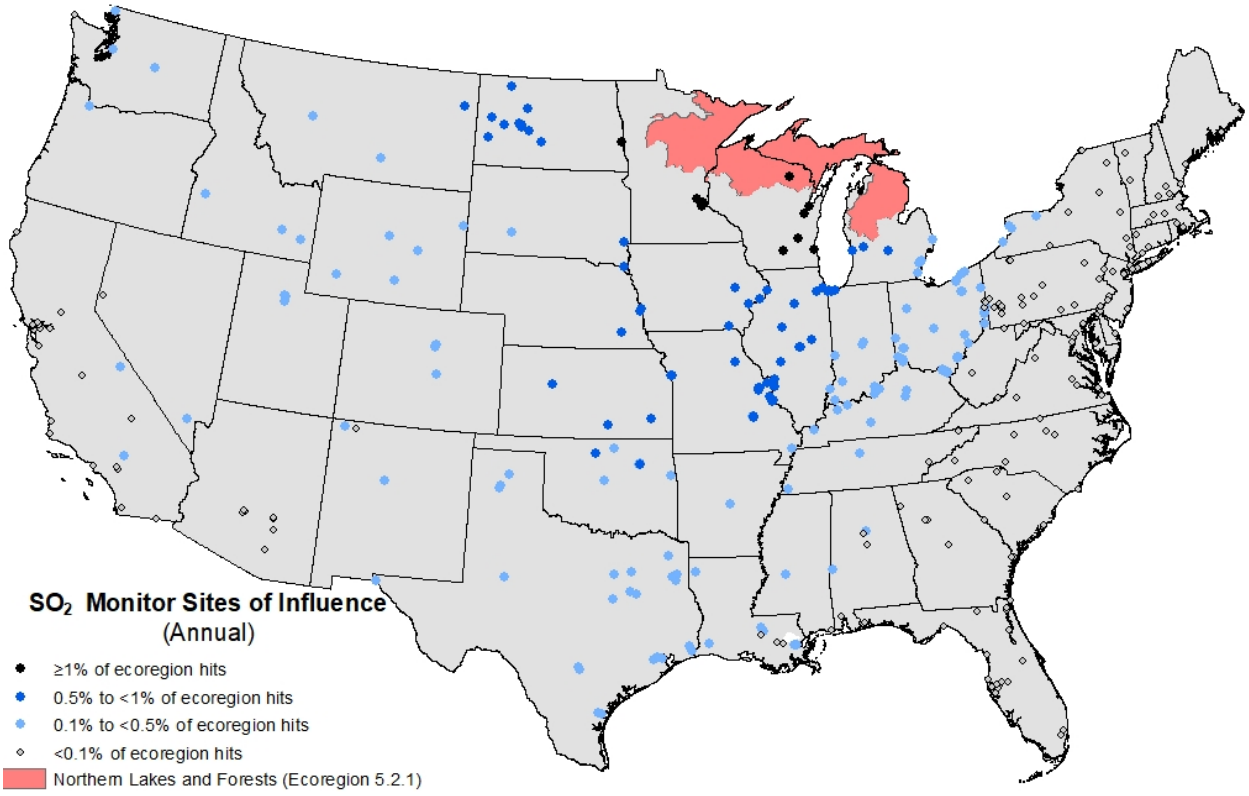

Figure 6A-5. Monitoring sites (annual SO_2 metric) of potential influence for ecoregion 5.2.1 (red shaded region).

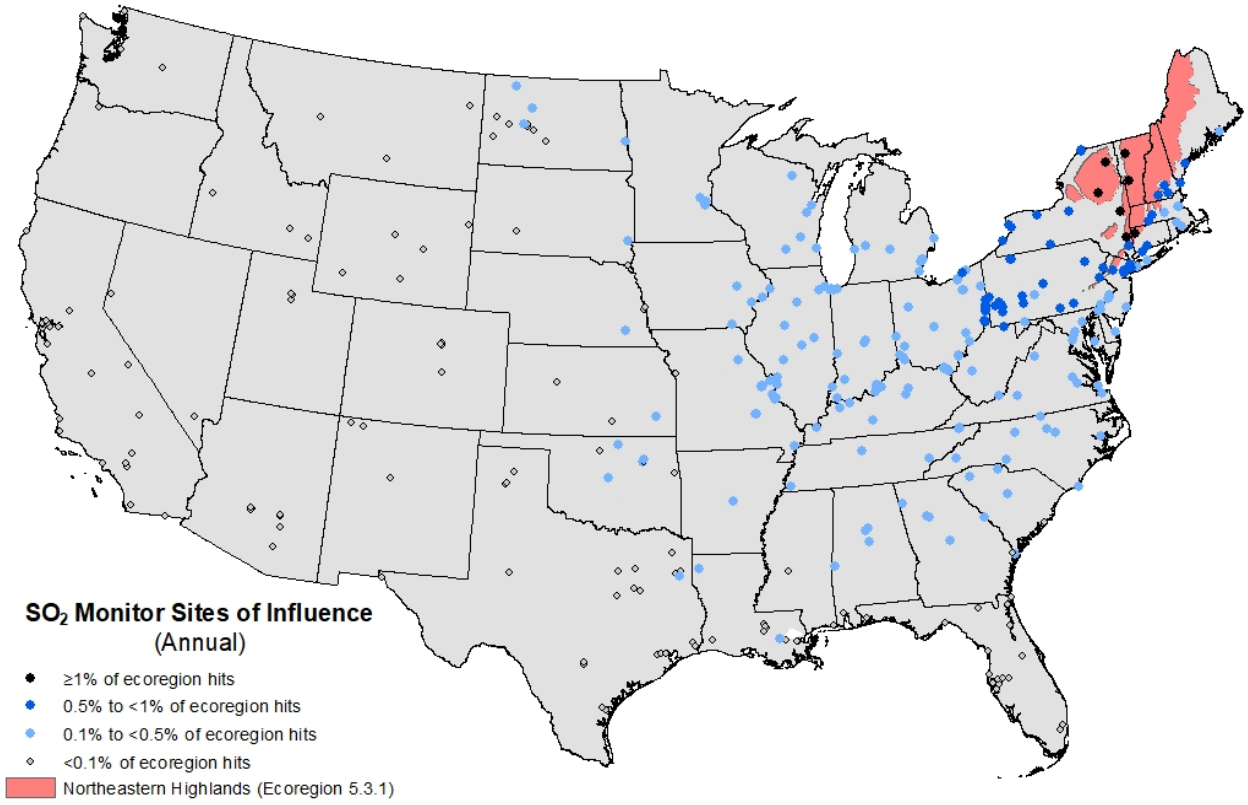

Figure 6A-6. Monitoring sites (annual SO_2 metric) of potential influence for ecoregion 5.3.1 (red shaded region).

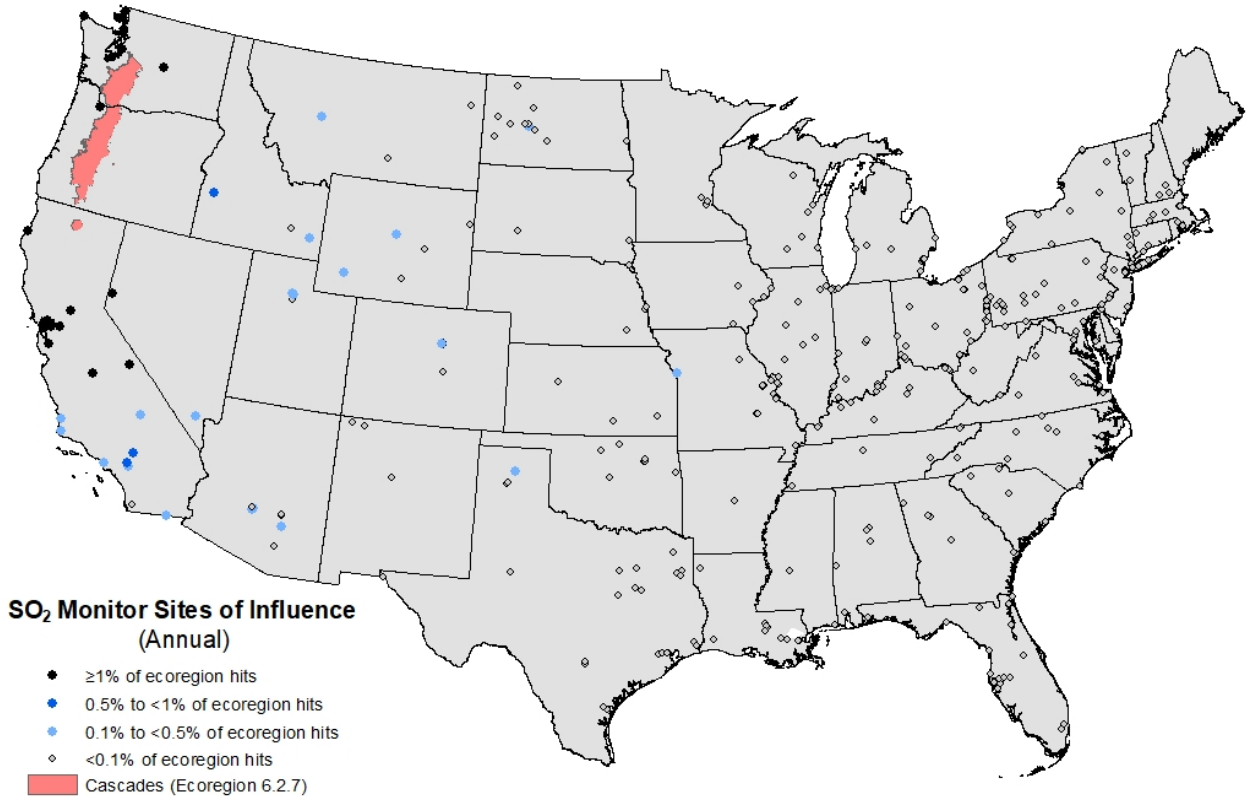

Figure 6A-7. Monitoring sites (annual SO_2 metric) of potential influence for ecoregion 6.2.7 (red shaded region).

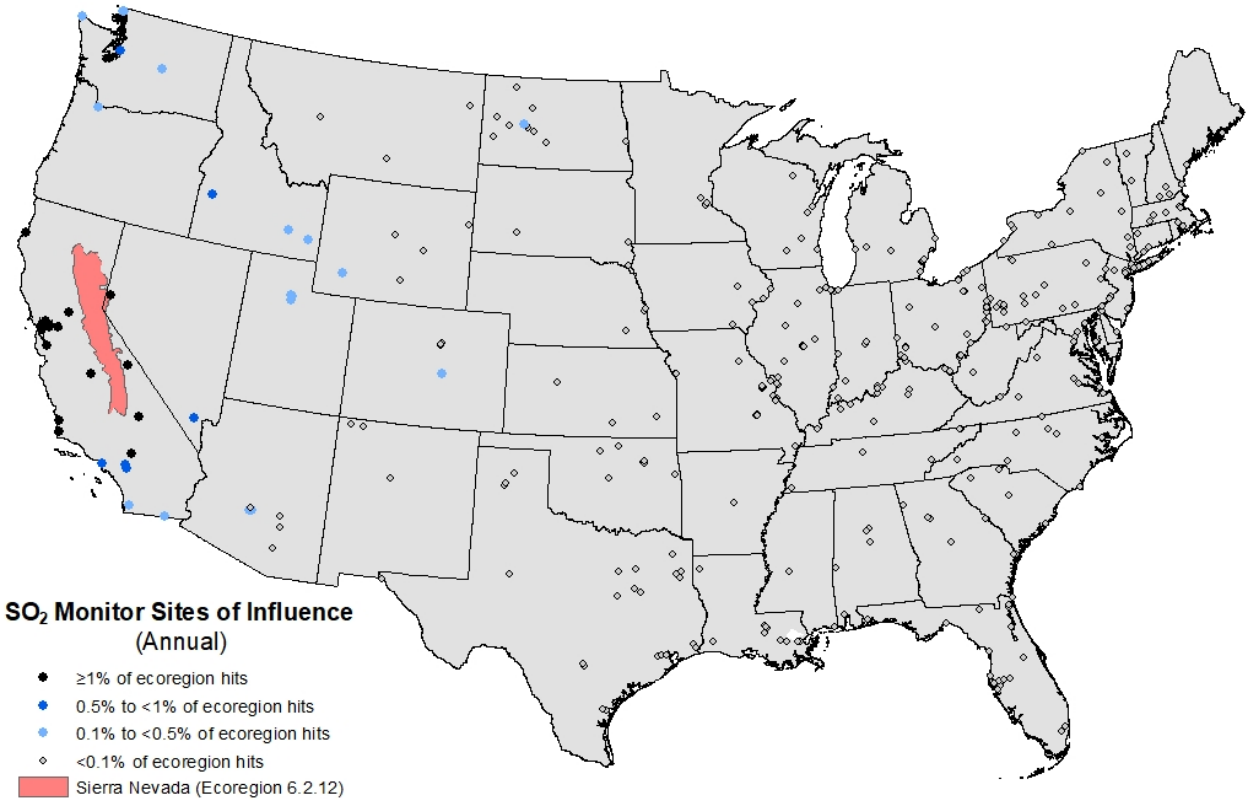

Figure 6A-8. Monitoring sites (annual SO_2 metric) of potential influence for ecoregion 6.2.12 (red shaded region).

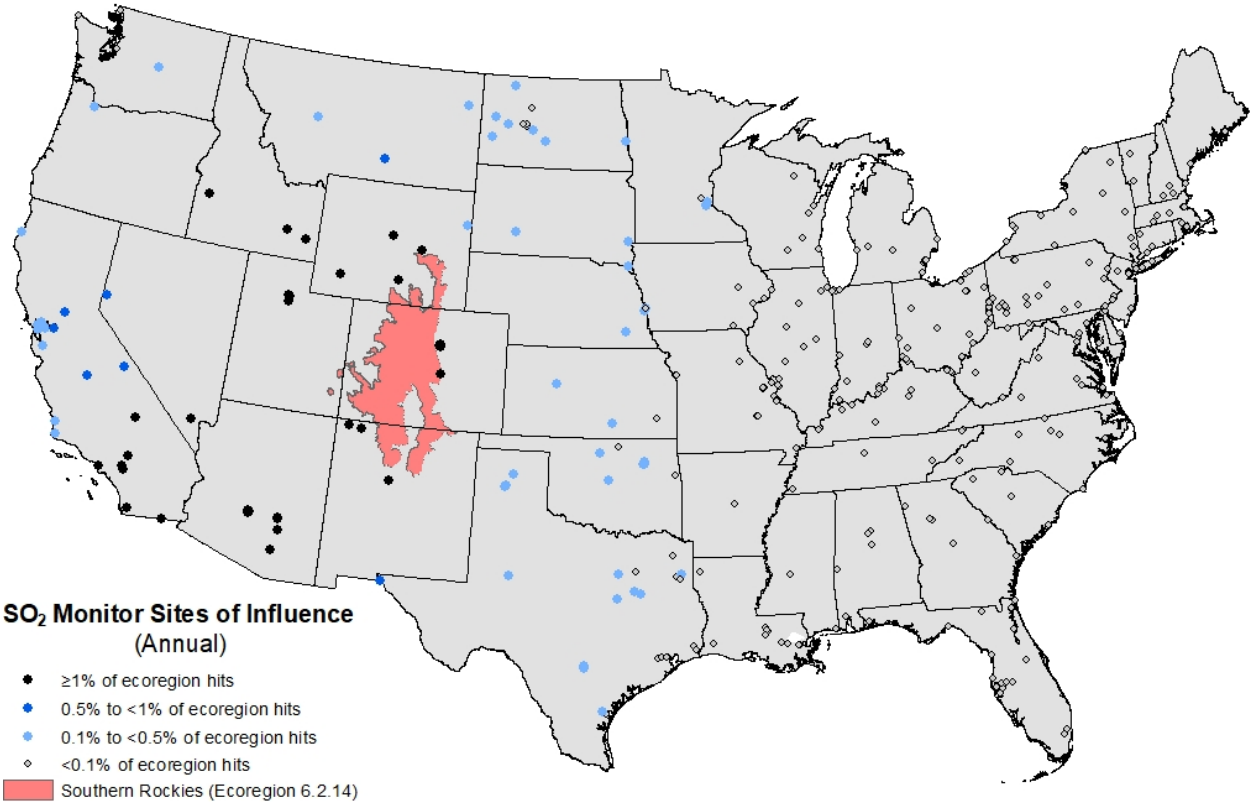

Figure 6A-9. Monitoring sites (annual SO_2 metric) of potential influence for ecoregion 6.2.14 (red shaded region).

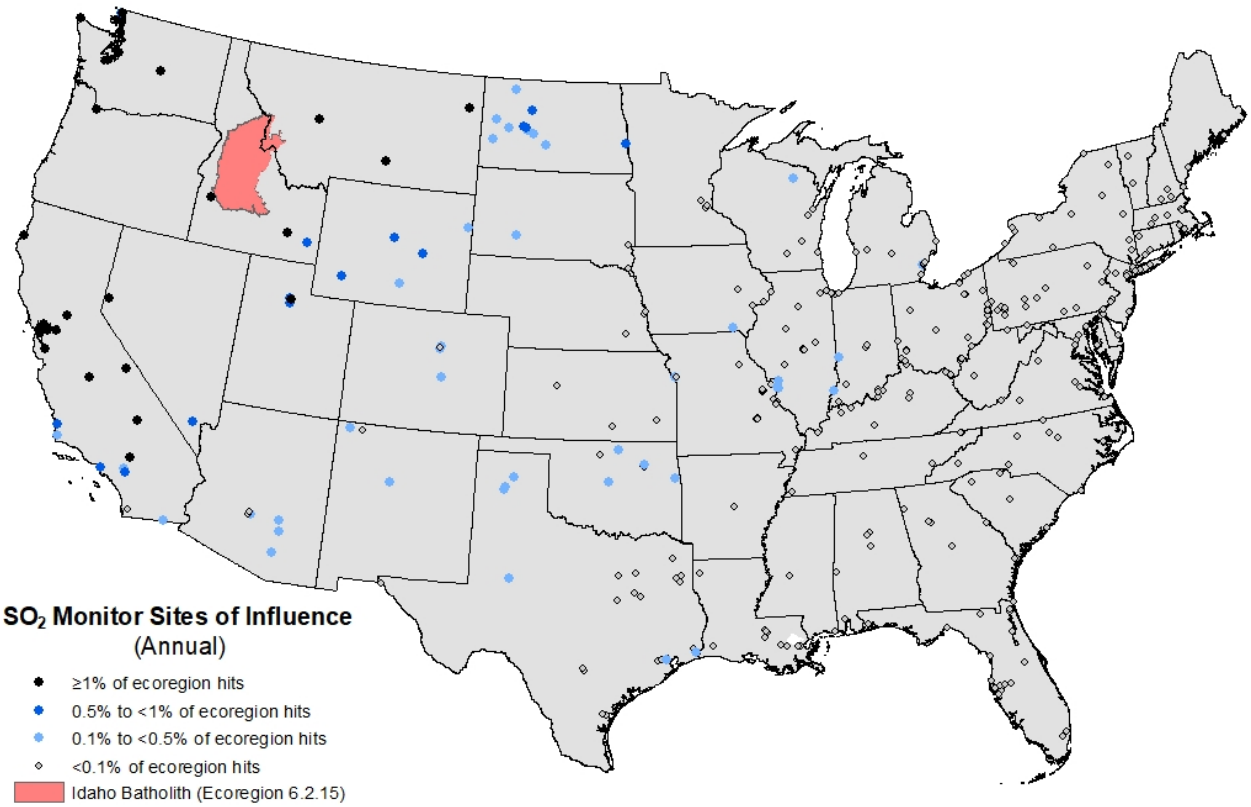

Figure 6A-10. Monitoring sites (annual SO_2 metric) of potential influence for ecoregion 6.2.15 (red shaded region).

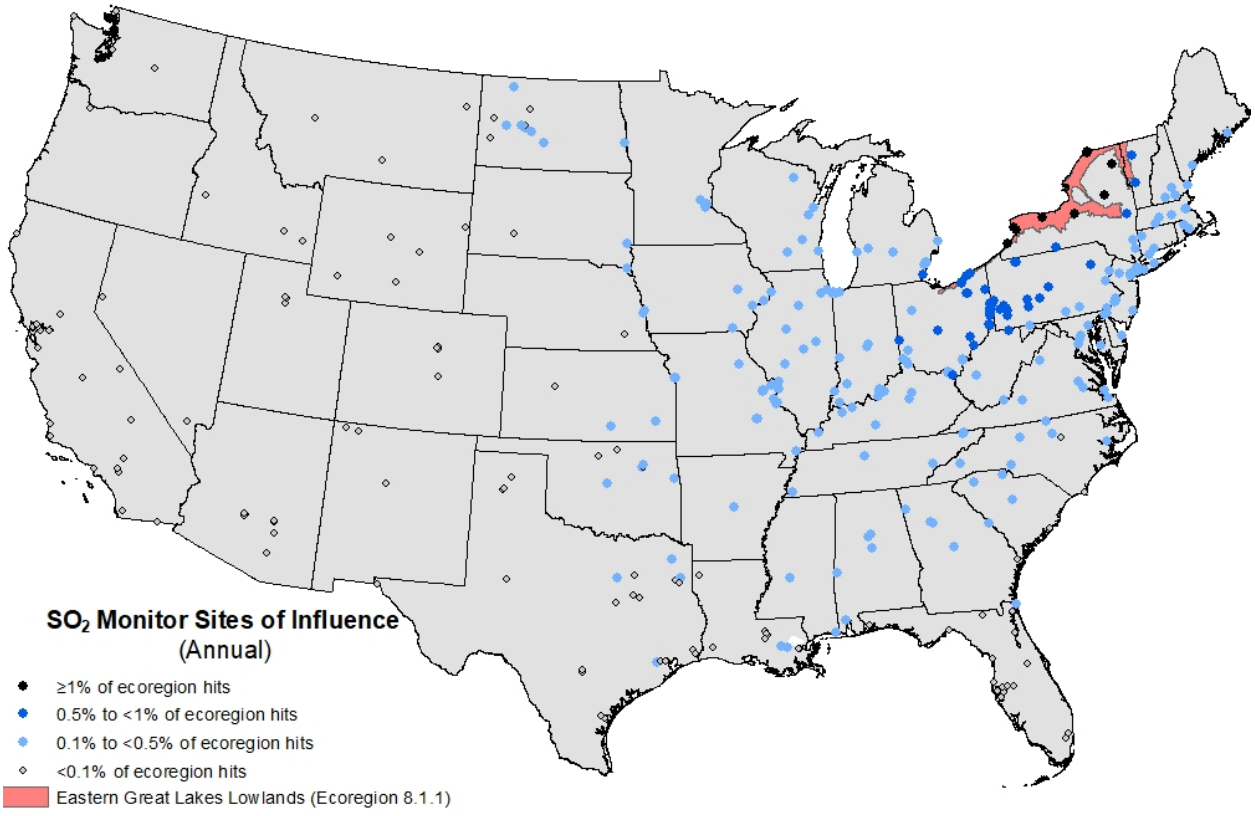

Figure 6A-11. Monitoring sites (annual SO_2 metric) of potential influence for ecoregion 8.1.1 (red shaded region).

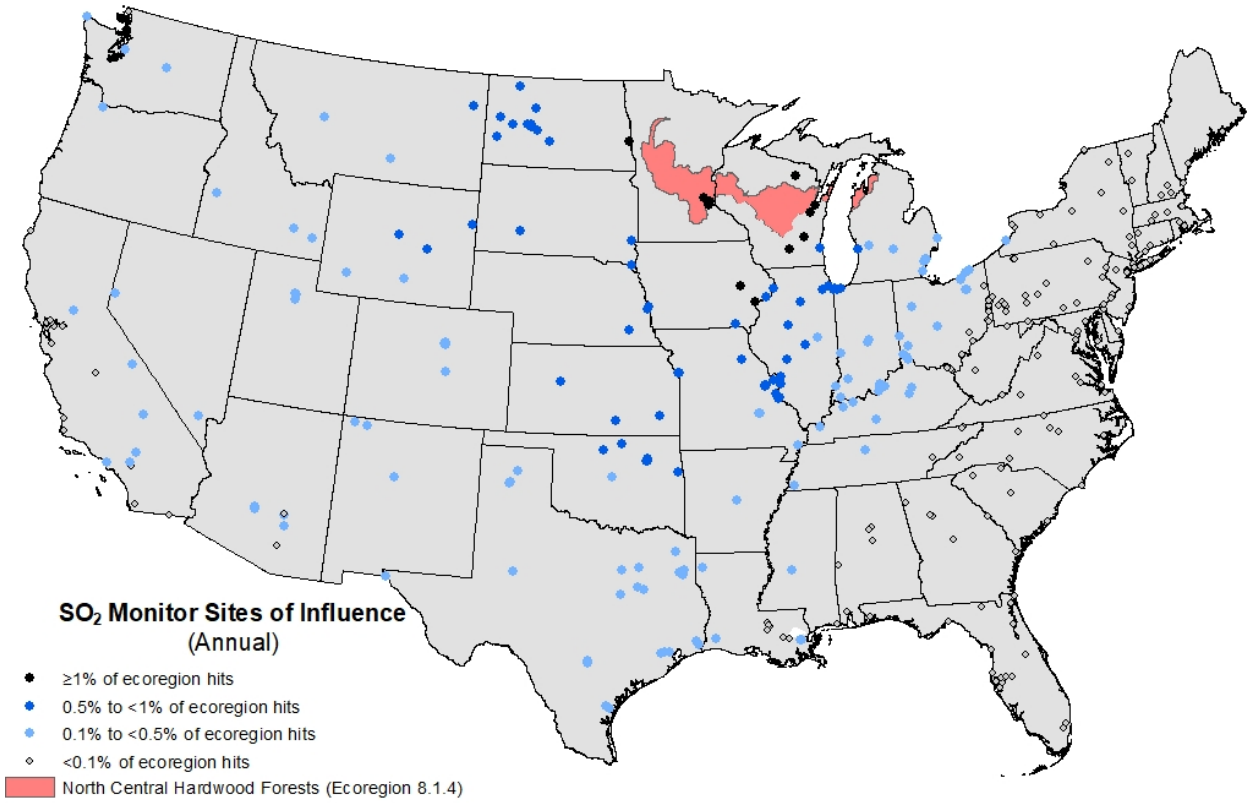

Figure 6A-12. Monitoring sites (annual SO_2 metric) of potential influence for ecoregion 8.1.4 (red shaded region).

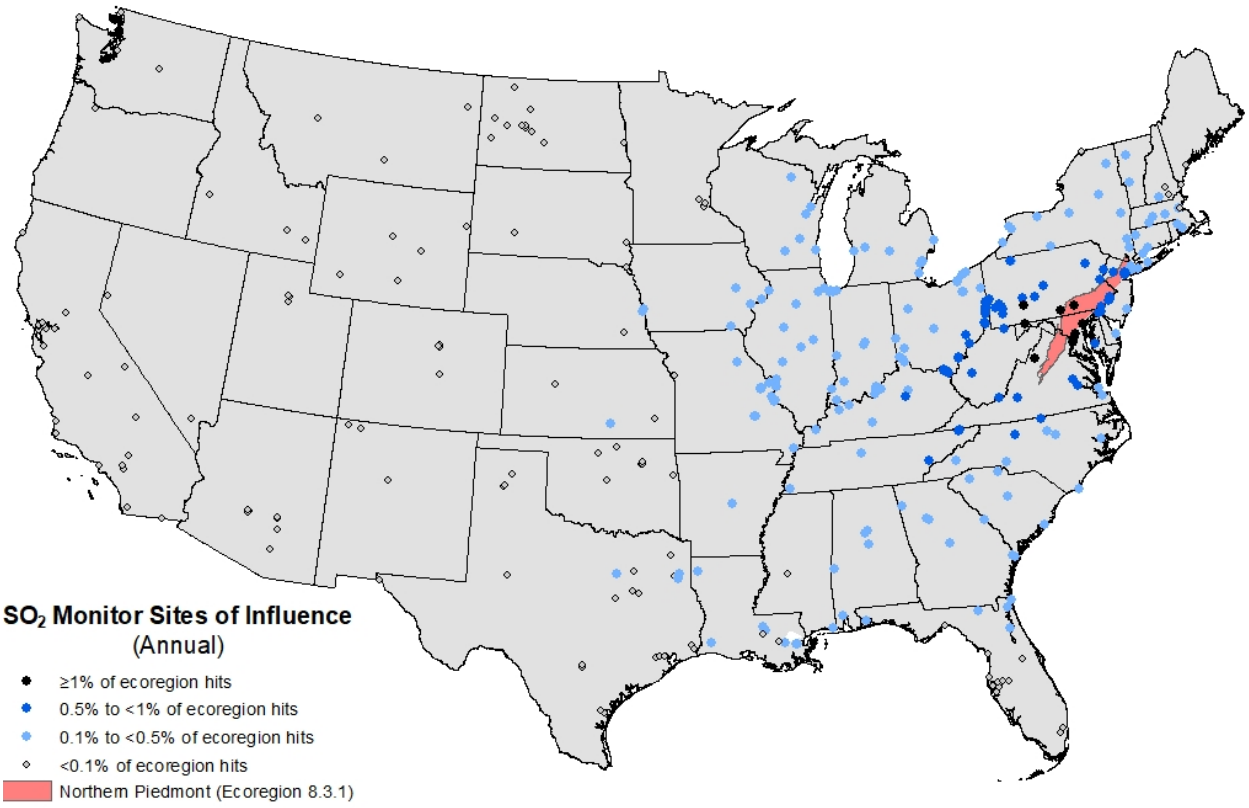

Figure 6A-13. Monitoring sites (annual SO_2 metric) of potential influence for ecoregion 8.3.1 (red shaded region).

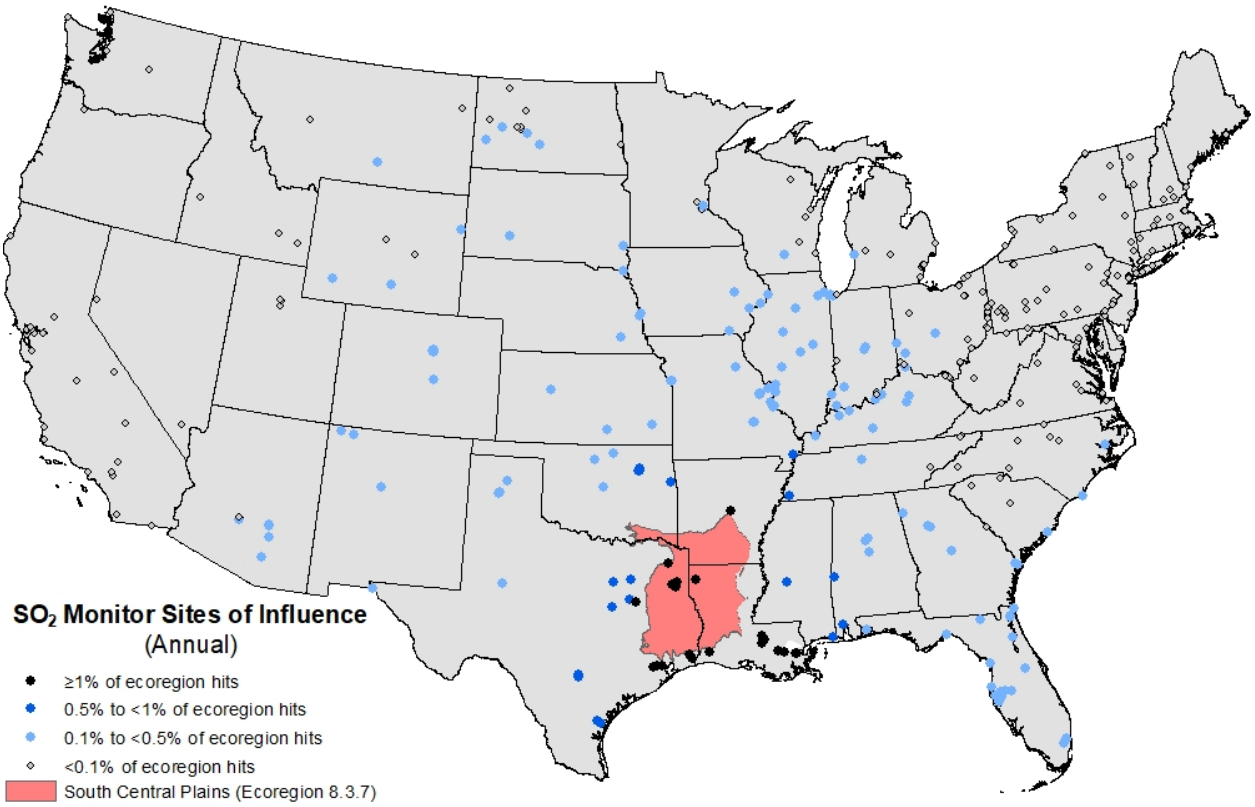

Figure 6A-14. Monitoring sites (annual SO_2 metric) of potential influence for ecoregion 8.3.7 (red shaded region).

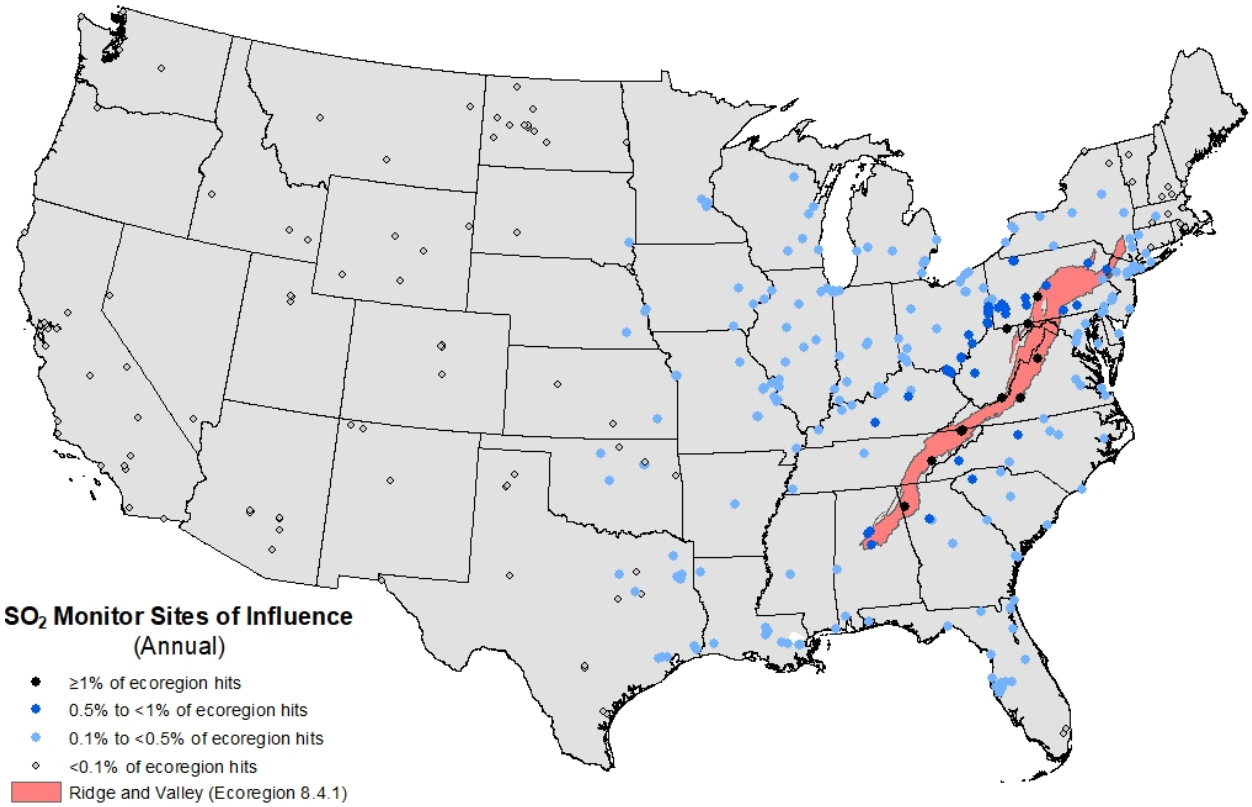

Figure 6A-15. Monitoring sites (annual SO_2 metric) of potential influence for ecoregion 8.4.1 (red shaded region).

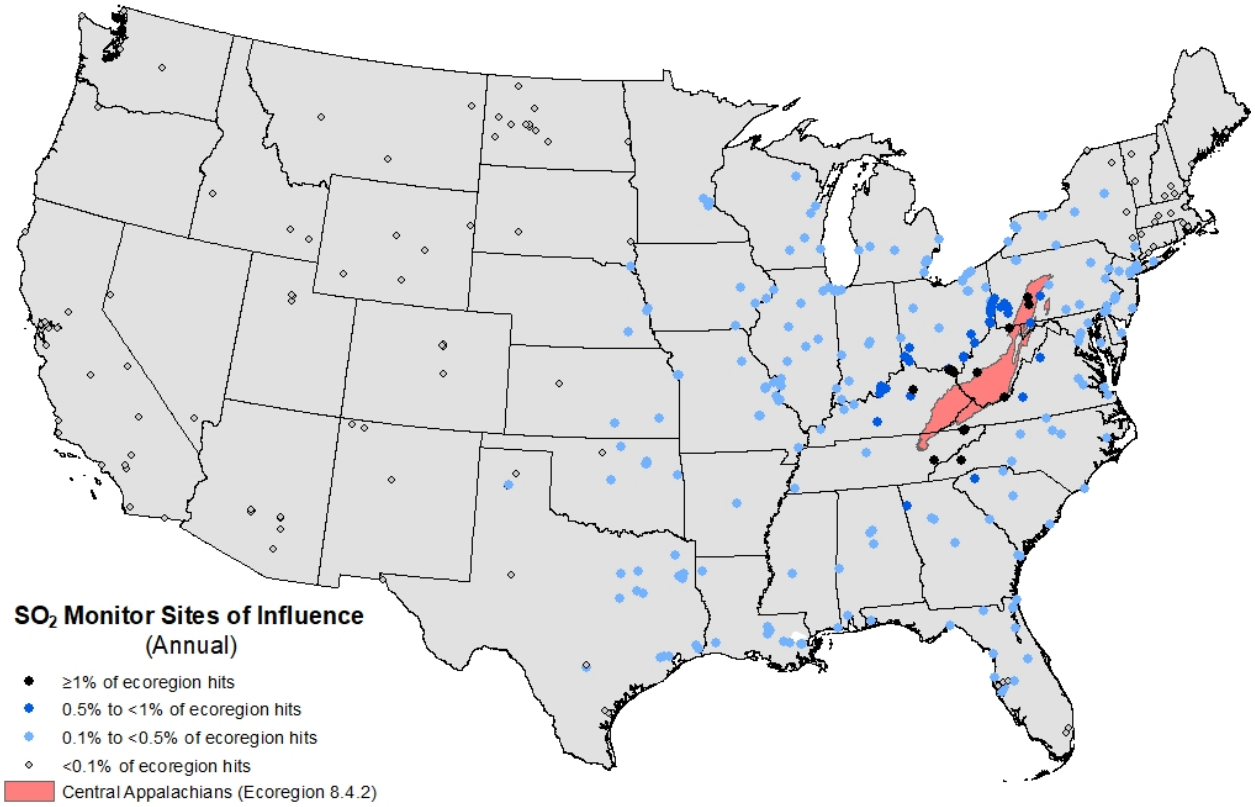

Figure 6A-16. Monitoring sites (annual SO_2 metric) of potential influence for ecoregion 8.4.2 (red shaded region).

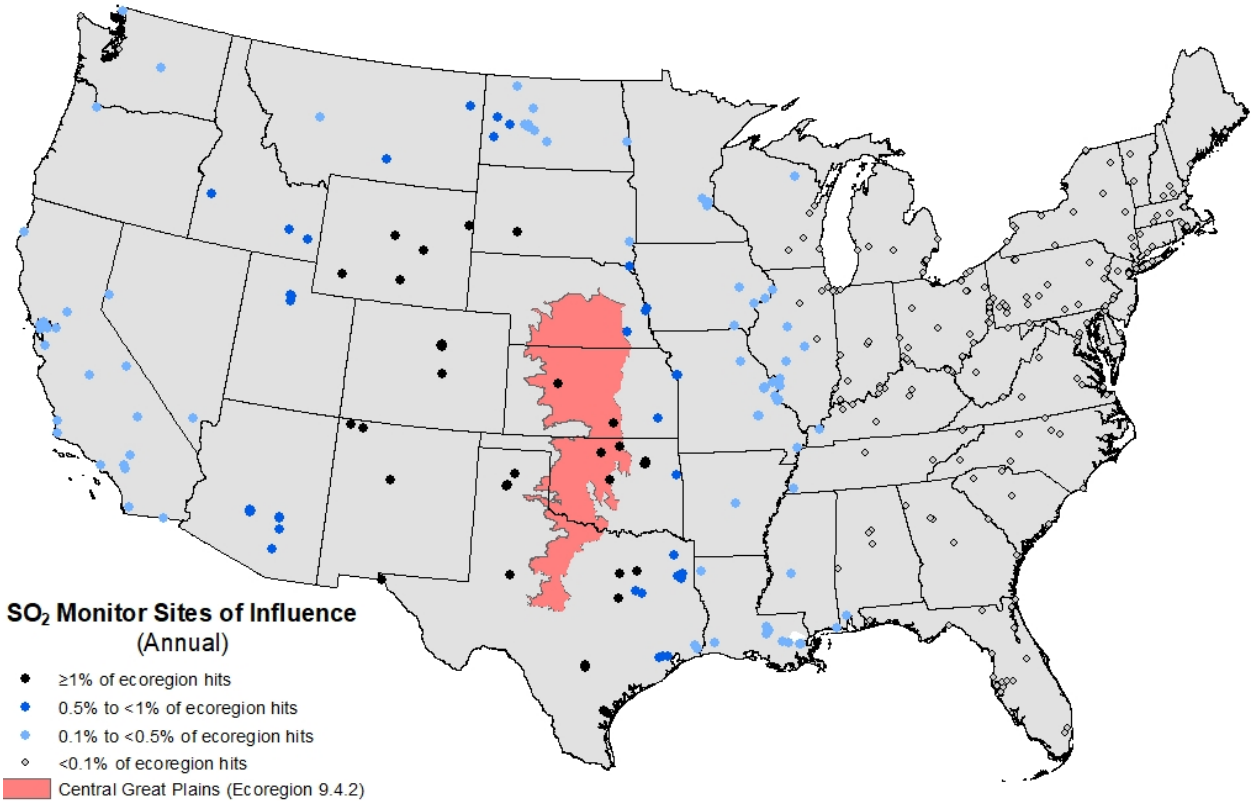

Figure 6A-17. Monitoring sites (annual SO_2 metric) of potential influence for ecoregion 9.4.2 (red shaded region).

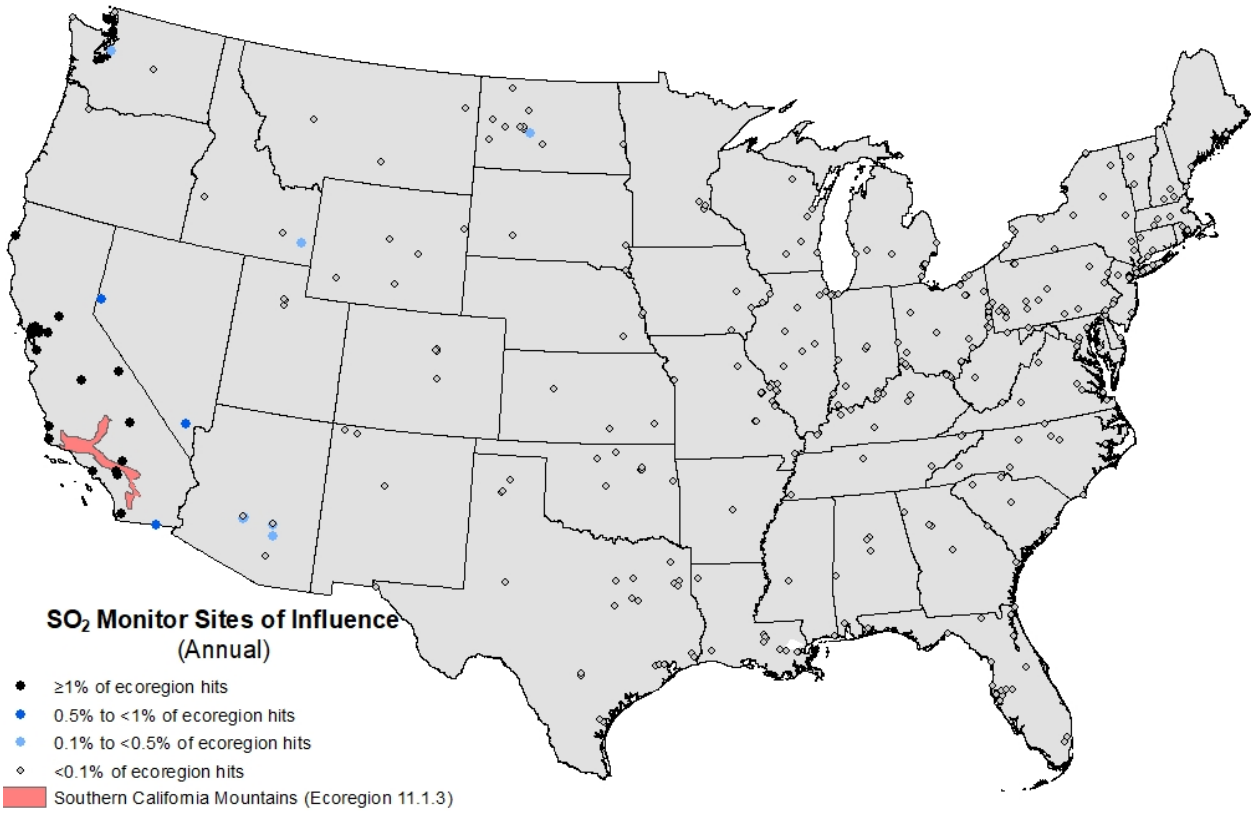

Figure 6A-18. Monitoring sites (annual SO_2 metric) of potential influence for ecoregion 11.1.3 (red shaded region).

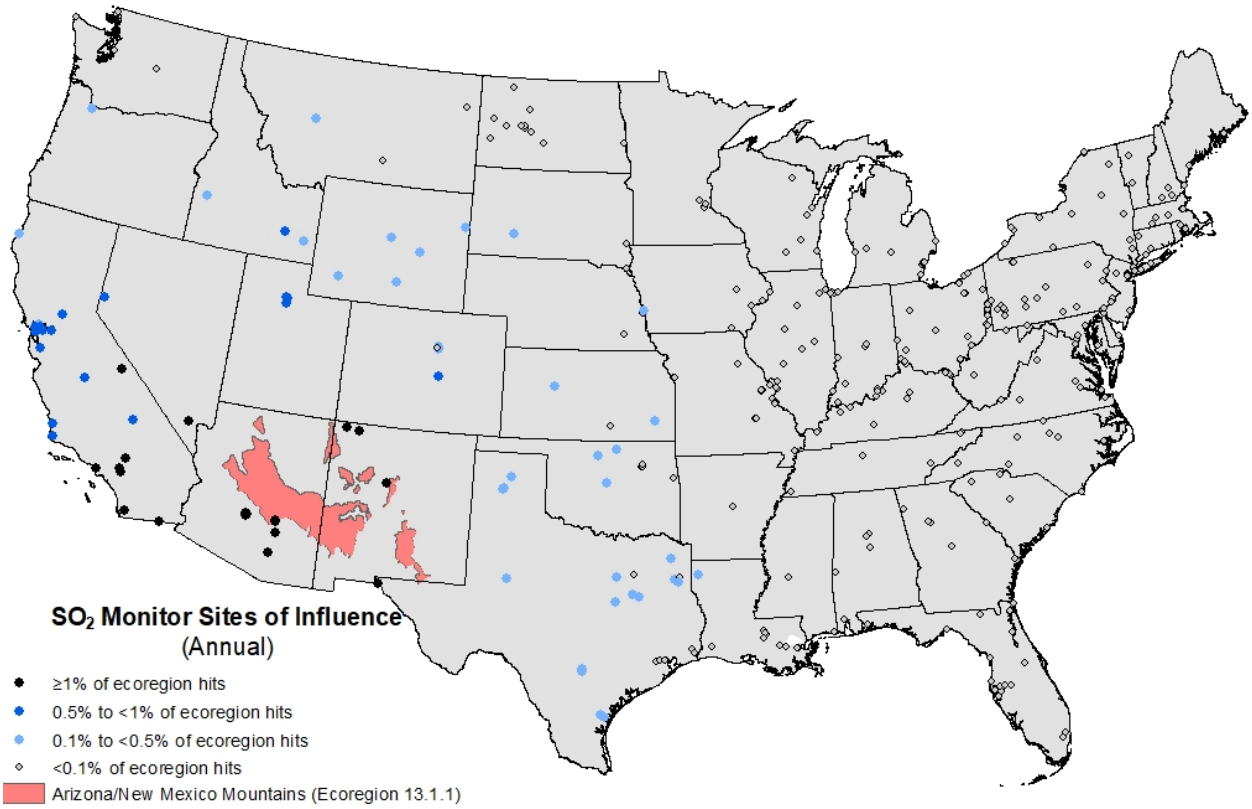

Figure 6A-19. Monitoring sites (annual SO_2 metric) of potential influence for ecoregion 13.1.1 (red shaded region).

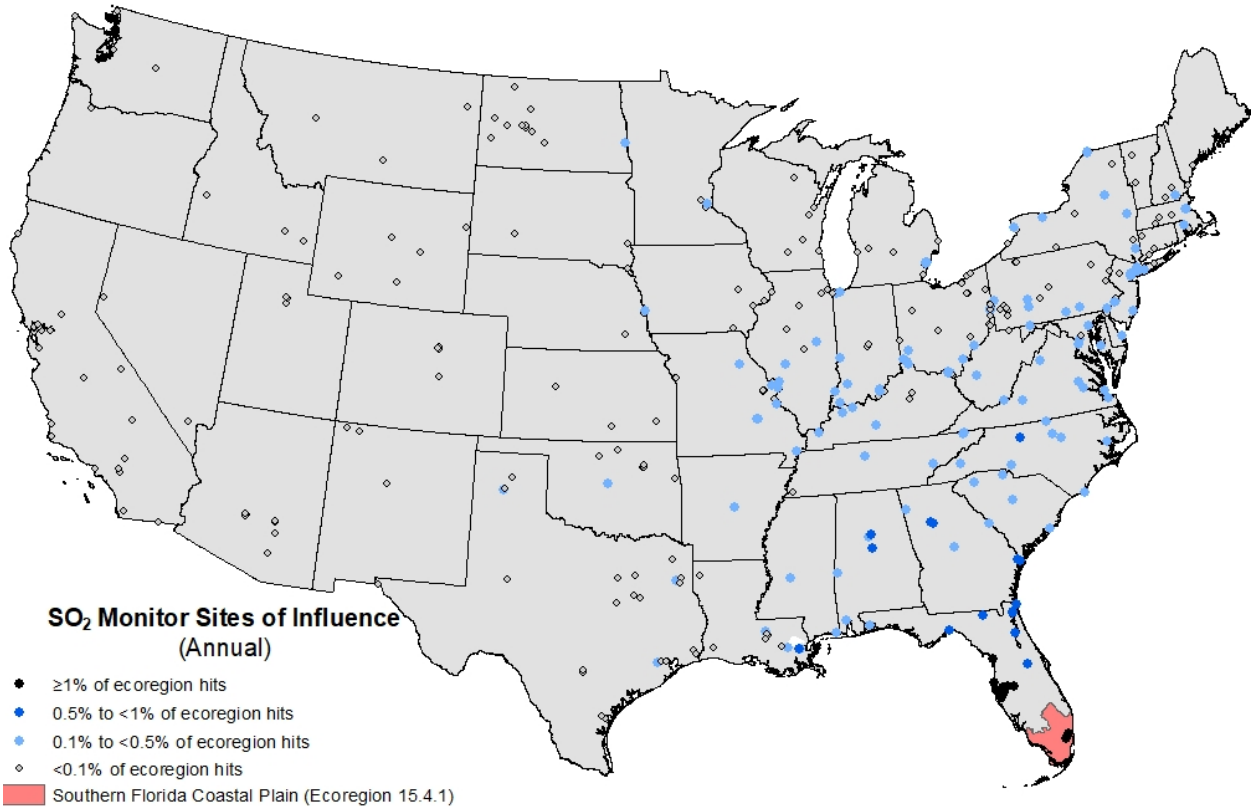

Figure 6A-20. Monitoring sites (annual SO_2 metric) of potential influence for ecoregion 15.4.1 (red shaded region).

6A.3. ESTIMATION OF ECOREGION AIR QUALITY METRICS

After the trajectories were generated and the air quality monitoring sites of influence were identified for each ecoregion-pollutant metric pair, the next step in this analysis was to investigate the relationship between air quality levels at the upwind sites and deposition levels in the downwind ecoregion. For each pollutant metric, two types of EAQMs were derived for each ecoregion based on the air quality data for that ecoregion's contributing monitors:

- *EAQM-max*: the highest value from any monitor within the sites of influence, and
- *EAQM-weighted*: a weighted average, where each monitor value is weighted by the percentage of HYSPLIT hits to the ecoregion.

Both versions of EAQMs have value. EAQM-max represents the highest EAQM within the upwind region potentially contributing to deposition in an ecoregion, and as such it enables an assessment of the relationship between deposition levels and *worst-case* monitored air quality that is associated with that level of deposition. Given that EAQM-weighted considers the relative contributions from different upwind directions, it is presumed to represent the *general-case* upwind air quality that is associated with downwind deposition. Design values at sites closer to

the ecoregion itself will have more weight as impactful trajectories from these locations are more common. Both types of EAQMs have inherent uncertainties related to the trajectories themselves, the methodology used to link upwind regions to downwind receptors (e.g., monitor inclusion criterion), and the density of the existing monitoring network. All EAQM values are averaged over 3 years. EAQMs were generated for the following periods: 2001-2003, 2006-2008, 2010-2012, 2014-2016, and 2018-2020. Both types of EAQMs were generated for each of the 84 Ecoregion III areas for four separate combinations of pollutant and averaging time:

- SO_2: annual 2nd high of individual 3-hour averages, averaged over 3-year periods
- SO_2: annual average of hourly data, averaged over 3-year periods
- NO_2: annual average of hourly data, averaged over 3-year periods
- $PM_{2.5}$: annual average of hourly data, averaged over 3-year periods

To provide further explanation of the EAQM calculation, we consider a specific example EAQM value and the individual steps that lead to its calculation. For this example, we will consider the annual SO_2 metric in 2020 for Ecoregion 5.2.1 (Northern Lakes and Forests).

- Step 1: Identify sites with valid annual SO_2 data for any year between 2000 and 2020.
- Step 2: Apply HYSPLIT to simulate 120-hour forward trajectories from these locations.
- Step 3: Evaluate the number of trajectory segments that reside in Ecoregion 5.2.1 and determine which sites contribute at least 0.5% of the total trajectory segments that impact the ecoregion. For this example, 74 sites meet the 0.5% criterion and are plotted as either black or dark blue circles in Figure 6A-5. The site with the most frequent trajectory impacts was site 55-041-0007 in Forest County, WI. The site of influence with the lowest hit percentage above 0.5% was 29-093-0034 in Iron County, MO.
- Step 4: For EAQM-max, determine which of the 74 potential sites of influence had the highest annual SO_2 metric value in each year. For 2018 through 2020, the maximum values were 3.31 ppb (Macon County, IL), 2.01 ppb (Mercer County, ND) and 2.13 ppb (Macon County, IL). The average of these three values (EAQM-max for 2018-2020) was then paired with the 2018-2020 3-year average deposition estimates from the ecoregion. Accordingly, for the analyses in Chapter 6 of the PA that consider the relationships between upwind air quality concentrations (i.e., EAQMs) and downwind deposition in an ecoregion, in this example for the 2018-2020 period we are pairing 2.49 ppb, as the 3-year average of the highest annual average SO_2 concentrations at sites of influence with the 2018-2020, 3-year-average annual total S deposition in Ecoregion 5.2.1. We recognize, however, that the monitoring network density may not always allow for the capture of all pollution concentrations that ultimately contribute to downwind deposition, and that atmospheric loading (and consequently subsequent deposition) is more a

function of the uniform distribution of air concentrations. As such, the EAQM-max associations should consider that caveat.

- Step 5: To calculate the EAQM-weighted value for the same time period, access the 2018, 2019 and 2020 annual average concentrations from the 74 potential sites of influence and derive a weighted average for each year from the site-specific concentrations weighted by the fraction of impacting trajectories arising from that site. Then take the 3-year average of the EAQM-weighted values for the three years. The resulting EAQM-weighted SO_2 for Ecoregion 5.2.1 in 2018-2020 was 0.77 ppb. For the analyses in Chapter 6 of the PA that consider the relationships between upwind air quality concentrations (i.e., EAQMs) and downwind deposition in an ecoregion, in this example we are concluding that as a regional generality, the annual average SO_2 value contributing to 2018-2020 total S deposition in Ecoregion 5.2.1 is 0.77 ppb.

As discussed further in Chapter 6, this analytical work culminates in a series of plots which display how the upwind EAQMs are related to median S and N deposition values for the downwind ecoregions. Again, the goal of this exercise is to examine the strengths of these associations and not to establish predictive relationships between EAQM values and deposition. The findings of this analysis are intended to help inform conclusions regarding the pollutants and concentration averaging times most strongly associated with eventual downwind deposition and might be useful in identifying policy options for controlling deposition with the potential for welfare effects.

6A.4. COMBINED EAQM AND DEPOSITION DATA

Linking the EAQM and ecoregion deposition data for the analysis in Chapter 6 was straightforward. As noted above, 8 sets of EAQM values were generated (i.e., 2 types for 4 different pollutant-forms) for five separate 3-year time periods between 2000 and 2020. Median ecoregion S and N deposition estimates (averages for the five 3-year periods) were then linked to the EAQM values for each ecoregion. The median TDep deposition value for a given year is derived from the estimates for all of the grid cells that comprise the ecoregion (at level III delineation).[9] Median deposition values were calculated using the zonal statistics tool in ArcMap. Grid cells from the TDep dataset were included if the centroid of the grid was within the ecoregion boundary.

The result was eight tables (one for each combination of the type of EAQM and the four pollutant metrics) that contain an EAQM value and TDep-based deposition value for each

[9] Deposition estimates for S and N were based on TDep v.2018.02 in all EAQM analyses (see https://nadp.slh.wisc.edu/committees/tdep/).

ecoregion and each of the 3-year time periods. The SO_2 tables (annual and 3-hour) include S deposition values. All other tables (i.e., NO_2, and $PM_{2.5}$) include N deposition values. An example EAQM table is shown in Table 6A-1.

Table 6A-1. EAQM-TDep table for a weighted annual SO_2 and S deposition.

Ecoregion	EAQM-weighted annual SO_2 (ppb)	Median ecoregion S dep (kg S/ha-yr)	Period
10.1.2	1.8	0.5	2001-2003
10.1.3	1.9	0.3	2001-2003
10.1.4	3.0	0.6	2001-2003
10.1.5	1.6	0.5	2001-2003
10.1.6	2.0	0.7	2001-2003
10.1.7	2.2	0.8	2001-2003
10.1.8	2.3	0.5	2001-2003
10.2.1	1.3	0.6	2001-2003
10.2.2	2.1	0.5	2001-2003
10.2.4	1.7	1.2	2001-2003
11.1.1	1.5	1.1	2001-2003
11.1.2	1.5	1.1	2001-2003
11.1.3	1.3	1.2	2001-2003
12.1.1	2.6	1.2	2001-2003
13.1.1	2.3	1.4	2001-2003
15.4.1	1.9	6.0	2001-2003
5.2.1	2.4	4.3	2001-2003
5.2.2	1.9	2.3	2001-2003
5.3.1	3.8	6.5	2001-2003
5.3.3	8.4	18.1	2001-2003
6.2.10	3.0	1.0	2001-2003
6.2.11	1.5	0.9	2001-2003
6.2.12	1.4	1.3	2001-2003
6.2.13	2.1	1.4	2001-2003
6.2.14	2.4	1.1	2001-2003
6.2.15	2.5	0.9	2001-2003
6.2.3	2.2	0.9	2001-2003
6.2.4	2.4	1.2	2001-2003
6.2.5	1.7	1.6	2001-2003
6.2.7	1.7	1.7	2001-2003

Ecoregion	EAQM-weighted annual SO_2 (ppb)	Median ecoregion S dep (kg S/ha-yr)	Period
6.2.8	1.5	0.4	2001-2003
6.2.9	1.7	0.5	2001-2003
7.1.7	1.8	2.1	2001-2003
7.1.8	1.5	2.4	2001-2003
7.1.9	1.8	1.6	2001-2003
8.1.1	6.6	11.0	2001-2003
8.1.10	7.7	18.4	2001-2003
8.1.3	6.6	11.9	2001-2003
8.1.4	2.0	4.6	2001-2003
8.1.5	2.6	5.4	2001-2003
8.1.6	4.2	9.6	2001-2003
8.1.7	4.7	9.6	2001-2003
8.1.8	3.9	4.5	2001-2003
8.2.1	3.3	7.0	2001-2003
8.2.2	4.7	9.9	2001-2003
8.2.3	3.9	9.8	2001-2003
8.2.4	5.1	14.8	2001-2003
8.3.1	6.4	14.9	2001-2003
8.3.2	4.3	10.5	2001-2003
8.3.3	4.7	13.5	2001-2003
8.3.4	3.9	11.7	2001-2003
8.3.5	2.9	9.7	2001-2003
8.3.6	3.4	8.6	2001-2003
8.3.7	2.7	7.3	2001-2003
8.3.8	2.2	6.4	2001-2003
8.4.1	5.6	14.1	2001-2003
8.4.2	6.3	16.2	2001-2003
8.4.3	8.3	20.4	2001-2003
8.4.4	4.1	11.1	2001-2003
8.4.5	3.1	6.3	2001-2003
8.4.6	2.6	6.0	2001-2003
8.4.7	2.6	5.5	2001-2003
8.4.8	2.6	6.2	2001-2003
8.4.9	4.0	14.7	2001-2003
8.5.1	3.7	10.5	2001-2003
8.5.2	3.3	7.4	2001-2003

Ecoregion	EAQM-weighted annual SO_2 (ppb)	Median ecoregion S dep (kg S/ha-yr)	Period
8.5.3	2.6	7.9	2001-2003
8.5.4	5.4	14.0	2001-2003
9.2.1	2.2	2.0	2001-2003
9.2.2	1.8	2.0	2001-2003
9.2.3	2.2	4.5	2001-2003
9.2.4	2.6	5.8	2001-2003
9.3.1	2.5	1.6	2001-2003
9.3.3	2.8	1.2	2001-2003
9.3.4	2.6	1.7	2001-2003
9.4.1	2.5	1.6	2001-2003
9.4.2	2.3	3.1	2001-2003
9.4.3	2.2	1.3	2001-2003
9.4.4	2.6	4.4	2001-2003
9.4.5	2.1	4.6	2001-2003
9.4.6	2.0	3.1	2001-2003
9.4.7	1.9	6.1	2001-2003
9.5.1	2.4	6.9	2001-2003
9.6.1	1.7	3.7	2001-2003
10.1.2	1.2	0.4	2006-2008
10.1.3	1.4	0.4	2006-2008
10.1.4	1.8	0.7	2006-2008
10.1.5	1.3	0.4	2006-2008
10.1.6	1.5	0.7	2006-2008
10.1.7	1.8	0.8	2006-2008
10.1.8	1.7	0.7	2006-2008
10.2.1	1.1	0.4	2006-2008
10.2.2	2.0	0.5	2006-2008
10.2.4	1.8	1.1	2006-2008
11.1.1	1.1	1.0	2006-2008
11.1.2	1.1	0.9	2006-2008
11.1.3	1.0	1.1	2006-2008
12.1.1	2.7	1.1	2006-2008
13.1.1	2.1	1.4	2006-2008
15.4.1	0.9	5.2	2006-2008
5.2.1	2.2	3.2	2006-2008
5.2.2	1.6	2.1	2006-2008

Ecoregion	EAQM-weighted annual SO_2 (ppb)	Median ecoregion S dep (kg S/ha-yr)	Period
5.3.1	3.0	5.8	2006-2008
5.3.3	6.1	15.1	2006-2008
6.2.10	1.8	1.1	2006-2008
6.2.11	1.1	1.1	2006-2008
6.2.12	1.1	1.1	2006-2008
6.2.13	1.6	1.4	2006-2008
6.2.14	1.7	1.2	2006-2008
6.2.15	1.5	1.2	2006-2008
6.2.3	1.4	1.0	2006-2008
6.2.4	1.5	1.3	2006-2008
6.2.5	1.2	1.6	2006-2008
6.2.7	1.2	1.7	2006-2008
6.2.8	1.1	0.5	2006-2008
6.2.9	1.2	0.5	2006-2008
7.1.7	1.2	1.6	2006-2008
7.1.8	1.1	2.1	2006-2008
7.1.9	1.3	1.5	2006-2008
8.1.1	4.8	8.8	2006-2008
8.1.10	5.7	15.1	2006-2008
8.1.3	4.8	10.2	2006-2008
8.1.4	1.9	3.4	2006-2008
8.1.5	2.3	5.0	2006-2008
8.1.6	3.5	8.3	2006-2008
8.1.7	3.4	8.4	2006-2008
8.1.8	2.9	4.6	2006-2008
8.2.1	3.0	6.4	2006-2008
8.2.2	4.2	8.6	2006-2008
8.2.3	3.3	9.0	2006-2008
8.2.4	4.0	12.0	2006-2008
8.3.1	5.2	12.6	2006-2008
8.3.2	3.5	9.3	2006-2008
8.3.3	4.1	11.0	2006-2008
8.3.4	3.5	9.6	2006-2008
8.3.5	2.7	8.1	2006-2008
8.3.6	3.2	6.7	2006-2008
8.3.7	2.3	6.8	2006-2008

Ecoregion	EAQM-weighted annual SO_2 (ppb)	Median ecoregion S dep (kg S/ha-yr)	Period
8.3.8	1.8	5.1	2006-2008
8.4.1	4.8	11.9	2006-2008
8.4.2	5.2	13.3	2006-2008
8.4.3	6.1	16.4	2006-2008
8.4.4	3.8	9.3	2006-2008
8.4.5	2.6	5.8	2006-2008
8.4.6	2.4	5.7	2006-2008
8.4.7	2.3	5.2	2006-2008
8.4.8	2.3	5.8	2006-2008
8.4.9	3.9	11.6	2006-2008
8.5.1	2.9	9.3	2006-2008
8.5.2	3.2	6.1	2006-2008
8.5.3	1.6	6.0	2006-2008
8.5.4	3.9	12.3	2006-2008
9.2.1	1.2	2.1	2006-2008
9.2.2	1.2	2.0	2006-2008
9.2.3	1.9	4.3	2006-2008
9.2.4	2.5	5.3	2006-2008
9.3.1	1.2	1.6	2006-2008
9.3.3	1.5	1.3	2006-2008
9.3.4	1.5	2.0	2006-2008
9.4.1	1.7	1.5	2006-2008
9.4.2	1.7	3.0	2006-2008
9.4.3	1.6	1.2	2006-2008
9.4.4	2.1	4.0	2006-2008
9.4.5	1.7	4.0	2006-2008
9.4.6	1.5	2.8	2006-2008
9.4.7	1.6	4.9	2006-2008
9.5.1	1.8	5.6	2006-2008
9.6.1	1.3	3.0	2006-2008
10.1.2	0.9	0.4	2010-2012
10.1.3	1.1	0.5	2010-2012
10.1.4	1.5	0.5	2010-2012
10.1.5	1.0	0.5	2010-2012
10.1.6	1.2	0.6	2010-2012
10.1.7	1.3	0.6	2010-2012

Ecoregion	EAQM-weighted annual SO_2 (ppb)	Median ecoregion S dep (kg S/ha-yr)	Period
10.1.8	1.2	0.6	2010-2012
10.2.1	0.8	0.4	2010-2012
10.2.2	1.4	0.5	2010-2012
10.2.4	1.3	1.1	2010-2012
11.1.1	0.8	0.9	2010-2012
11.1.2	0.8	0.8	2010-2012
11.1.3	0.7	1.1	2010-2012
12.1.1	2.2	0.9	2010-2012
13.1.1	1.6	1.2	2010-2012
15.4.1	0.6	4.2	2010-2012
5.2.1	1.5	2.4	2010-2012
5.2.2	1.1	1.5	2010-2012
5.3.1	2.0	3.0	2010-2012
5.3.3	3.2	7.2	2010-2012
6.2.10	1.3	0.9	2010-2012
6.2.11	0.9	1.0	2010-2012
6.2.12	0.8	1.2	2010-2012
6.2.13	1.3	1.2	2010-2012
6.2.14	1.2	0.9	2010-2012
6.2.15	1.1	1.1	2010-2012
6.2.3	1.0	0.8	2010-2012
6.2.4	1.1	1.0	2010-2012
6.2.5	0.9	1.3	2010-2012
6.2.7	0.9	1.4	2010-2012
6.2.8	0.8	0.5	2010-2012
6.2.9	0.9	0.5	2010-2012
7.1.7	1.0	1.4	2010-2012
7.1.8	0.9	2.0	2010-2012
7.1.9	0.9	1.4	2010-2012
8.1.1	2.7	4.0	2010-2012
8.1.10	3.2	8.1	2010-2012
8.1.3	2.7	4.8	2010-2012
8.1.4	1.3	2.6	2010-2012
8.1.5	1.4	3.4	2010-2012
8.1.6	2.1	5.3	2010-2012
8.1.7	1.7	3.8	2010-2012

Ecoregion	EAQM-weighted annual SO_2 (ppb)	Median ecoregion S dep (kg S/ha-yr)	Period
8.1.8	1.7	2.4	2010-2012
8.2.1	1.8	4.0	2010-2012
8.2.2	2.7	5.2	2010-2012
8.2.3	2.0	5.4	2010-2012
8.2.4	2.4	7.1	2010-2012
8.3.1	2.6	5.3	2010-2012
8.3.2	2.3	6.2	2010-2012
8.3.3	2.2	6.2	2010-2012
8.3.4	1.5	4.3	2010-2012
8.3.5	1.6	4.3	2010-2012
8.3.6	2.2	4.6	2010-2012
8.3.7	1.7	4.9	2010-2012
8.3.8	1.2	3.8	2010-2012
8.4.1	2.7	5.3	2010-2012
8.4.2	2.9	7.0	2010-2012
8.4.3	3.3	8.3	2010-2012
8.4.4	1.7	4.4	2010-2012
8.4.5	1.9	4.6	2010-2012
8.4.6	1.7	4.5	2010-2012
8.4.7	1.7	4.2	2010-2012
8.4.8	1.6	4.7	2010-2012
8.4.9	2.1	5.5	2010-2012
8.5.1	1.8	5.1	2010-2012
8.5.2	2.4	4.2	2010-2012
8.5.3	1.1	4.4	2010-2012
8.5.4	2.2	5.6	2010-2012
9.2.1	1.0	1.7	2010-2012
9.2.2	0.9	1.4	2010-2012
9.2.3	1.3	3.0	2010-2012
9.2.4	1.7	4.1	2010-2012
9.3.1	1.0	1.4	2010-2012
9.3.3	1.1	1.0	2010-2012
9.3.4	1.2	1.5	2010-2012
9.4.1	1.3	1.3	2010-2012
9.4.2	1.2	2.2	2010-2012
9.4.3	1.2	1.0	2010-2012

Ecoregion	EAQM-weighted annual SO_2 (ppb)	Median ecoregion S dep (kg S/ha-yr)	Period
9.4.4	1.5	2.9	2010-2012
9.4.5	1.1	3.0	2010-2012
9.4.6	1.1	2.2	2010-2012
9.4.7	1.0	3.8	2010-2012
9.5.1	1.2	4.3	2010-2012
9.6.1	0.9	2.5	2010-2012
10.1.2	0.7	0.5	2014-2016
10.1.3	0.9	0.5	2014-2016
10.1.4	1.4	0.6	2014-2016
10.1.5	0.7	0.5	2014-2016
10.1.6	1.0	0.6	2014-2016
10.1.7	1.5	0.6	2014-2016
10.1.8	1.0	0.6	2014-2016
10.2.1	0.6	0.4	2014-2016
10.2.2	1.5	0.4	2014-2016
10.2.4	1.6	1.2	2014-2016
11.1.1	0.6	0.8	2014-2016
11.1.2	0.8	0.8	2014-2016
11.1.3	0.4	1.0	2014-2016
12.1.1	2.3	0.9	2014-2016
13.1.1	2.0	1.0	2014-2016
15.4.1	0.4	4.3	2014-2016
5.2.1	1.1	1.9	2014-2016
5.2.2	0.8	1.1	2014-2016
5.3.1	1.0	2.0	2014-2016
5.3.3	1.7	4.1	2014-2016
6.2.10	1.1	0.9	2014-2016
6.2.11	0.6	1.1	2014-2016
6.2.12	0.8	1.1	2014-2016
6.2.13	0.9	1.3	2014-2016
6.2.14	1.4	0.8	2014-2016
6.2.15	0.8	0.9	2014-2016
6.2.3	0.8	0.8	2014-2016
6.2.4	0.8	1.0	2014-2016
6.2.5	0.6	1.4	2014-2016
6.2.7	0.7	1.5	2014-2016

Ecoregion	EAQM-weighted annual SO_2 (ppb)	Median ecoregion S dep (kg S/ha-yr)	Period
6.2.8	0.7	0.5	2014-2016
6.2.9	0.7	0.6	2014-2016
7.1.7	0.6	2.1	2014-2016
7.1.8	0.5	2.0	2014-2016
7.1.9	0.6	1.7	2014-2016
8.1.1	1.4	2.7	2014-2016
8.1.10	1.6	5.0	2014-2016
8.1.3	1.5	2.8	2014-2016
8.1.4	1.0	2.0	2014-2016
8.1.5	1.1	2.6	2014-2016
8.1.6	1.3	3.3	2014-2016
8.1.7	0.8	2.4	2014-2016
8.1.8	0.9	1.6	2014-2016
8.2.1	1.2	2.7	2014-2016
8.2.2	1.5	3.2	2014-2016
8.2.3	1.3	4.1	2014-2016
8.2.4	1.3	4.1	2014-2016
8.3.1	1.3	3.3	2014-2016
8.3.2	1.5	4.3	2014-2016
8.3.3	1.3	4.2	2014-2016
8.3.4	0.8	2.6	2014-2016
8.3.5	0.9	3.5	2014-2016
8.3.6	1.2	4.0	2014-2016
8.3.7	0.9	4.7	2014-2016
8.3.8	0.6	4.4	2014-2016
8.4.1	1.4	3.2	2014-2016
8.4.2	1.4	4.1	2014-2016
8.4.3	1.5	4.8	2014-2016
8.4.4	0.8	2.6	2014-2016
8.4.5	1.0	3.2	2014-2016
8.4.6	0.9	3.3	2014-2016
8.4.7	0.9	3.4	2014-2016
8.4.8	0.9	4.1	2014-2016
8.4.9	1.2	3.5	2014-2016
8.5.1	0.9	3.4	2014-2016
8.5.2	1.3	3.9	2014-2016

Ecoregion	EAQM-weighted annual SO_2 (ppb)	Median ecoregion S dep (kg S/ha-yr)	Period
8.5.3	0.8	3.9	2014-2016
8.5.4	1.1	3.8	2014-2016
9.2.1	0.8	1.3	2014-2016
9.2.2	0.7	1.2	2014-2016
9.2.3	0.9	2.6	2014-2016
9.2.4	0.9	3.0	2014-2016
9.3.1	0.9	1.2	2014-2016
9.3.3	1.0	0.9	2014-2016
9.3.4	1.3	1.4	2014-2016
9.4.1	1.4	1.3	2014-2016
9.4.2	1.0	2.2	2014-2016
9.4.3	1.4	1.1	2014-2016
9.4.4	0.9	2.5	2014-2016
9.4.5	0.6	3.1	2014-2016
9.4.6	0.8	2.5	2014-2016
9.4.7	0.6	4.0	2014-2016
9.5.1	0.6	4.7	2014-2016
9.6.1	0.6	3.1	2014-2016
10.1.2	0.6	0.3	2018-2020
10.1.3	0.8	0.3	2018-2020
10.1.4	1.2	0.4	2018-2020
10.1.5	0.6	0.3	2018-2020
10.1.6	0.7	0.3	2018-2020
10.1.7	0.9	0.3	2018-2020
10.1.8	0.8	0.4	2018-2020
10.2.1	0.6	0.3	2018-2020
10.2.2	0.9	0.3	2018-2020
10.2.4	1.3	0.9	2018-2020
11.1.1	0.5	0.7	2018-2020
11.1.2	0.6	0.7	2018-2020
11.1.3	0.5	0.8	2018-2020
12.1.1	1.4	0.5	2018-2020
13.1.1	1.1	0.6	2018-2020
15.4.1	0.7	3.8	2018-2020
5.2.1	0.7	1.3	2018-2020
5.2.2	0.8	0.9	2018-2020

Ecoregion	EAQM-weighted annual SO_2 (ppb)	Median ecoregion S dep (kg S/ha-yr)	Period
5.3.1	0.6	1.3	2018-2020
5.3.3	0.9	2.4	2018-2020
6.2.10	1.1	0.7	2018-2020
6.2.11	0.5	0.9	2018-2020
6.2.12	0.5	1.0	2018-2020
6.2.13	0.7	0.8	2018-2020
6.2.14	0.9	0.5	2018-2020
6.2.15	0.6	0.6	2018-2020
6.2.3	0.8	0.5	2018-2020
6.2.4	0.8	0.8	2018-2020
6.2.5	0.9	1.1	2018-2020
6.2.7	0.6	1.2	2018-2020
6.2.8	0.6	0.5	2018-2020
6.2.9	0.6	0.4	2018-2020
7.1.7	0.9	1.2	2018-2020
7.1.8	0.5	1.5	2018-2020
7.1.9	0.5	1.1	2018-2020
8.1.1	0.8	1.6	2018-2020
8.1.10	0.8	2.8	2018-2020
8.1.3	0.7	1.7	2018-2020
8.1.4	0.7	1.4	2018-2020
8.1.5	0.8	1.9	2018-2020
8.1.6	0.8	2.2	2018-2020
8.1.7	0.5	1.9	2018-2020
8.1.8	0.5	1.2	2018-2020
8.2.1	0.7	2.0	2018-2020
8.2.2	0.9	2.1	2018-2020
8.2.3	0.9	2.4	2018-2020
8.2.4	1.2	2.6	2018-2020
8.3.1	0.8	2.1	2018-2020
8.3.2	1.4	3.0	2018-2020
8.3.3	1.4	2.7	2018-2020
8.3.4	0.8	1.9	2018-2020
8.3.5	0.8	2.6	2018-2020
8.3.6	1.9	3.2	2018-2020
8.3.7	1.1	3.6	2018-2020

Ecoregion	EAQM-weighted annual SO_2 (ppb)	Median ecoregion S dep (kg S/ha-yr)	Period
8.3.8	0.6	3.6	2018-2020
8.4.1	1.1	2.1	2018-2020
8.4.2	1.0	2.3	2018-2020
8.4.3	1.0	2.9	2018-2020
8.4.4	0.9	1.9	2018-2020
8.4.5	1.4	2.6	2018-2020
8.4.6	1.1	2.8	2018-2020
8.4.7	1.1	3.0	2018-2020
8.4.8	1.1	3.5	2018-2020
8.4.9	1.2	2.6	2018-2020
8.5.1	0.7	2.4	2018-2020
8.5.2	1.9	3.2	2018-2020
8.5.3	0.8	3.2	2018-2020
8.5.4	0.6	2.7	2018-2020
9.2.1	0.9	1.2	2018-2020
9.2.2	0.9	1.1	2018-2020
9.2.3	0.8	1.9	2018-2020
9.2.4	0.7	2.3	2018-2020
9.3.1	1.0	1.1	2018-2020
9.3.3	1.0	0.8	2018-2020
9.3.4	1.1	1.4	2018-2020
9.4.1	1.1	1.0	2018-2020
9.4.2	1.0	1.8	2018-2020
9.4.3	1.1	0.6	2018-2020
9.4.4	0.8	1.9	2018-2020
9.4.5	0.7	2.6	2018-2020
9.4.6	0.8	2.1	2018-2020
9.4.7	0.6	3.4	2018-2020
9.5.1	0.6	4.3	2018-2020
9.6.1	0.7	2.4	2018-2020

6A.5. IMPACTS OF THREE KEY ASPECTS OF METHODOLOGY ON FINDINGS

As noted earlier in this appendix, three aspects of the analytical methodology used to compare upwind air quality (EAQM) and downwind deposition in an ecoregion were examined with regard to their influence on analysis findings. Specifically, we examined two durations for the forward parcel trajectories (48-hours and 120-hours), two different meteorological input data sets (NARR-32 and NAM-12) with differing resolution, and three different monitor inclusion criteria (hit rates) ranging from from 1% of total hits to 0.1% of total hits. Each of these methodological changes, when moving from the original analysis to the final analysis, had the effect of allowing more distant upwind sites to be included in the EAQM calculations of air quality across potential sites of influence. Again, like other elements of the EAQM analysis, these methodological assumptions about the potential scope of the sites of influence introduce uncertainty. Sensitivity analyses were performed to evaluate the effect of these changes (length of trajectory plus finer resolution meteorological data, hit threshold) on what the EAQM approach concluded about the association between upwind air quality and downwind deposition.

Figure 6A-21 shows the association between annual SO_2 EAQM values and S deposition across the 84 ecoregions and 5 time periods, based on a 48-hour duration for the trajectory analysis, the NARR-32 inputs, and a monitor inclusion criterion of 1%. Figure 6A-22 shows the association between annual SO_2 EAQM values and S deposition across the 84 ecoregions and 5 time periods, based on 120-hour duration for the trajectory analysis, the NAM-12 input data, and a minimum hit rate of 0.5% for monitoring site inclusion criterion. In both analyses, similar themes emerge. It is clear from both figures that the EAQM SO_2 and TDep S deposition association is strongest for the 47 eastern ecoregions and their upwind monitoring sites of influence, and essentially non-existent for the 37 western ecoregions and their upwind monitoring sites of influence. In both cases, we can conclude that the relationship between upwind air quality and downwind deposition was stronger in the earlier periods than the most recent 2018-2020 period. It can be noted that the r-value improves slightly with the inclusion of more distant sites (i.e., the final analysis configuration), from 0.45 to 0.56. Figures 6A-23 and 6A-24 limit the EAQM-TDep comparisons to sites in the eastern U.S. and the associations are equally strong in both iterations of the methodology (r-values = 0.85, slopes ~ 2.2). We also looked at how the results varied by methodology for other associations (e.g., annual NO_2 and N deposition) and concluded that the overall strength of association between upwind air quality and downwind deposition were not strongly affected by the choice of trajectory length, meteorological inputs, or monitor inclusion criteria. All of the outputs, both original analysis and final analysis (for 3 different monitor inclusion criteria) are shown in 6A.6.

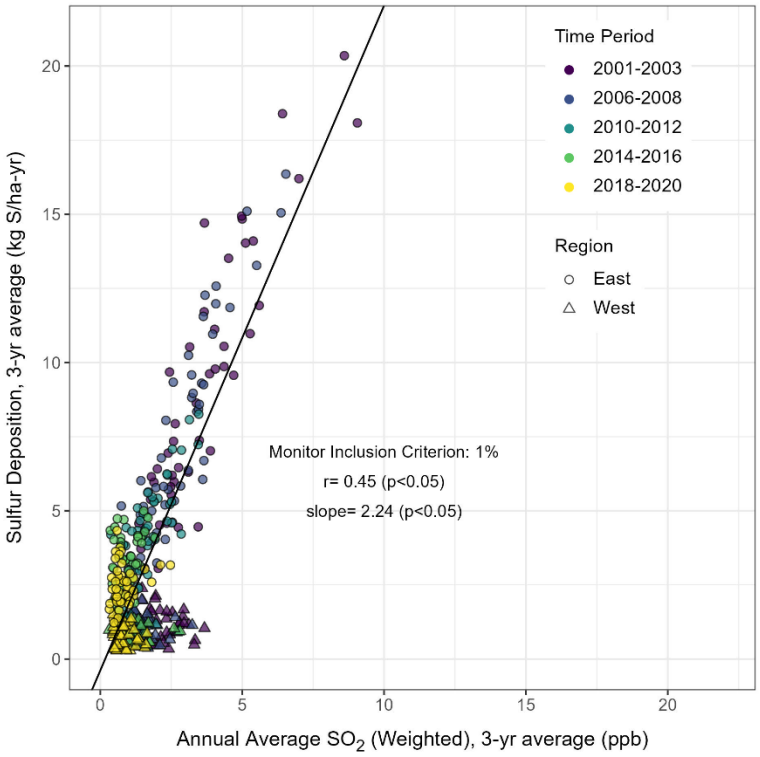

Figure 6A-21. Annual SO_2 EAQM-weighted values and TDep S deposition in 84 ecoregions (48-hr trajectories, NARR-32, 1% monitor inclusion criteria).

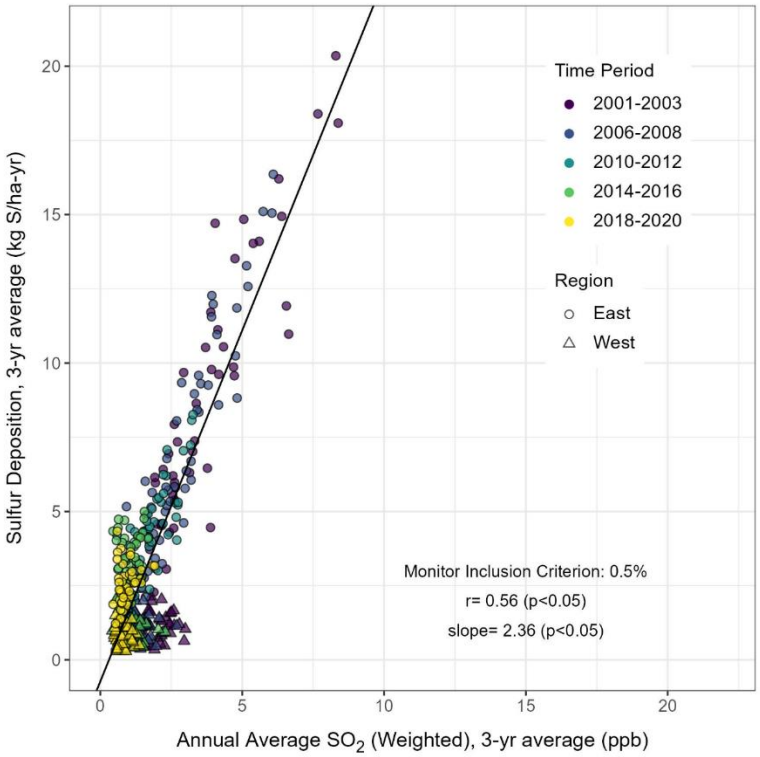

Figure 6A-22. Annual SO_2 EAQM-weighted values and TDep S deposition in 84 ecoregions (120-hr trajectories, NAM-12, 0.5% monitor inclusion criteria).

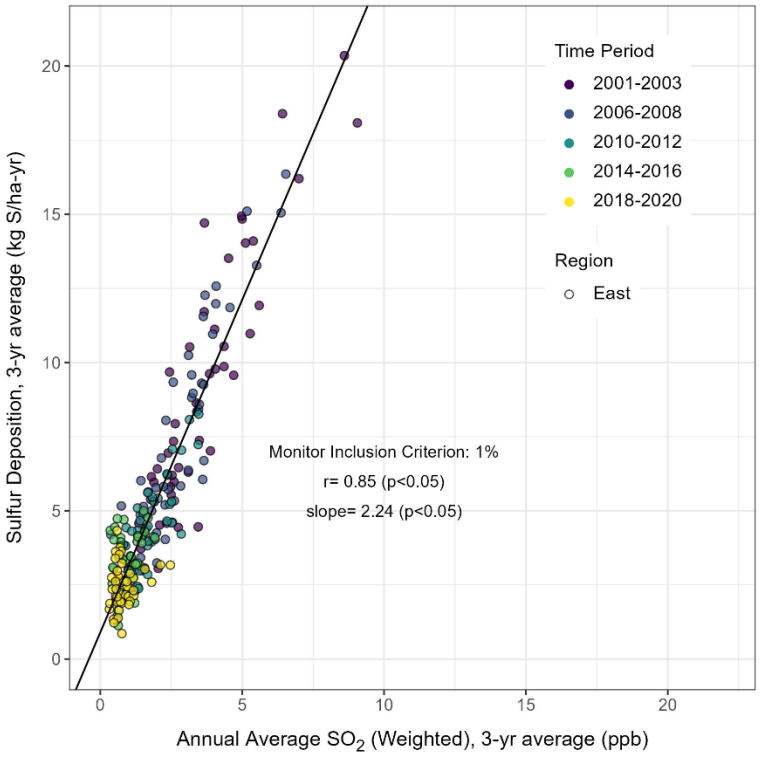

Figure 6A-23. Annual SO_2 EAQM-weighted values and TDep S deposition in eastern ecoregions (48-hr trajectories, NARR-32, 1% monitor inclusion criteria).

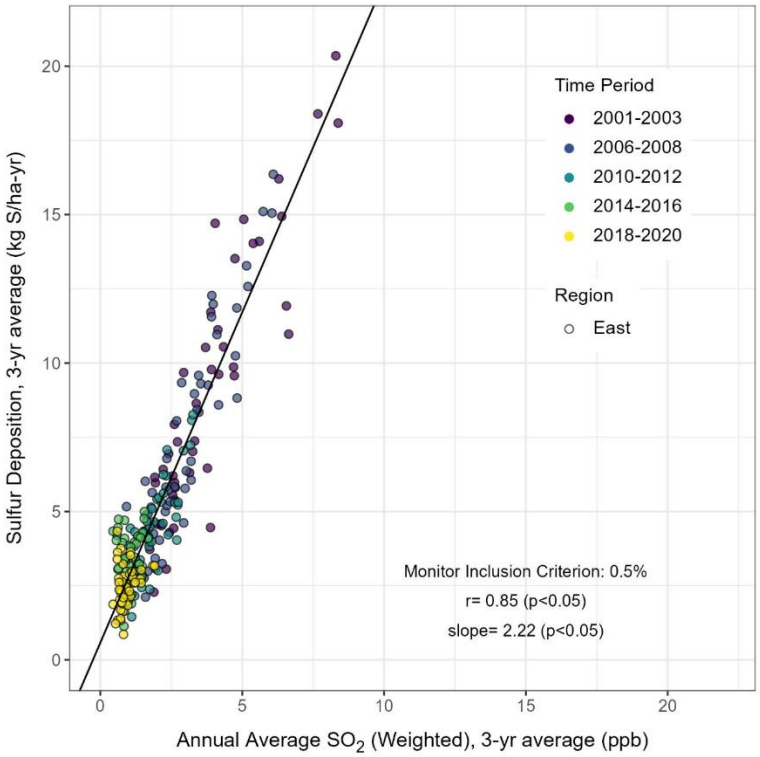

Figure 6A-24. Annual SO_2 EAQM-weighted values and TDep S deposition in eastern ecoregions (120-hr trajectories, NAM-12, 0.5% monitor inclusion criteria).

6A.6. RESULTS OF HYSPLIT EAQM ANALYSES

6A.6.1. SO_2 3-hr Metric – 120-hr

Table 6A-2. Correlation coefficients of TDep-estimated S deposition and 3-hr SO_2 EAQMs generated by HYSPLIT analysis at three monitor inclusion criteria, 120-hr trajectories.

Sulfur Deposition and SO_2 (3-hr Standard)					
3-hr Max-All Ecoregions- Monitor Inclusion Criteria: 1%	Correlation Coefficient (r) = 0.29*	*3-hr Max-All Ecoregions- Monitor Inclusion Criteria: 0.5%*	Correlation Coefficient (r) = 0.51*	*3-hr Max-All Ecoregions- Monitor Inclusion Criteria: 0.1%*	Correlation Coefficient (r) = 0.50*
Year	r	Year	r	Year	r
2001 - 2003	0.12	2001 - 2003	0.49*	2001 - 2003	0.84*
2006 - 2008	0.49*	2006 - 2008	0.69*	2006 - 2008	0.84*
2010 - 2012	0.25*	2010 - 2012	0.25*	2010 - 2012	-0.12
2014 - 2016	0.15	2014 - 2016	0.23*	2014 - 2016	0.57*
2018 - 2020	0.17	2018 - 2020	0.54*	2018 - 2020	0.76*
3-hr Max-All Ecoregions- Monitor Inclusion Criteria: 1%[a]	r = 0.29*	*3-hr Max-All Ecoregions- Monitor Inclusion Criteria: 0.5%*[a]	r = 0.52*	*3-hr Max-All Ecoregions- Monitor Inclusion Criteria: 0.1%*[a]	r = 0.59*
Year	r	Year	r	Year	r
2001 - 2003	0.12	2001 - 2003	0.49*	2001 - 2003	0.84*
2006 - 2008	0.49*	2006 - 2008	0.69*	2006 - 2008	0.84*
2010 - 2012	0.25*	2010 - 2012	0.25*	2010 - 2012	-0.12
2014 - 2016	0.15	2014 - 2016	0.23*	2014 - 2016	0.57*
2018 - 2020	0.10	2018 - 2020	0.40*	2018 - 2020	-0.09*
3-hr Max-East Ecoregions- Monitor Inclusion Criteria: 1%	r = 0.42*	*3-hr Max-East Ecoregions- Monitor Inclusion Criteria: 0.5%*	r = 0.32*	*3-hr Max-East Ecoregions- Monitor Inclusion Criteria: 0.1%*	r = -0.11
Year	r	Year	r	Year	r
2001 - 2003	0.29	2001 - 2003	0.28	2001 - 2003	0.63*
2006 - 2008	0.26	2006 - 2008	0.05	2006 - 2008	0.56*
2010 - 2012	0.05	2010 - 2012	-0.29*	2010 - 2012	-0.18
2014 - 2016	0.15	2014 - 2016	-0.38*	2014 - 2016	-0.18
2018 - 2020	0.18	2018 - 2020	0.33*	2018 - 2020	NA
3-hr Max-East Ecoregions- Monitor Inclusion Criteria: 1%[a]	r = 0.45*	*3-hr Max-East Ecoregions- Monitor Inclusion Criteria: 0.5%*[a]	r = 0.42*	*3-hr Max-East Ecoregions- Monitor Inclusion Criteria: 0.1%*[a]	r = 0.47*
Year	r	Year	r	Year	r
2001 - 2003	0.29	2001 - 2003	0.28	2001 - 2003	0.63*
2006 - 2008	0.26	2006 - 2008	0.05	2006 - 2008	0.56*
2010 - 2012	0.06	2010 - 2012	-0.29*	2010 - 2012	-0.18
2014 - 2016	0.15	2014 - 2016	-0.38*	2014 - 2016	-0.18
2018 - 2020	0.11	2018 - 2020	0.04	2018 - 2020	NA
3-hr Max-West Ecoregions- Monitor Inclusion Criteria: 1%	r = -0.01	*3-hr Max-West Ecoregions- Monitor Inclusion Criteria: 0.5%*	r = 0.07	*3-hr Max-West Ecoregions- Monitor Inclusion Criteria: 0.1%*	r = -0.01
Year	r	Year	r	Year	r
2001 - 2003	-0.18	2001 - 2003	-0.06	2001 - 2003	0.06

2006 - 2008	0.06	2006 - 2008	0.18	2006 - 2008	0.15
2010 - 2012	-0.16	2010 - 2012	-0.09	2010 - 2012	0.09
2014 - 2016	-0.15	2014 - 2016	-0.09	2014 - 2016	-0.14
2018 - 2020	0.21	2018 - 2020	0.10	2018 - 2020	-0.03
3-hr Max-West Ecoregions- Monitor Inclusion Criteria: 1% [a]	No outliers in West dataset	*3-hr Max-West Ecoregions- Monitor Inclusion Criteria: 0.5%* [a]	No outliers in West dataset	*3-hr Max-West Ecoregions- Monitor Inclusion Criteria: 0.1%* [a]	r = -0.02
Year	r	Year	r	Year	r
2001 - 2003		2001 - 2003		2001 - 2003	0.06
2006 - 2008		2006 - 2008		2006 - 2008	0.15
2010 - 2012		2010 - 2012		2010 - 2012	0.09
2014 - 2016		2014 - 2016		2014 - 2016	-0.14
2018 - 2020		2018 - 2020		2018 - 2020	-0.15
Weighted 3-hr Average-All Ecoregions- Monitor Inclusion Criteria: 1%	r = 0.60*	*Weighted 3-hr Average-All Ecoregions- Monitor Inclusion Criteria: 0.5%*	r = 0.70*	*Weighted 3-hr Average-All Ecoregions- Monitor Inclusion Criteria: 0.1%*	r = 0.72*
Year	r	Year	r	Year	r
2001 - 2003	0.74*	2001 - 2003	0.86*	2001 - 2003	0.90*
2006 - 2008	0.81*	2006 - 2008	0.89*	2006 - 2008	0.91*
2010 - 2012	0.64*	2010 - 2012	0.77*	2010 - 2012	0.81*
2014 - 2016	0.34*	2014 - 2016	0.38*	2014 - 2016	0.42*
2018 - 2020	0.38*	2018 - 2020	0.54*	2018 - 2020	0.58*
Weighted 3-hr Average-East Ecoregions- Monitor Inclusion Criteria: 1%	r = 0.77*	*Weighted 3-hr Average-East Ecoregions- Monitor Inclusion Criteria: 0.5%*	r = 0.83*	*Weighted 3-hr Average-East Ecoregions- Monitor Inclusion Criteria: 0.1%*	r = 0.84*
Year	r	Year	r	Year	r
2001 - 2003	0.70*	2001 - 2003	0.86*	2001 - 2003	0.92*
2006 - 2008	0.63*	2006 - 2008	0.78*	2006 - 2008	0.86*
2010 - 2012	0.57*	2010 - 2012	0.76*	2010 - 2012	0.78*
2014 - 2016	0.43*	2014 - 2016	0.24	2014 - 2016	0.24
2018 - 2020	0.32*	2018 - 2020	0.41*	2018 - 2020	0.43*
Weighted 3-hr Average-West Ecoregions- Monitor Inclusion Criteria: 1%	r = 0.19*	*Weighted 3-hr Average-West Ecoregions- Monitor Inclusion Criteria: 0.5%*	r = 0.20*	*Weighted 3-hr Average-West Ecoregions- Monitor Inclusion Criteria: 0.1%*	r = 0.21*
Year	r	Year	r	Year	r
2001 - 2003	0.16	2001 - 2003	0.15	2001 - 2003	0.15
2006 - 2008	0.23	2006 - 2008	0.31	2006 - 2008	0.32
2010 - 2012	0.05	2010 - 2012	0.06	2010 - 2012	0.07
2014 - 2016	-0.17	2014 - 2016	-0.16	2014 - 2016	-0.19
2018 - 2020	0.15	2018 - 2020	0.15	2018 - 2020	0.12

*p< 0.05

[a] Note: There are several outlier points in this comparison where the EAQM-max annual average SO_2 value exceeds 20 ppb in the 2018-2020 period. These points have been removed from these analyses. These data are driven by a monitor in southeastern MO where annual average SO_2 has exceeded 20 ppb in recent years. Any downwind ecoregion that is linked to this upwind monitor will have an EAQM-max with this value. A preliminary analysis suggests that these observed SO_2 data are due to a new source that was not modeled in the CMAQ simulation that informed the TDep estimates of deposition. As there is no deposition monitor in the immediate vicinity of the source it is unlikely that the TDep estimates are capturing the impacts of this source. For that reason, we concluded it was appropriate not to consider these data in our evaluation of the concentration-deposition relationship.

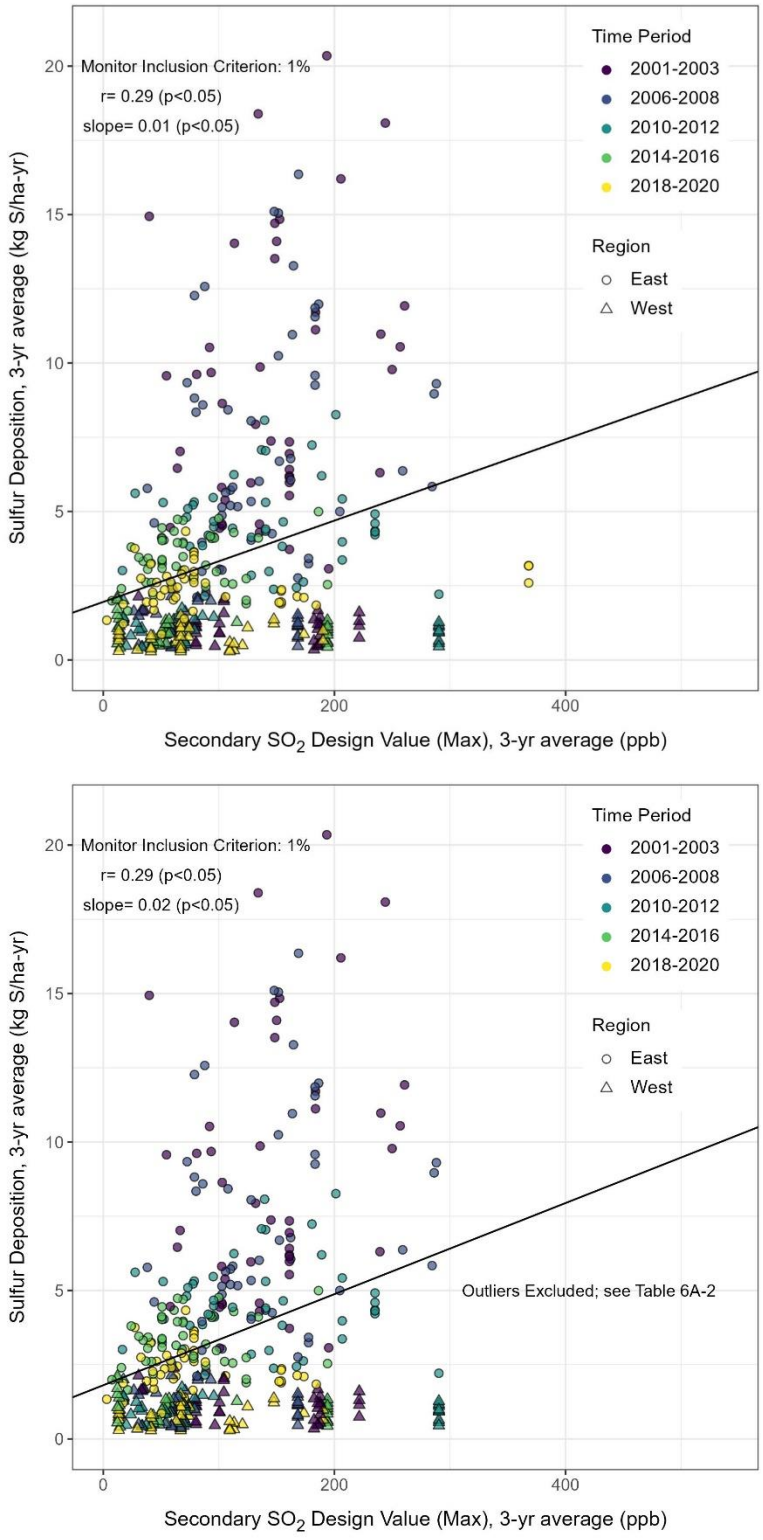

Figure 6A-25. The 3-hr SO_2 EAQM-max values and TDep S deposition in 84 ecoregions (120-hr trajectories, NAM-12, 1% monitor inclusion criteria): all values (upper), outliers excluded (lower).

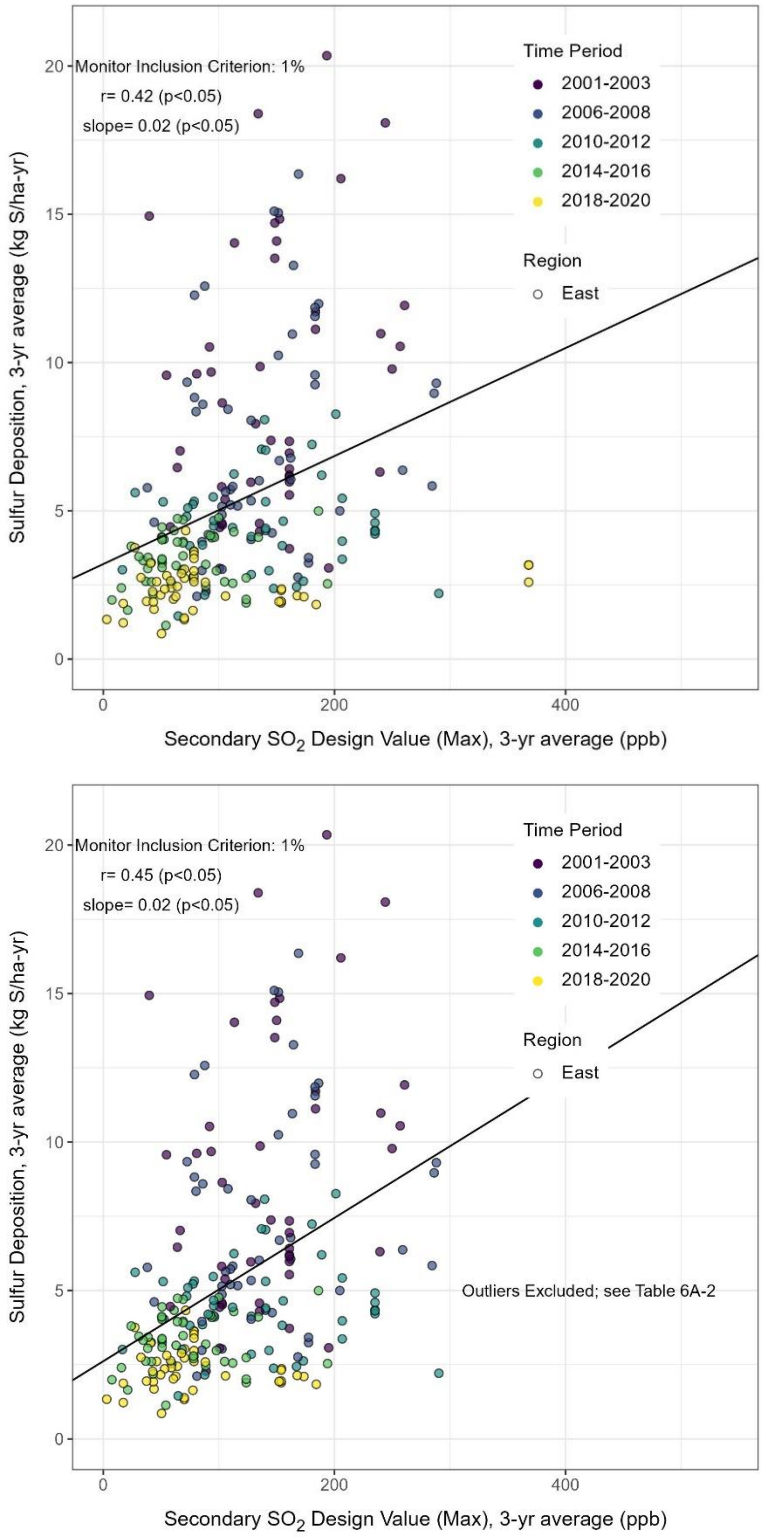

Figure 6A-26. The 3-hr SO_2 EAQM-max values and TDep S deposition in eastern ecoregions (120-hr trajectories, NAM-12, 1% monitor inclusion criteria): all values (upper), outliers excluded (lower).

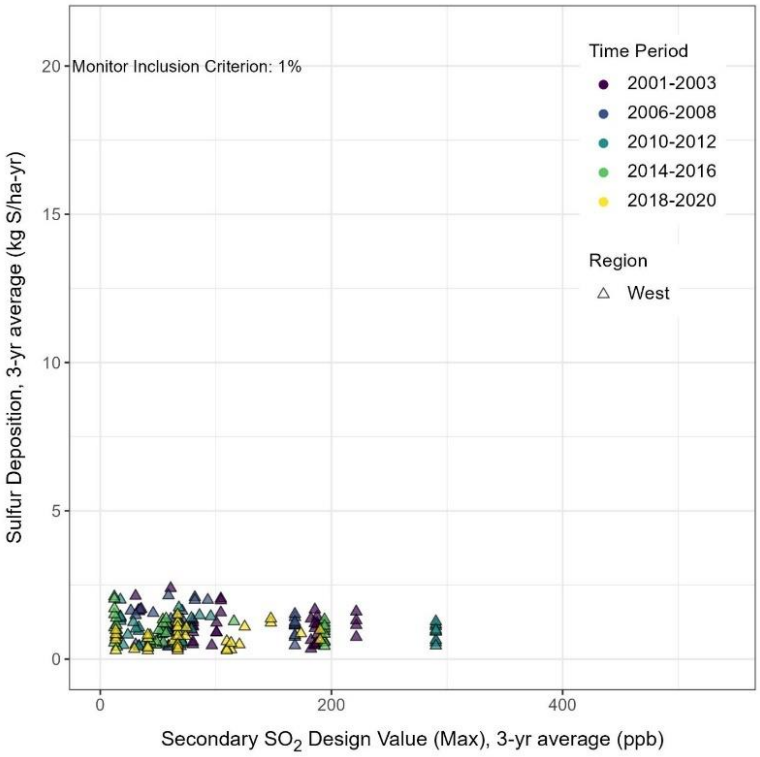

Figure 6A-27. The 3-hr SO_2 EAQM-max values and TDep S deposition in western ecoregions (120-hr trajectories, NAM-12, 1% monitor inclusion criteria).

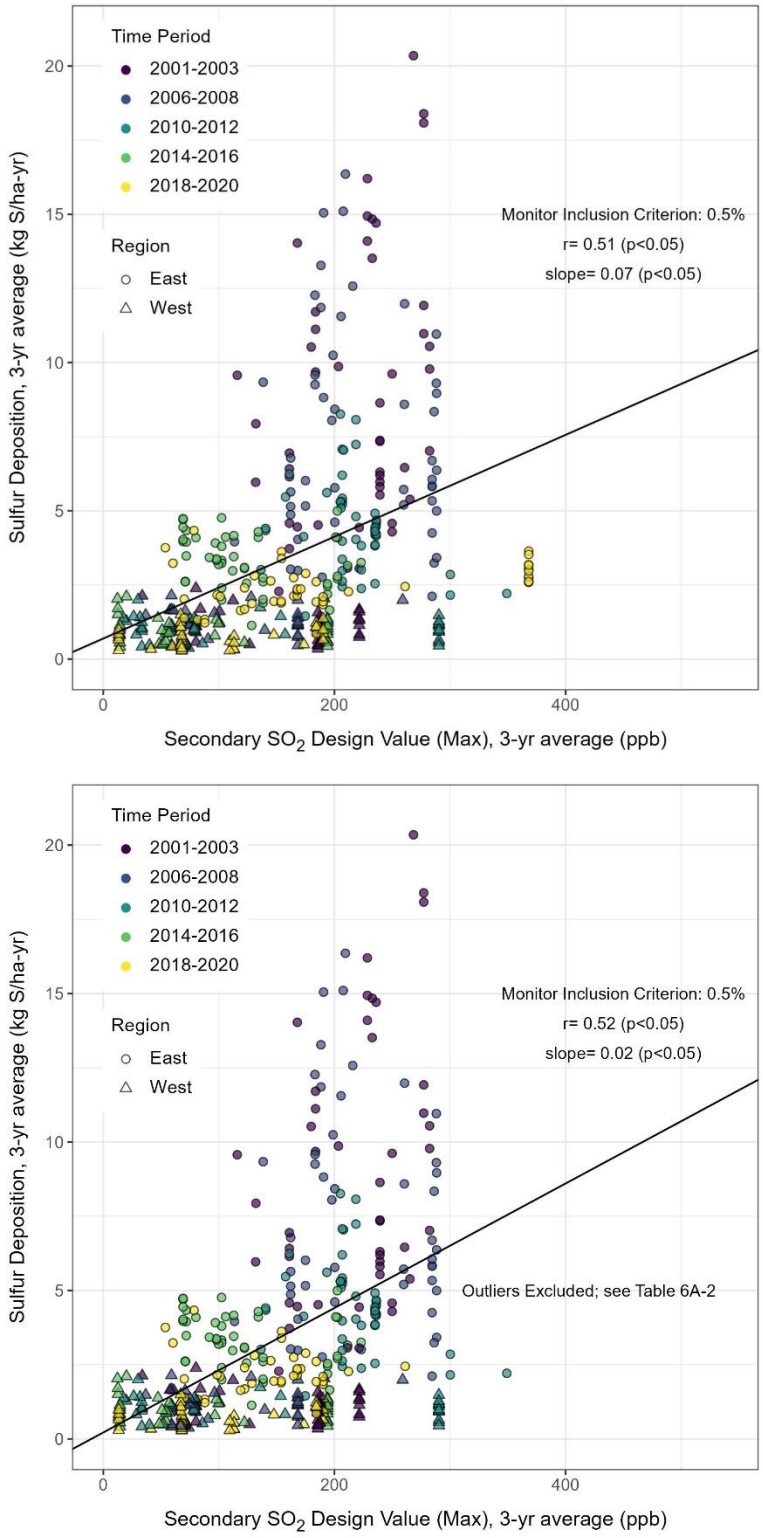

Figure 6A-28. The 3-hr SO_2 EAQM-max values and TDep S deposition in 84 ecoregions (120-hr trajectories, NAM-12, 0.5% monitor inclusion criteria): all values (upper), outliers excluded (lower).

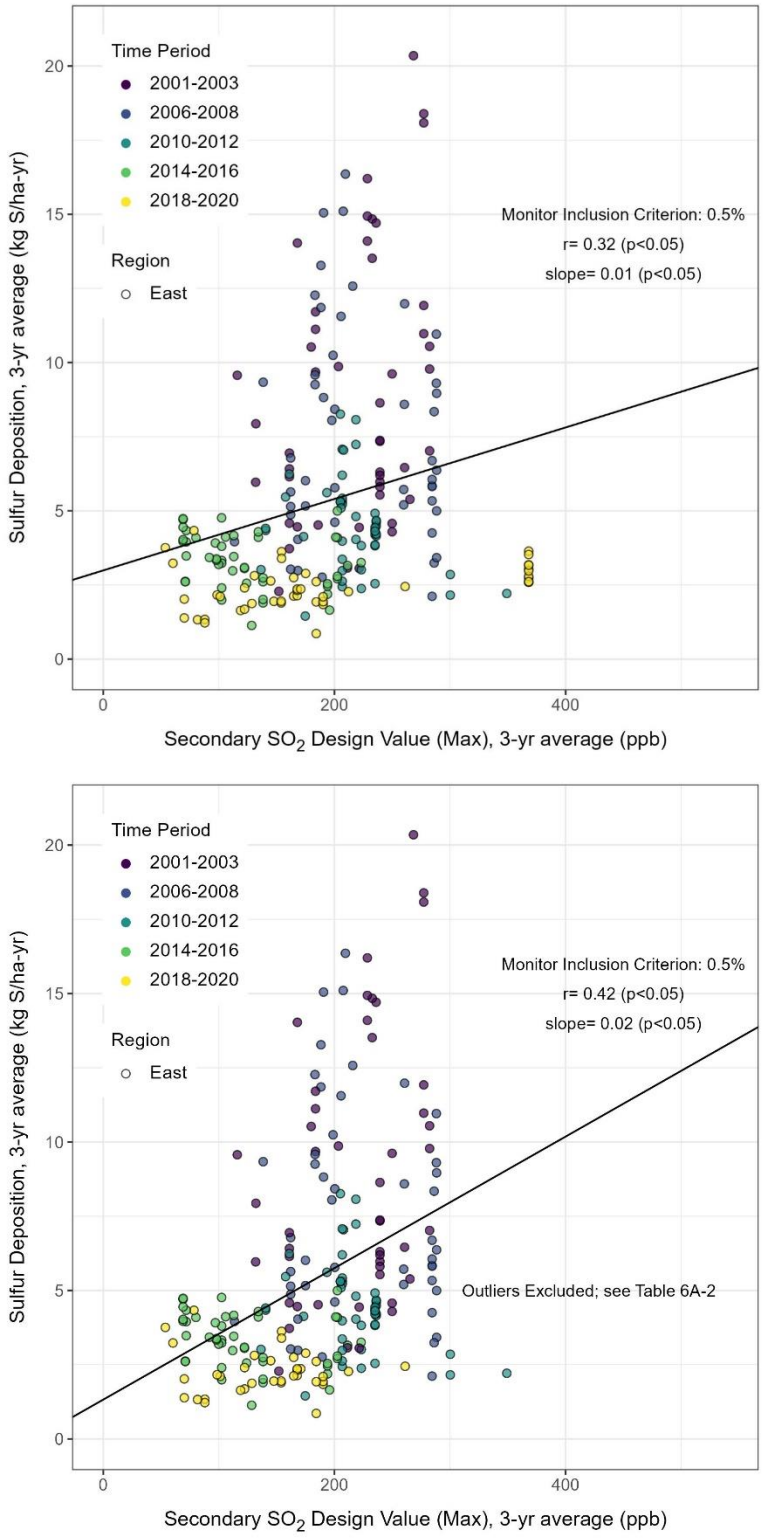

Figure 6A-29. The 3-hr SO_2 EAQM-max values and TDep S deposition in eastern ecoregions (120-hr trajectories, NAM-12, 0.5% monitor inclusion criteria): all values (upper), outliers excluded (lower).

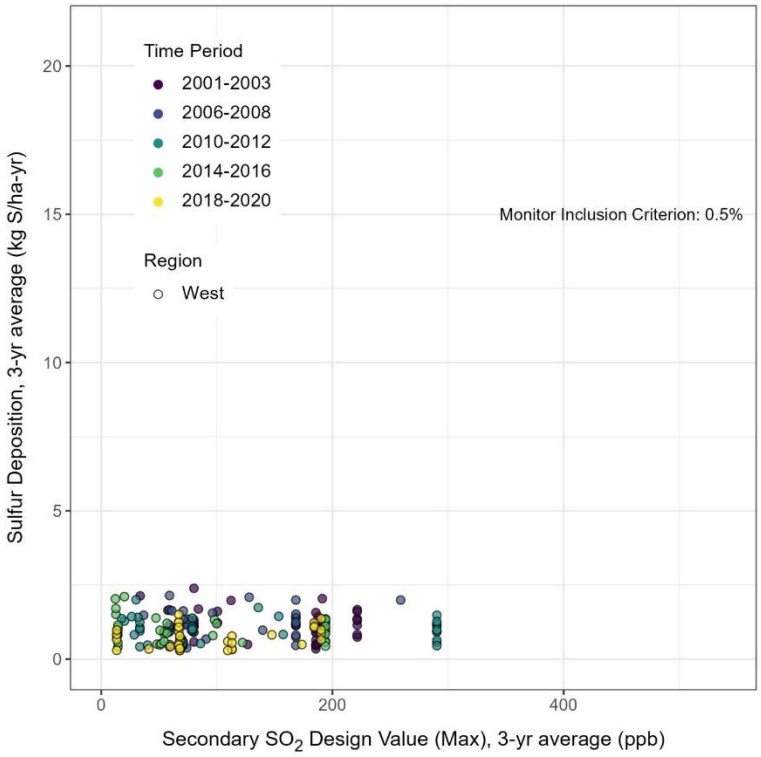

Figure 6A-30. The 3-hr SO_2 EAQM-max values and TDep S deposition in western ecoregions (120-hr trajectories, NAM-12, 0.5% monitor inclusion criteria).

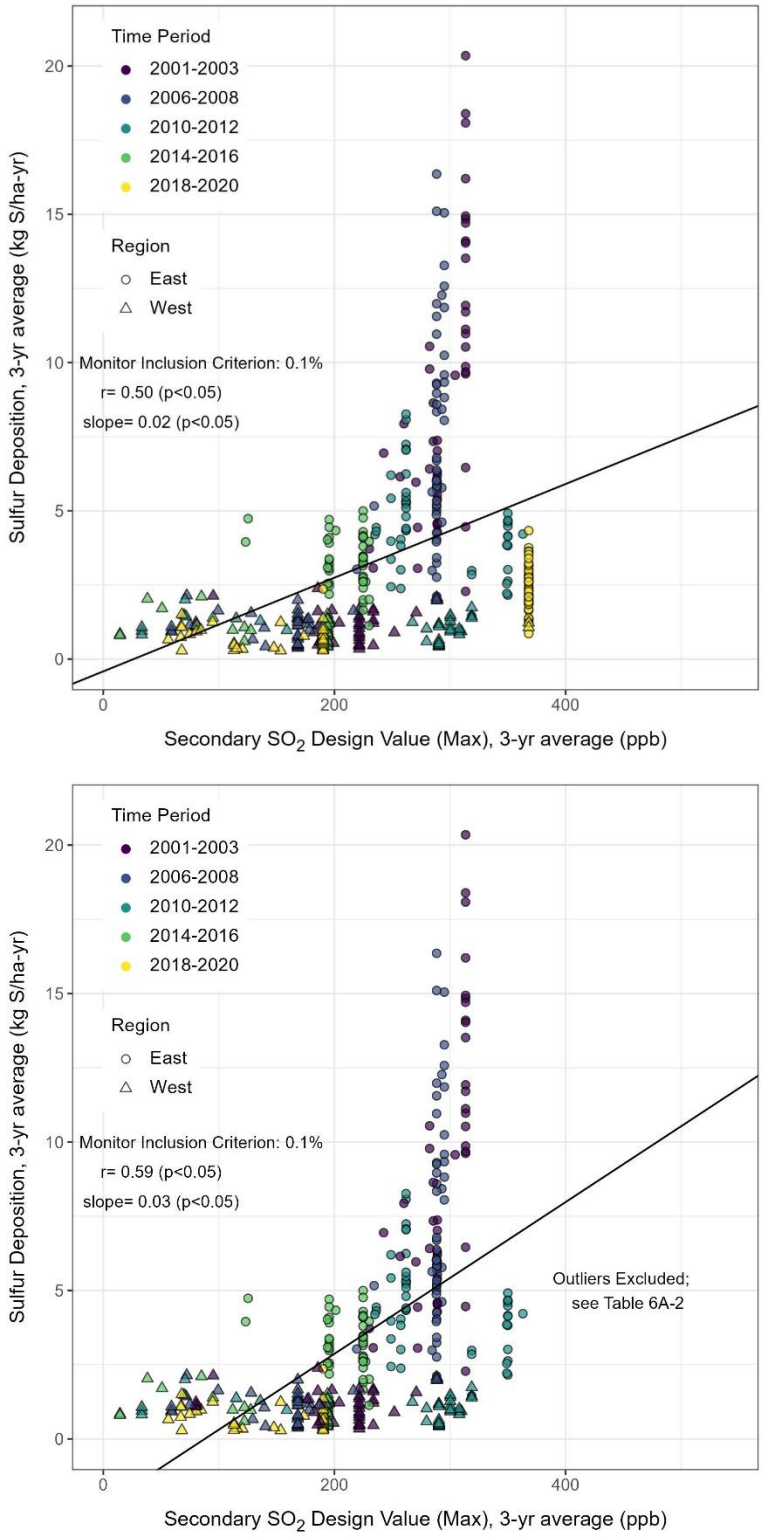

Figure 6A-31. The 3-hr SO_2 EAQM-max values and TDep S deposition in 84 ecoregions (120-hr trajectories, NAM-12, 0.1% monitor inclusion criteria): all values (upper), outliers excluded (lower).

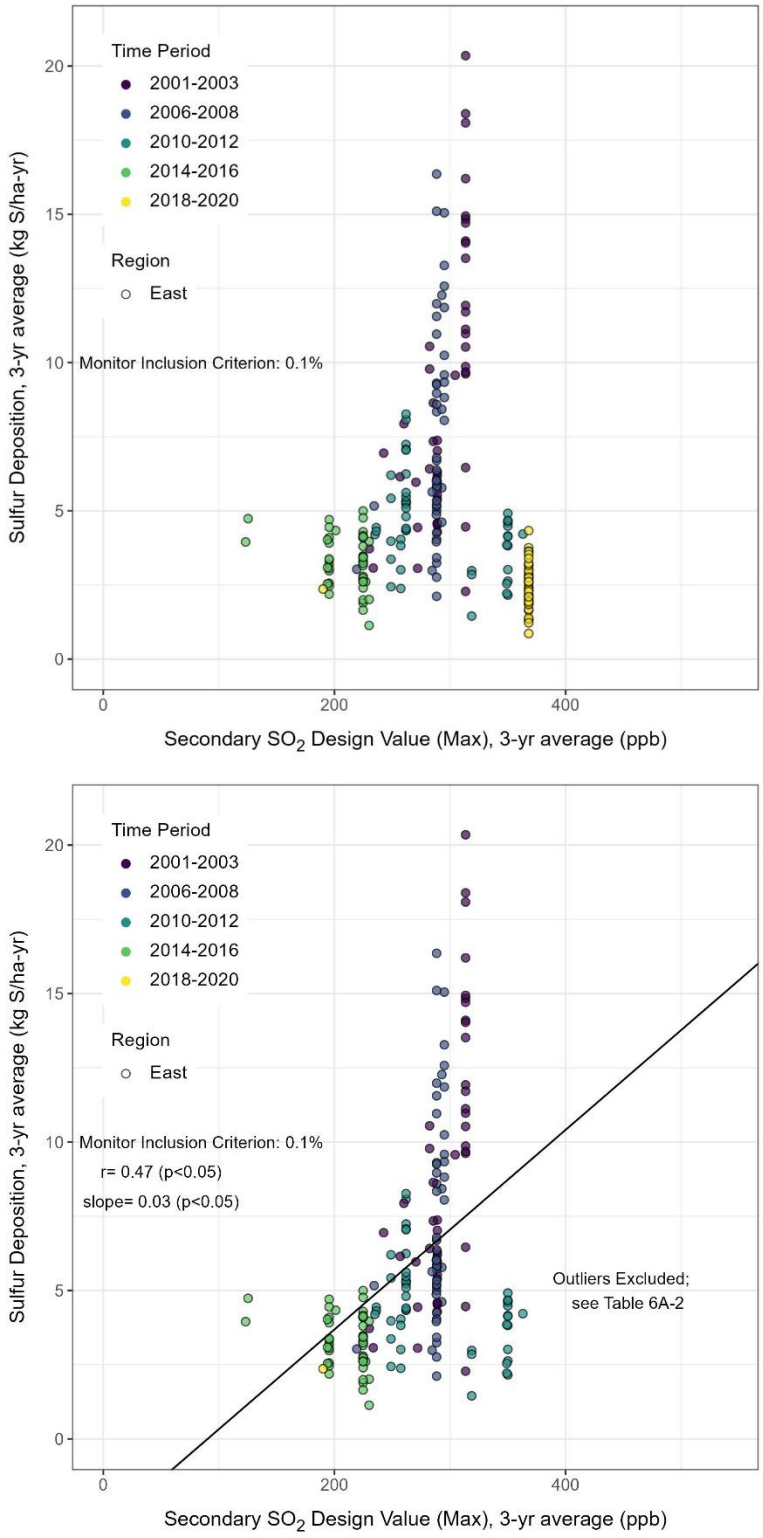

Figure 6A-32. The 3-hr SO_2 EAQM-max values and TDep S deposition in eastern ecoregions (120-hr trajectories, NAM-12, 0.1% monitor inclusion criteria): all values (upper), outliers excluded (lower).

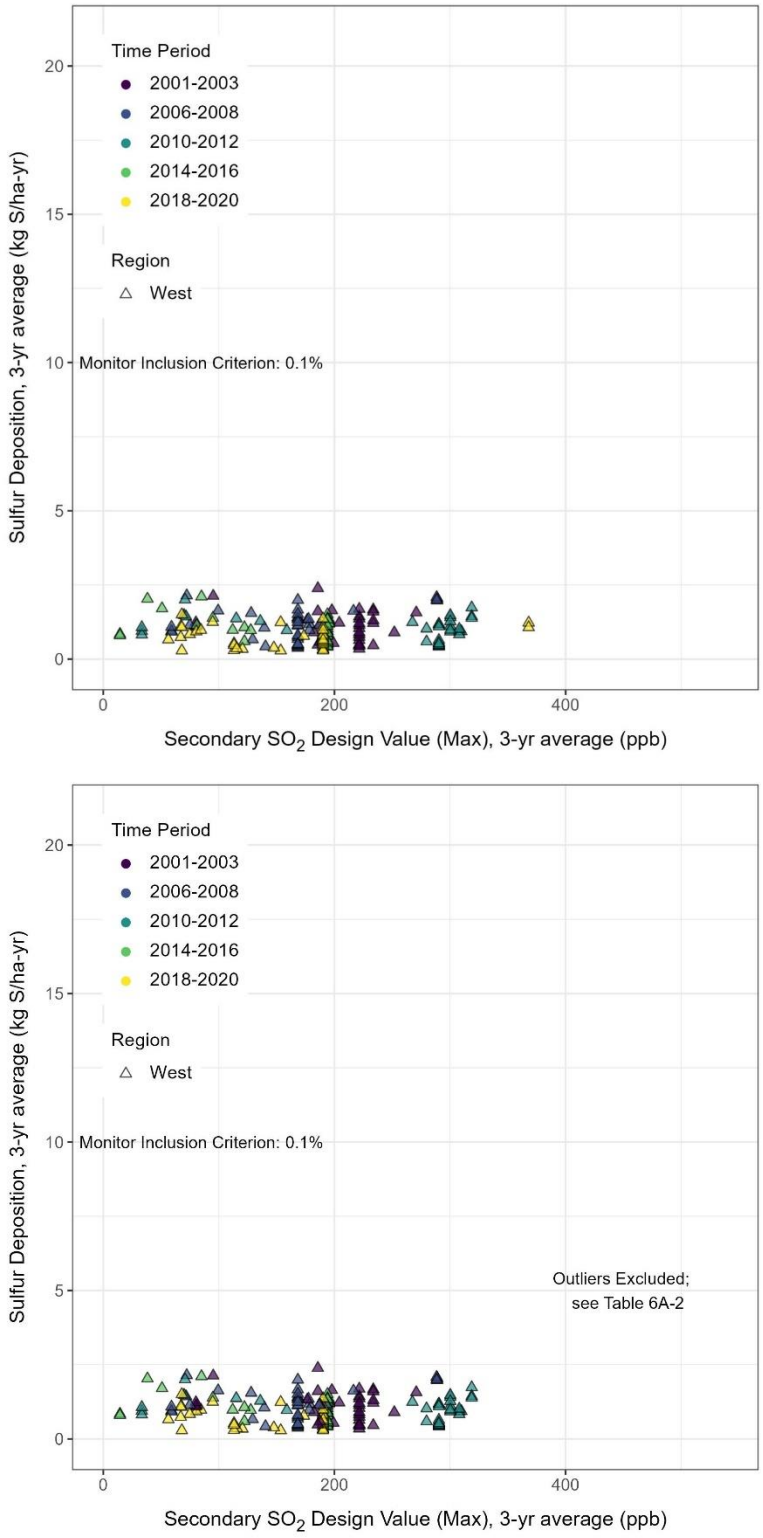

Figure 6A-33. The 3-hr SO_2 EAQM-max values and TDep S deposition in western ecoregions (120-hr trajectories, NAM-12, 0.1% monitor inclusion criteria): all values (upper), outliers excluded (lower).

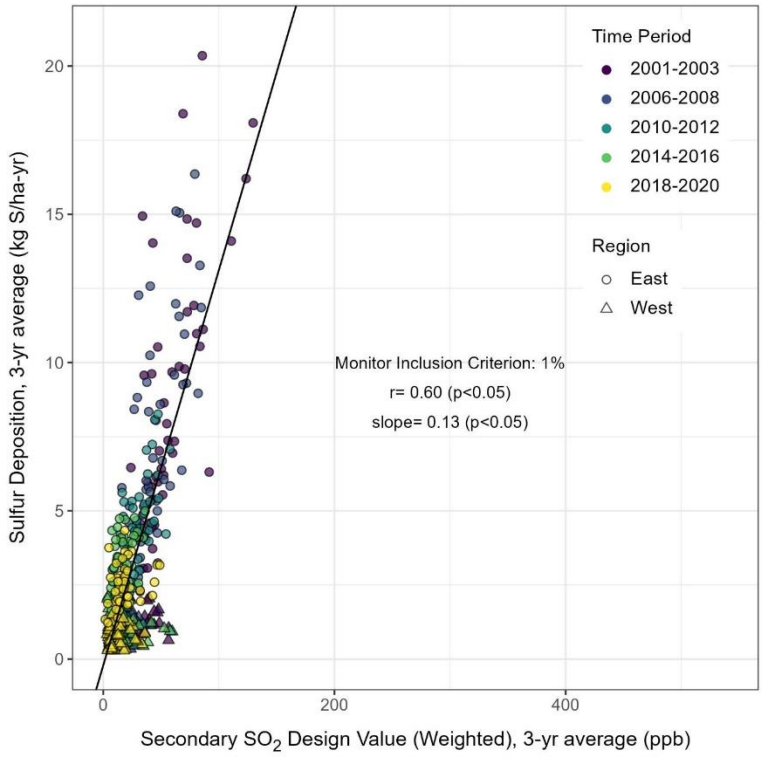

Figure 6A-34. The 3-hr SO_2 EAQM-weighted values and TDep S deposition in 84 ecoregions (120-hr trajectories, NAM-12, 1% monitor inclusion criteria).

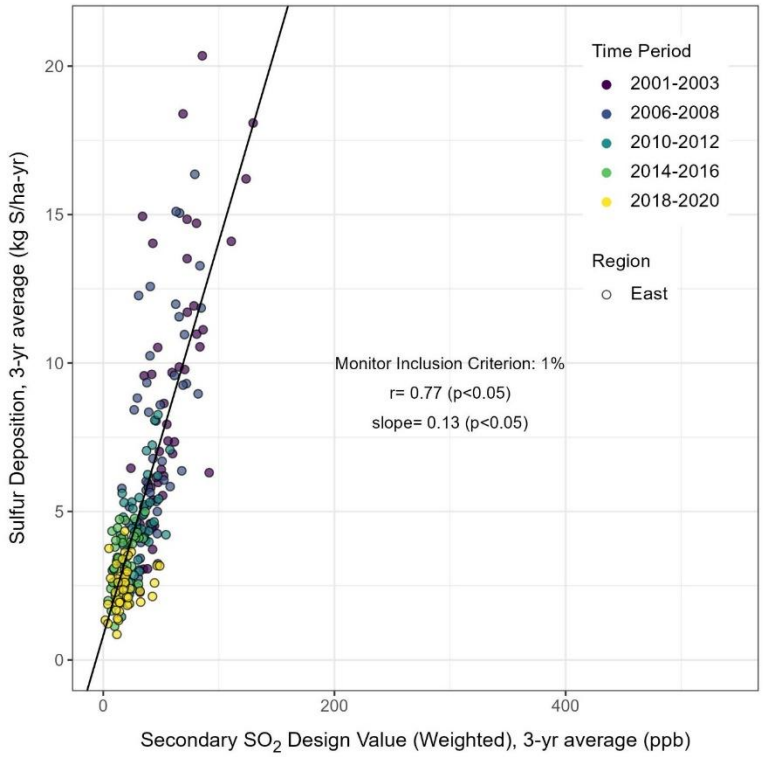

Figure 6A-35. The 3-hr SO_2 EAQM-weighted values and TDep S deposition in eastern ecoregions (120-hr trajectories, NAM-12, 1% monitor inclusion criteria).

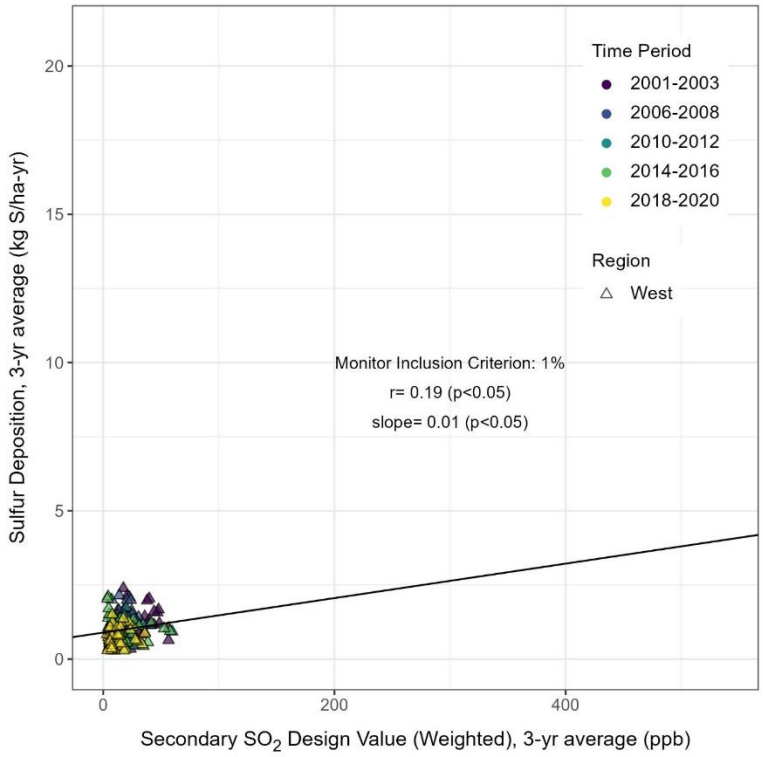

Figure 6A-36. The 3-hr SO_2 EAQM-weighted values and TDep S deposition in western ecoregions (120-hr trajectories, NAM-12, 1% monitor inclusion criteria).

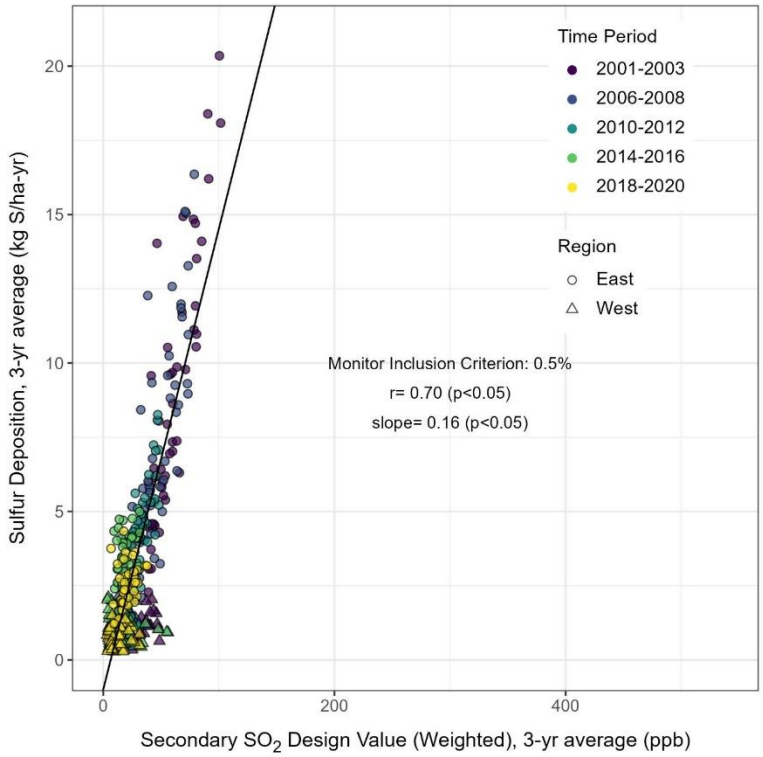

Figure 6A-37. The 3-hr SO_2 EAQM-weighted values and TDep S deposition in 84 ecoregions (120-hr trajectories, NAM-12, 0.5% monitor inclusion criteria).

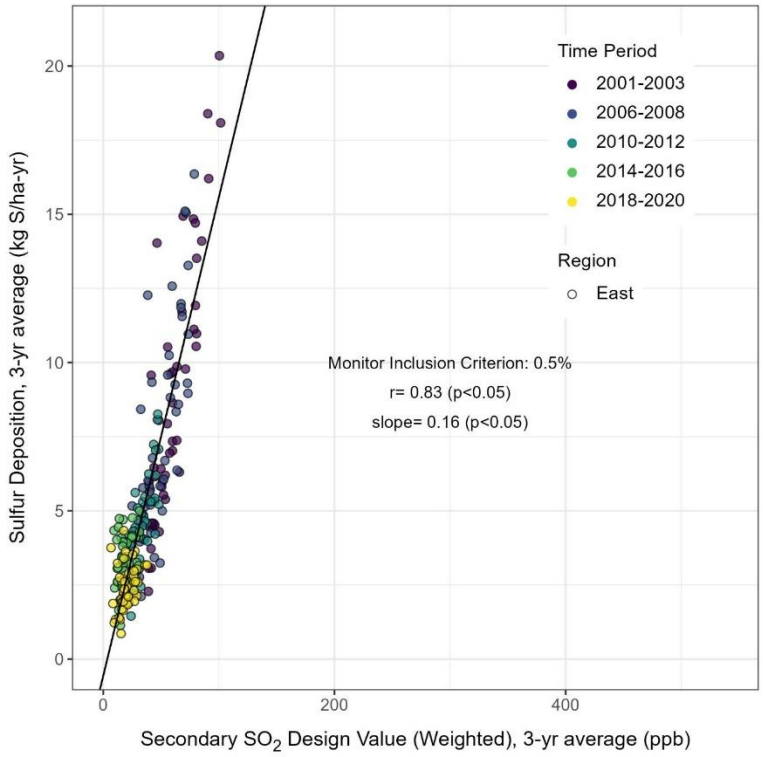

Figure 6A-38. The 3-hr SO_2 EAQM-weighted values and TDep S deposition in eastern ecoregions (120-hr trajectories, NAM-12, 0.5% monitor inclusion criteria).

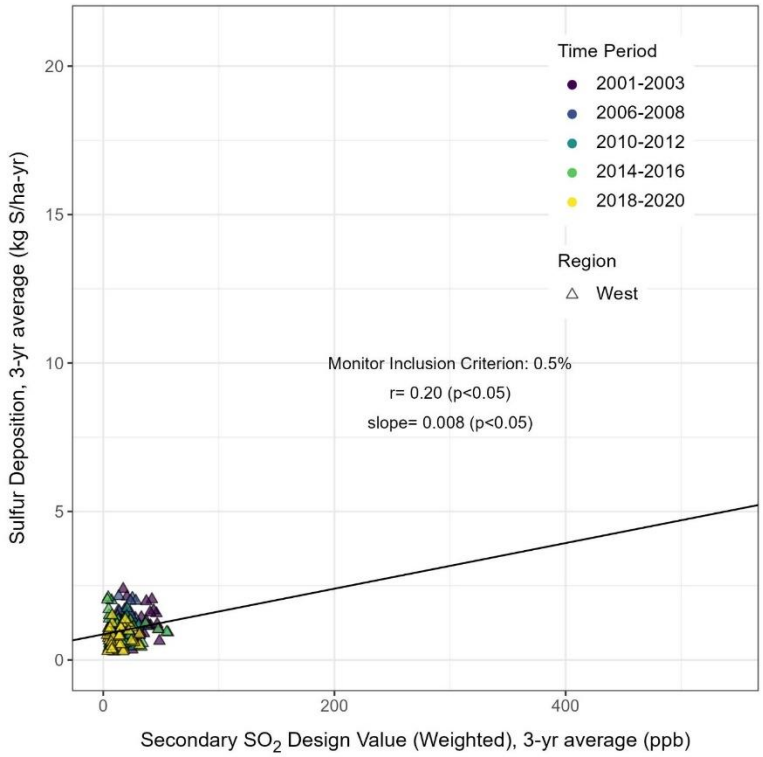

Figure 6A-39. The 3-hr SO_2 EAQM-weighted values and TDep S deposition in western ecoregions (120-hr trajectories, NAM-12, 0.5% monitor inclusion criteria).

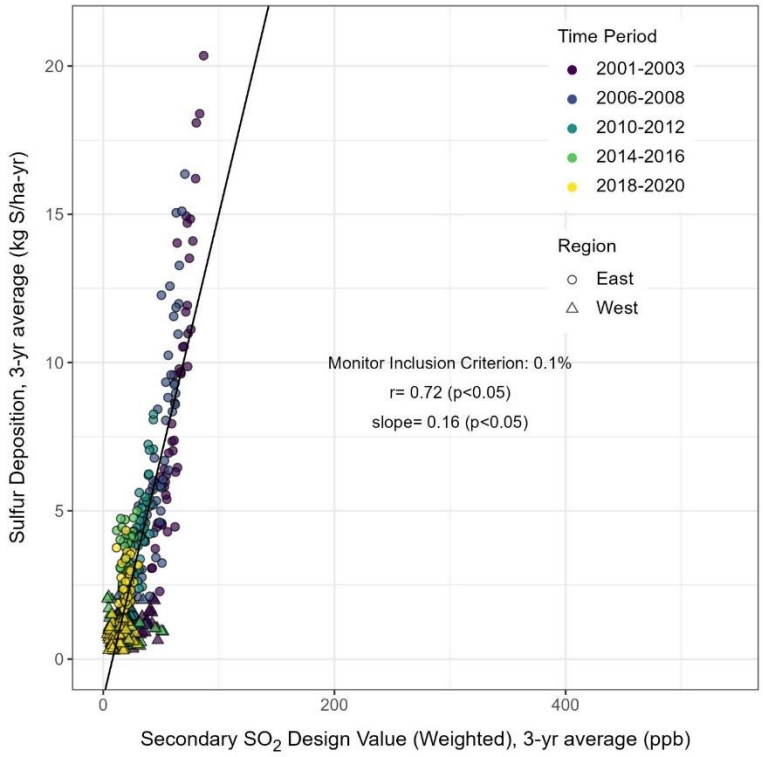

Figure 6A-40. The 3-hr SO_2 EAQM-weighted values and TDep S deposition in 84 ecoregions (120-hr trajectories, NAM-12, 0.1% monitor inclusion criteria).

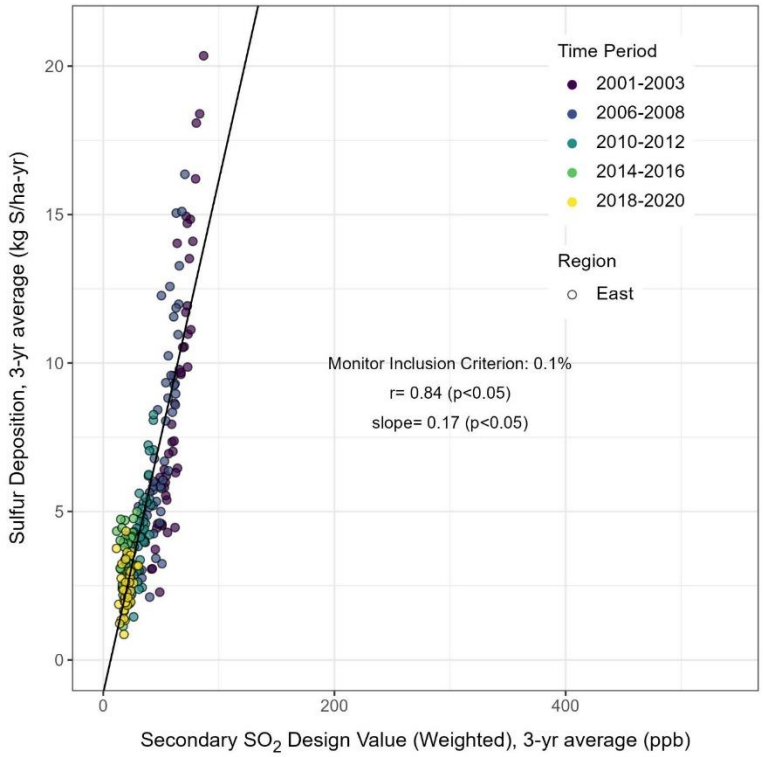

Figure 6A-41. The 3-hr SO_2 EAQM-weighted values and TDep S deposition in eastern ecoregions (120-hr trajectories, NAM-12, 0.1% monitor inclusion criteria).

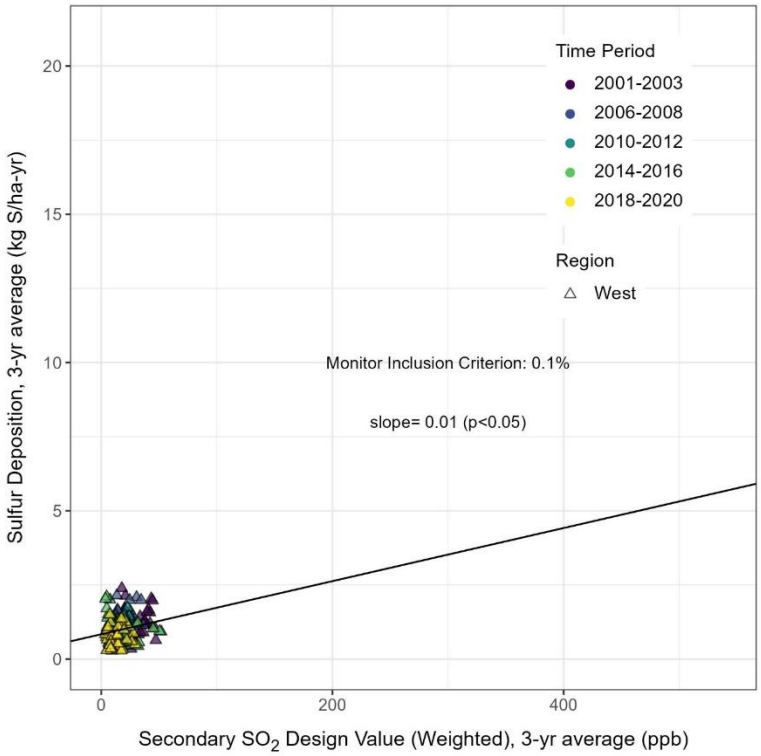

Figure 6A-42. The 3-hr SO_2 EAQM-weighted values and TDep S deposition in western ecoregions (120-hr trajectories, NAM-12, 0.1% monitor inclusion criteria).

6A.6.2. SO_2 3-hr Metric – 48-hr

Table 6A-3. Correlation coefficients of TDep estimates of sulfur deposition and 3-hr SO_2 EAQMs generated by HYSPLIT analysis, 48-hr trajectories. Data are also split by year and by region (East/West).

Sulfur Deposition and SO_2 (3-hr Standard)			
Annual Max-All Ecoregions- Monitor Inclusion Criteria: 1%	Correlation Coefficient (r) = 0.39*	*Weighted Annual Average-All Ecoregions- Monitor Inclusion Criteria: 1%*	Correlation Coefficient (r) = 0.59*
Year	r	Year	r
2001 - 2003	0.27*	2001 - 2003	0.72*
2006 - 2008	0.69*	2006 - 2008	0.80*
2010 - 2012	0.32*	2010 - 2012	0.64*
2014 - 2016	0.17	2014 - 2016	0.32*
2018 - 2020	0.32*	2018 - 2020	0.38*
Annual Max-East Ecoregions- Monitor Inclusion Criteria: 1%	r = 0.44*	*Weighted Annual Average-East Ecoregions- Monitor Inclusion Criteria: 1%*	r = 0.80*
Year	r	Year	r
2001 - 2003	0.29*	2001 - 2003	0.80*
2006 - 2008	0.33*	2006 - 2008	0.68*
2010 - 2012	0.22	2010 - 2012	0.67*
2014 - 2016	0.11	2014 - 2016	0.39*
2018 - 2020	0.18	2018 - 2020	0.35*
Annual Max-West Ecoregions- Monitor Inclusion Criteria: 1%	r = -0.07	*Weighted Annual Average-West Ecoregions- Monitor Inclusion Criteria: 1%*	r = 0.14
Year	r	Year	r
2001 - 2003	-0.31	2001 - 2003	0.05
2006 - 2008	-0.03	2006 - 2008	0.11
2010 - 2012	-0.20	2010 - 2012	0.01
2014 - 2016	-0.33*	2014 - 2016	-0.24
2018 - 2020	0.10	2018 - 2020	0.07
*p< 0.05			

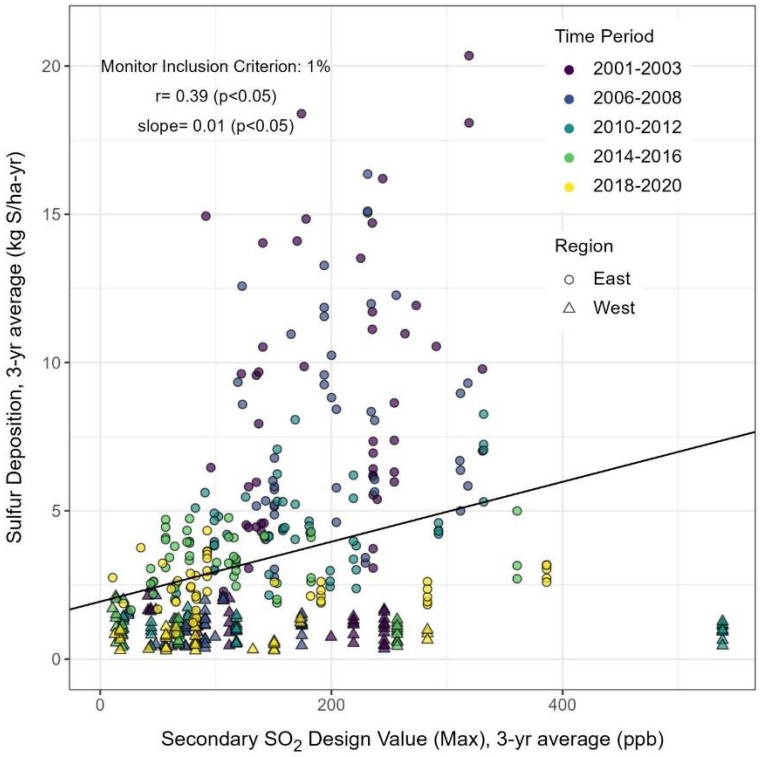

Figure 6A-43. The 3-hr SO_2 EAQM-max values and TDep S deposition in 84 ecoregions (48-hr trajectories, NARR-32, 1% monitor inclusion criteria).

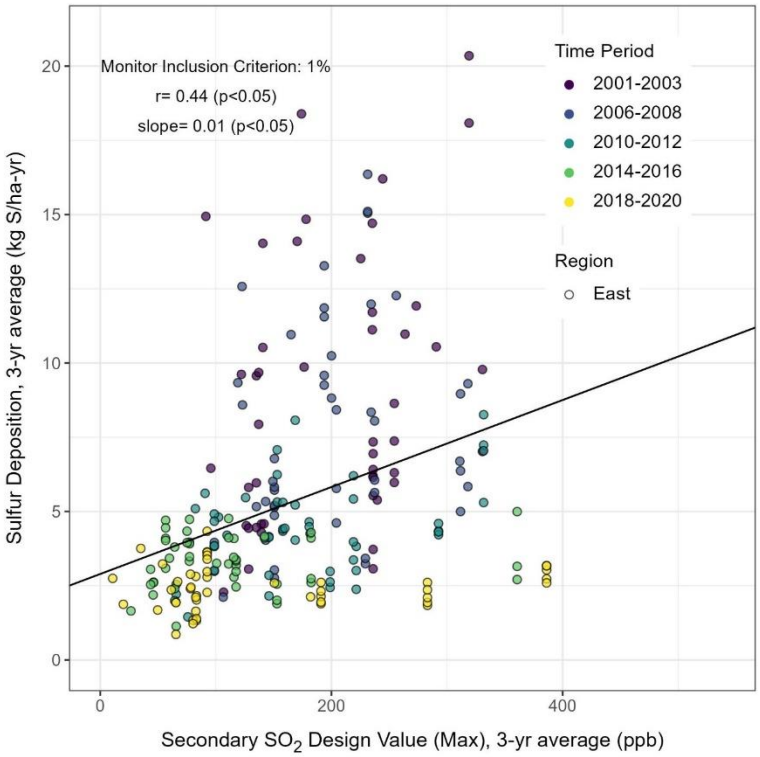

Figure 6A-44. The 3-hr SO_2 EAQM-max values and TDep S deposition in eastern ecoregions (48-hr trajectories, NARR-32, 1% monitor inclusion criteria).

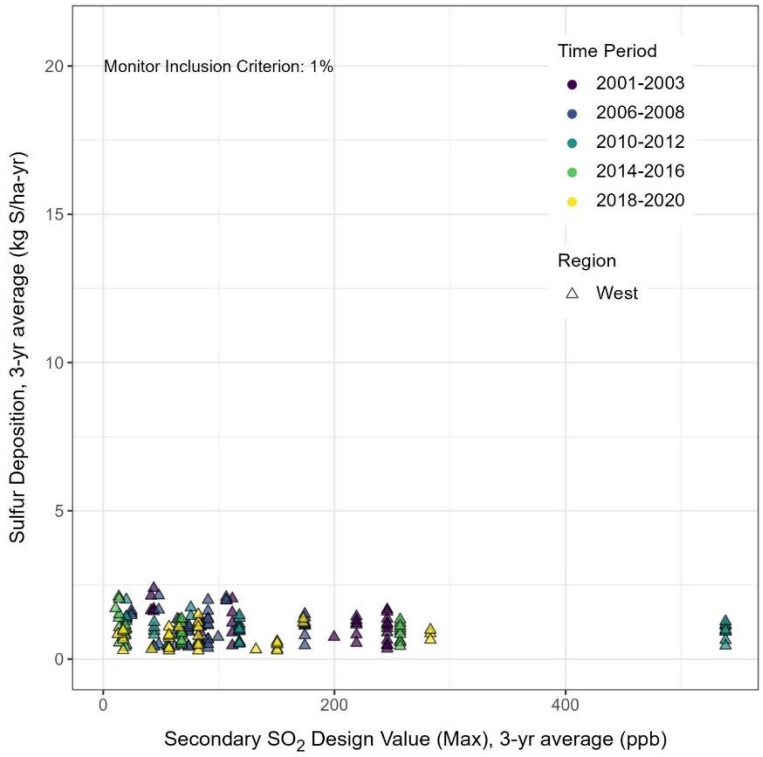

Figure 6A-45. The 3-hr SO_2 EAQM-max values and TDep S deposition in western ecoregions (48-hr trajectories, NARR-32, 1% monitor inclusion criteria).

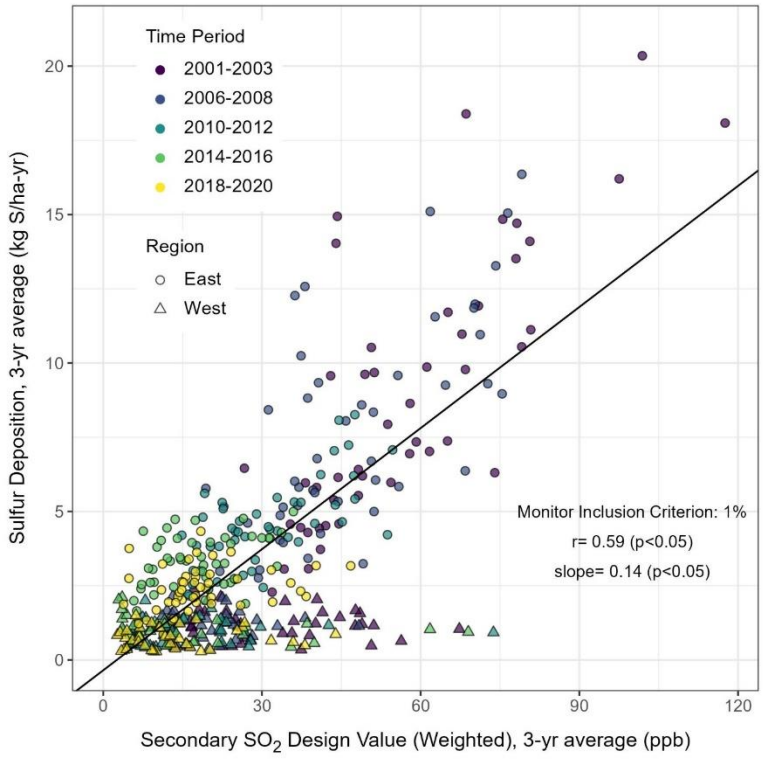

Figure 6A-46. The 3-hr SO_2 EAQM-weightedvalues and TDep S deposition in 84 ecoregions (48-hr trajectories, NARR-32, 1% monitor inclusion criteria).

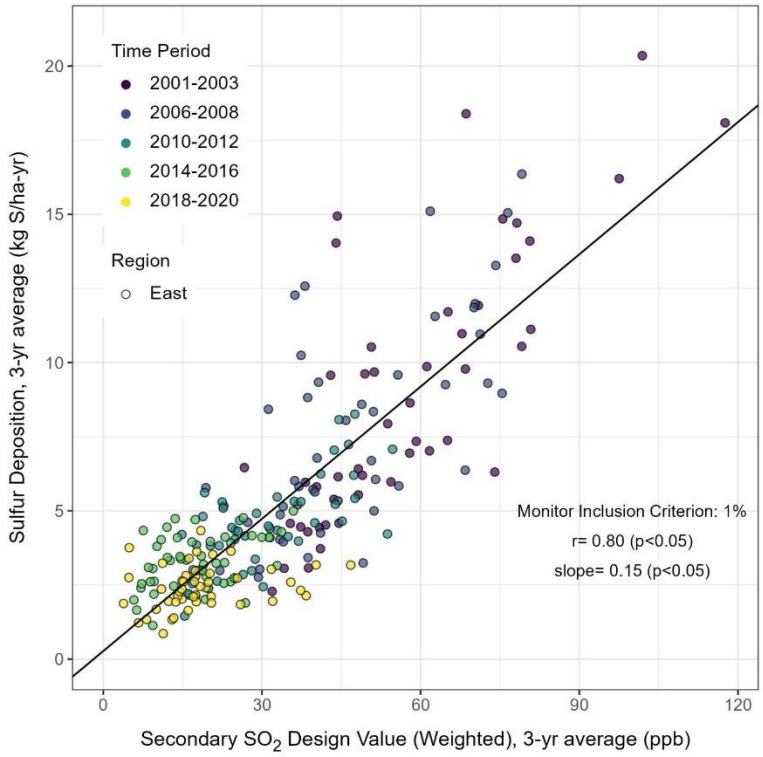

Figure 6A-47. The 3-hr SO_2 EAQM-weighted values and TDep S deposition in eastern ecoregions (48-hr trajectories, NARR-32, 1% monitor inclusion criteria).

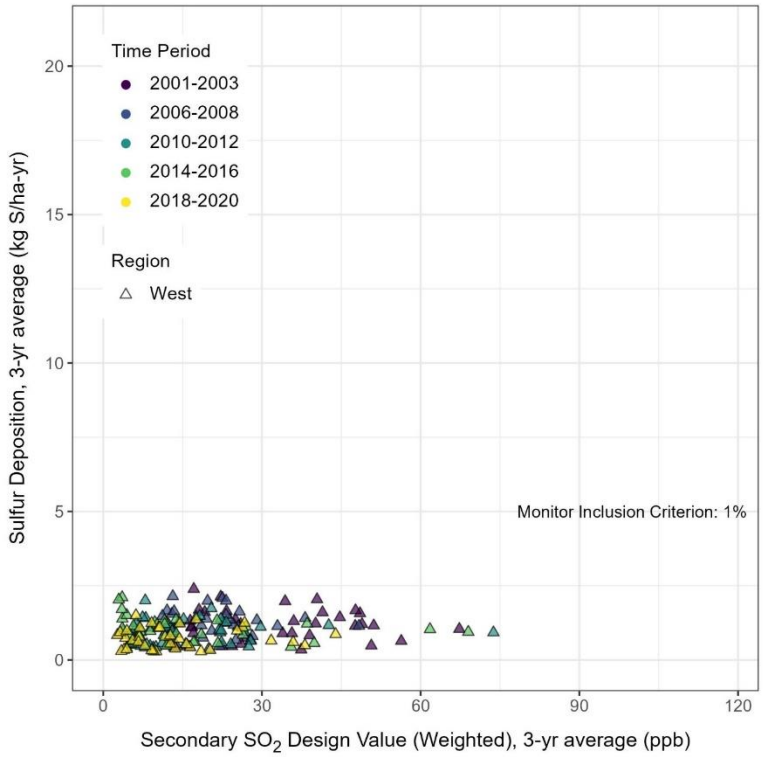

Figure 6A-48. The 3-hr SO_2 EAQM-weighted values and TDep S deposition in western ecoregions (48-hr trajectories, NARR-32, 1% monitor inclusion criteria).

6A.6.3. SO_2 Annual Metric – 120-hr

Table 6A-4. Correlation coefficients of TDep estimates of sulfur deposition and annual SO_2 EAQMs generated by HYSPLIT analysis at three different monitor inclusion criteria, 120-hr trajectories.

Sulfur Deposition and SO_2					
Annual Max-All Ecoregions- Monitor Inclusion Criteria: 1%	Correlation Coefficient (r) = 0.24*	*Annual Max-All Ecoregions- Monitor Inclusion Criteria: 0.5%*	Correlation Coefficient (r) = 0.48*	*Annual Max-All Ecoregions- Monitor Inclusion Criteria: 0.1%*	Correlation Coefficient (r) = 0.51*
Year	r	Year	r	Year	r
2001 - 2003	0.33*	2001 - 2003	0.62*	2001 - 2003	0.87*
2006 - 2008	0.48*	2006 - 2008	0.69*	2006 - 2008	0.87*
2010 - 2012	0.19	2010 - 2012	0.28*	2010 - 2012	0.72*
2014 - 2016	-0.24*	2014 - 2016	-0.05	2014 - 2016	-0.28*
2018 - 2020	-0.34*	2018 - 2020	0.19	2018 - 2020	0.76*
Annual Max-All Ecoregions- Monitor Inclusion Criteria: 1% [a]	r = 0.24*	*Annual Max-All Ecoregions- Monitor Inclusion Criteria: 0.5%* [a]	r = 0.49*	*Annual Max-All Ecoregions- Monitor Inclusion Criteria: 0.1%* [a]	r = 0.61*
Year	r	Year	r	Year	r
2001 - 2003	0.33*	2001 - 2003	0.62*	2001 - 2003	0.87*
2006 - 2008	0.48*	2006 - 2008	0.69*	2006 - 2008	0.87*
2010 - 2012	0.19*	2010 - 2012	0.28*	2010 - 2012	0.72*
2014 - 2016	-0.24*	2014 - 2016	-0.05	2014 - 2016	-0.28*
2018 - 2020	-0.45*	2018 - 2020	-0.10	2018 - 2020	-0.10
Annual Max-East Ecoregions- Monitor Inclusion Criteria: 1%	r = 0.65*	*Annual Max-East Ecoregions- Monitor Inclusion Criteria: 0.5%*	r = 0.53*	*Annual Max-East Ecoregions- Monitor Inclusion Criteria: 0.1%*	r = 0.05
Year	r	Year	r	Year	r
2001 - 2003	0.69*	2001 - 2003	0.78*	2001 - 2003	0.73*
2006 - 2008	0.55*	2006 - 2008	0.59*	2006 - 2008	0.74*
2010 - 2012	0.18	2010 - 2012	-0.43*	2010 - 2012	0.01
2014 - 2016	0.02	2014 - 2016	-0.44*	2014 - 2016	-0.09
2018 - 2020	0.23	2018 - 2020	0.23	2018 - 2020	0.17
Annual Max-East Ecoregions- Monitor Inclusion Criteria: 1%[a]	r = 0.68*	*Annual Max-East Ecoregions- Monitor Inclusion Criteria: 0.5%*[a]	r = 0.65*	*Annual Max-East Ecoregions- Monitor Inclusion Criteria: 0.1%*[a]	r = 0.72*
Year	r	Year	r	Year	r
2001 - 2003	0.69*	2001 - 2003	0.78*	2001 - 2003	0.73*
2006 - 2008	0.56*	2006 - 2008	0.59*	2006 - 2008	0.74*
2010 - 2012	0.18	2010 - 2012	-0.43*	2010 - 2012	-0.01
2014 - 2016	0.12	2014 - 2016	-0.44*	2014 - 2016	-0.09
2018 - 2020	0.15	2018 - 2020	-0.13	2018 - 2020	N/A (only 2 datapoints)
Annual Max-West Ecoregions- Monitor Inclusion Criteria: 1%	r = -0.07	*Annual Max-West Ecoregions- Monitor Inclusion Criteria: 0.5%*	r = 0.04	*Annual Max-West Ecoregions- Monitor Inclusion Criteria: 0.1%*	r = 0.04
Year	r	Year	r	Year	r
2001 - 2003	-0.19	2001 - 2003	-0.11	2001 - 2003	-0.09
2006 - 2008	-0.07	2006 - 2008	0.12	2006 - 2008	0.05
2010 - 2012	-0.26	2010 - 2012	-0.07	2010 - 2012	-0.12
2014 - 2016	-0.19	2014 - 2016	-0.06	2014 - 2016	-0.27

2018 - 2020	-0.07	2018 - 2020	0.03	2018 - 2020	-0.09
Annual Max-West Ecoregions (outliers excluded)- Monitor Inclusion Criteria: 1%	No outliers in West dataset	*Annual Max-West Ecoregions (outliers excluded)- Monitor Inclusion Criteria: 0.5%*	No outliers in West dataset	*Annual Max-West Ecoregions (outliers excluded)- Monitor Inclusion Criteria: 0.1%* [a]	r = 0.03
Year	r	Year	r	Year	r
2001 - 2003		2001 - 2003		2001 - 2003	-0.09
2006 - 2008		2006 - 2008		2006 - 2008	0.05
2010 - 2012		2010 - 2012		2010 - 2012	-0.12
2014 - 2016		2014 - 2016		2014 - 2016	-0.27
2018 - 2020		2018 - 2020		2018 - 2020	-0.23
Weighted Annual Average-All Ecoregions- Monitor Inclusion Criteria: 1%	r = 0.47*	*Weighted Annual Average-All Ecoregions- Monitor Inclusion Criteria: 0.5%*	r = 0.56*	*Weighted Annual Average-All Ecoregions- Monitor Inclusion Criteria: 0.1%*	r = 0.59*
Year	r	Year	r	Year	r
2001 - 2003	0.66*	2001 - 2003	0.77*	2001 - 2003	0.85*
2006 - 2008	0.72*	2006 - 2008	0.81*	2006 - 2008	0.86*
2010 - 2012	0.58*	2010 - 2012	0.71*	2010 - 2012	0.75*
2014 - 2016	0.07	2014 - 2016	0.16	2014 - 2016	0.24*
2018 - 2020	-0.04	2018 - 2020	0.22*	2018 - 2020	0.34
Weighted Annual Average-East Ecoregions- Monitor Inclusion Criteria: 1%	r = 0.85*	*Weighted Annual Average-East Ecoregions- Monitor Inclusion Criteria: 0.5%*	r = 0.85*	*Weighted Annual Average-East Ecoregions- Monitor Inclusion Criteria: 0.1%*	r = 0.84*
Year	r	Year	r	Year	r
2001 - 2003	0.90*	2001 - 2003	0.89*	2001 - 2003	0.84*
2006 - 2008	0.88*	2006 - 2008	0.9*	2006 - 2008	0.85*
2010 - 2012	0.75*	2010 - 2012	0.75*	2010 - 2012	0.72*
2014 - 2016	0.32*	2014 - 2016	0.19	2014 - 2016	0.2
2018 - 2020	0.21	2018 - 2020	0.30*	2018 - 2020	0.31*
Weighted Annual Average-West Ecoregions- Monitor Inclusion Criteria: 1%	r = 0.14	*Weighted Annual Average-West Ecoregions- Monitor Inclusion Criteria: 0.5%*	r = 0.19*	*Weighted Annual Average-West Ecoregions- Monitor Inclusion Criteria: 0.1%*	r = 0.20*
Year	r	Year	r	Year	r
2001 - 2003	-0.01	2001 - 2003	0.04	2001 - 2003	0.07
2006 - 2008	-0.17	2006 - 2008	-0.07	2006 - 2008	0.06
2010 - 2012	-0.23	2010 - 2012	-0.12	2010 - 2012	-0.11
2014 - 2016	-0.19	2014 - 2016	-0.14	2014 - 2016	-0.17
2018 - 2020	0.04	2018 - 2020	0.04	2018 - 2020	0.02

*p< 0.05

[a] Note: There are several outlier points in this comparison where the EAQM-max annual average SO_2 value exceeds 20 ppb in the 2018-2020 period. These points have been removed from these analyses. These data are driven by a monitor in southeastern MO where annual average SO_2 has exceeded 20 ppb in recent years. Any downwind ecoregion that is linked to this upwind monitor will have an EAQM-max with this value. A preliminary analysis suggests that these observed SO_2 data are due to a new source that was not modeled in the CMAQ simulation that informed the TDep estimates of deposition. As there is no deposition monitor in the immediate vicinity of the source it is unlikely that the TDep estimates are capturing the impacts of this source. For that reason, we concluded it was appropriate not to consider these data in our evaluation of the concentration-deposition relationship.

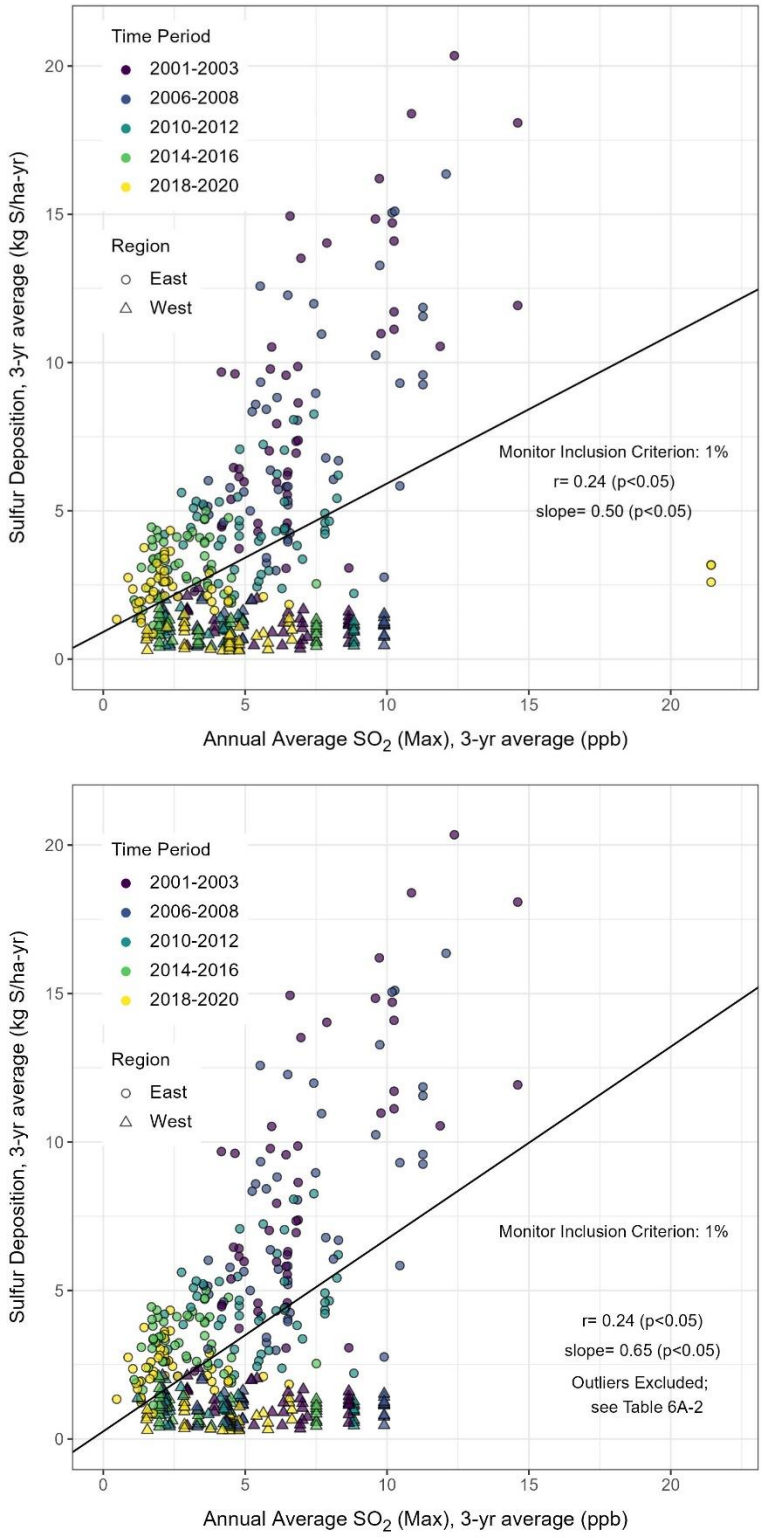

Figure 6A-49. Annual SO_2 EAQM-max values and TDep S deposition in 84 ecoregions (120-hr trajectories, NAM-12, 1% monitor inclusion criteria): all values (upper), outliers excluded (lower).

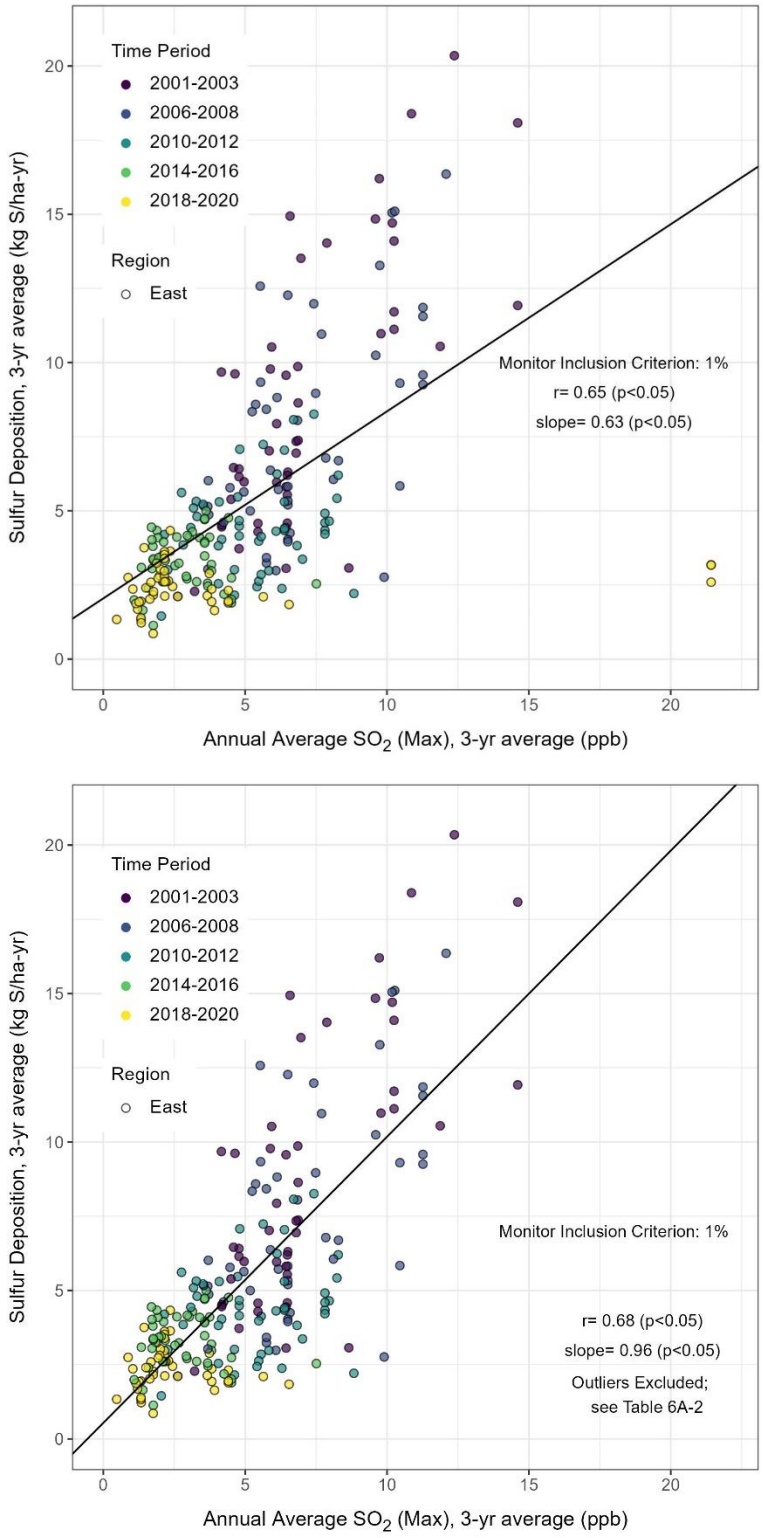

Figure 6A-50. Annual SO_2 EAQM-max values and TDep S deposition in eastern ecoregions (120-hr trajectories, NAM-12, 1% monitor inclusion criteria): all values (upper), outliers excluded (lower).

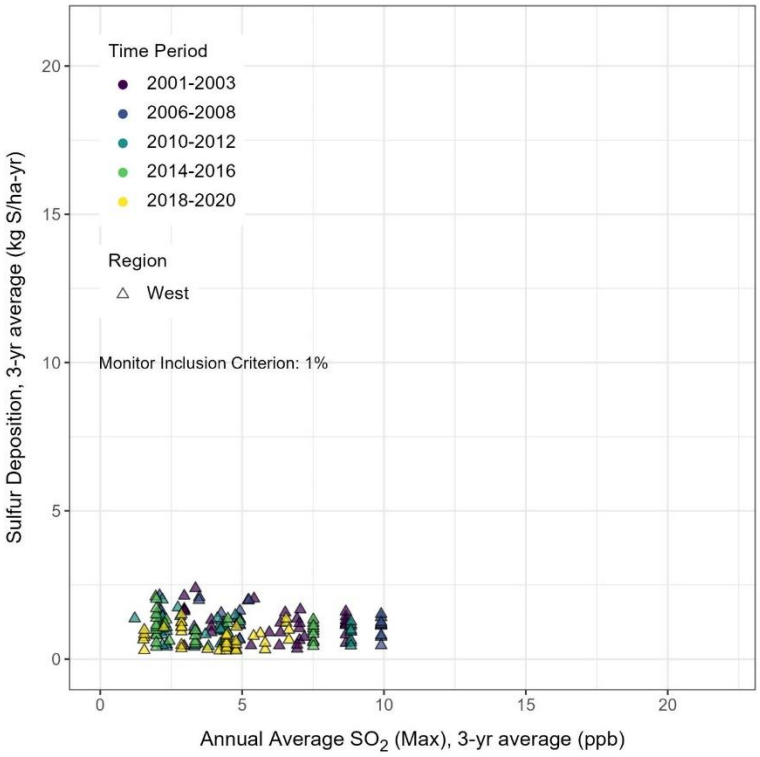

Figure 6A-51. Annual SO_2 EAQM-max values and TDep S deposition in western ecoregions (120-hr trajectories, NAM-12, 1% monitor inclusion criteria).

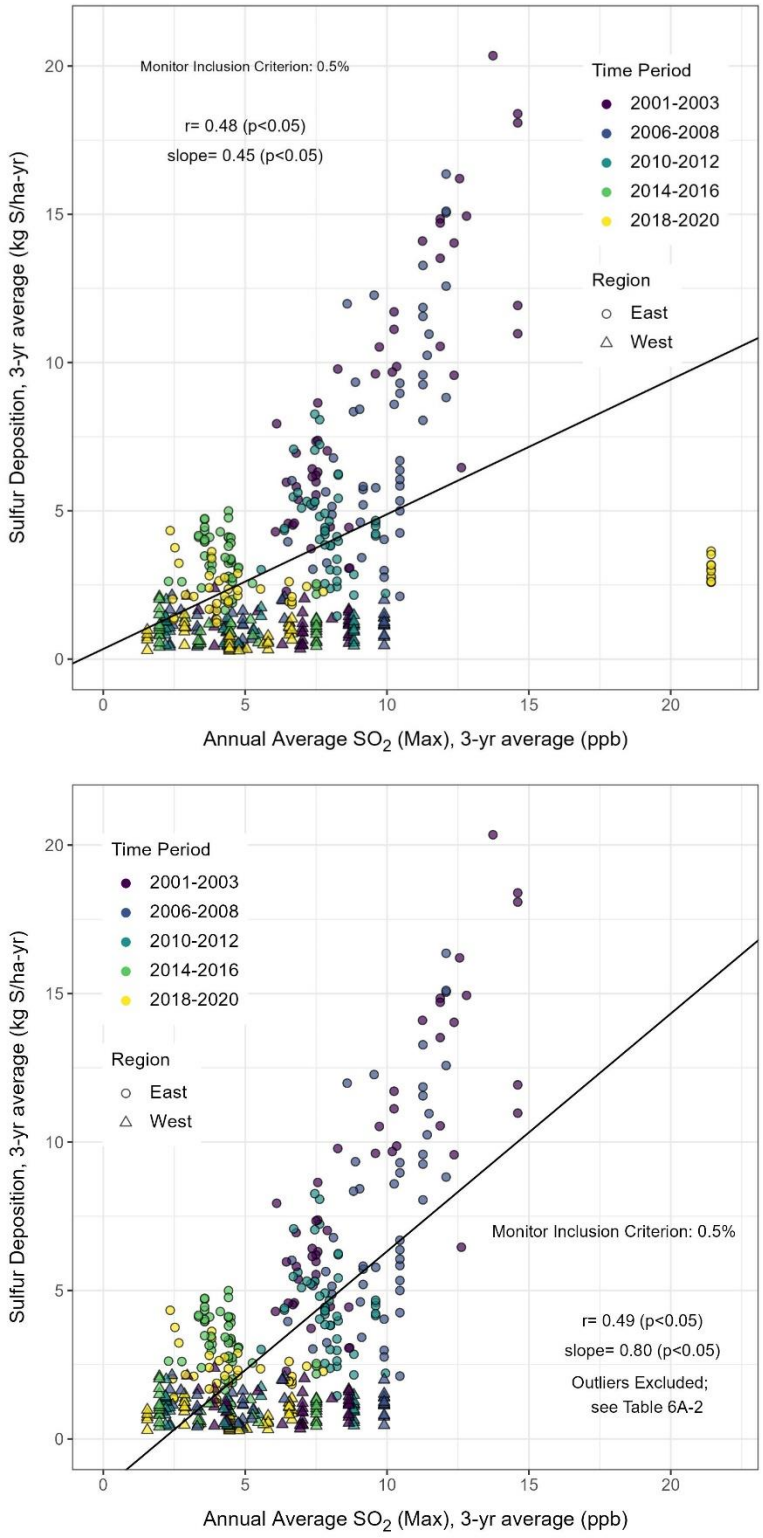

Figure 6A-52. Annual SO_2 EAQM-max values and TDep S deposition in 84 ecoregions (120-hr trajectories, NAM-12, 0.5% monitor inclusion criteria): all values (upper), outliers excluded (lower).

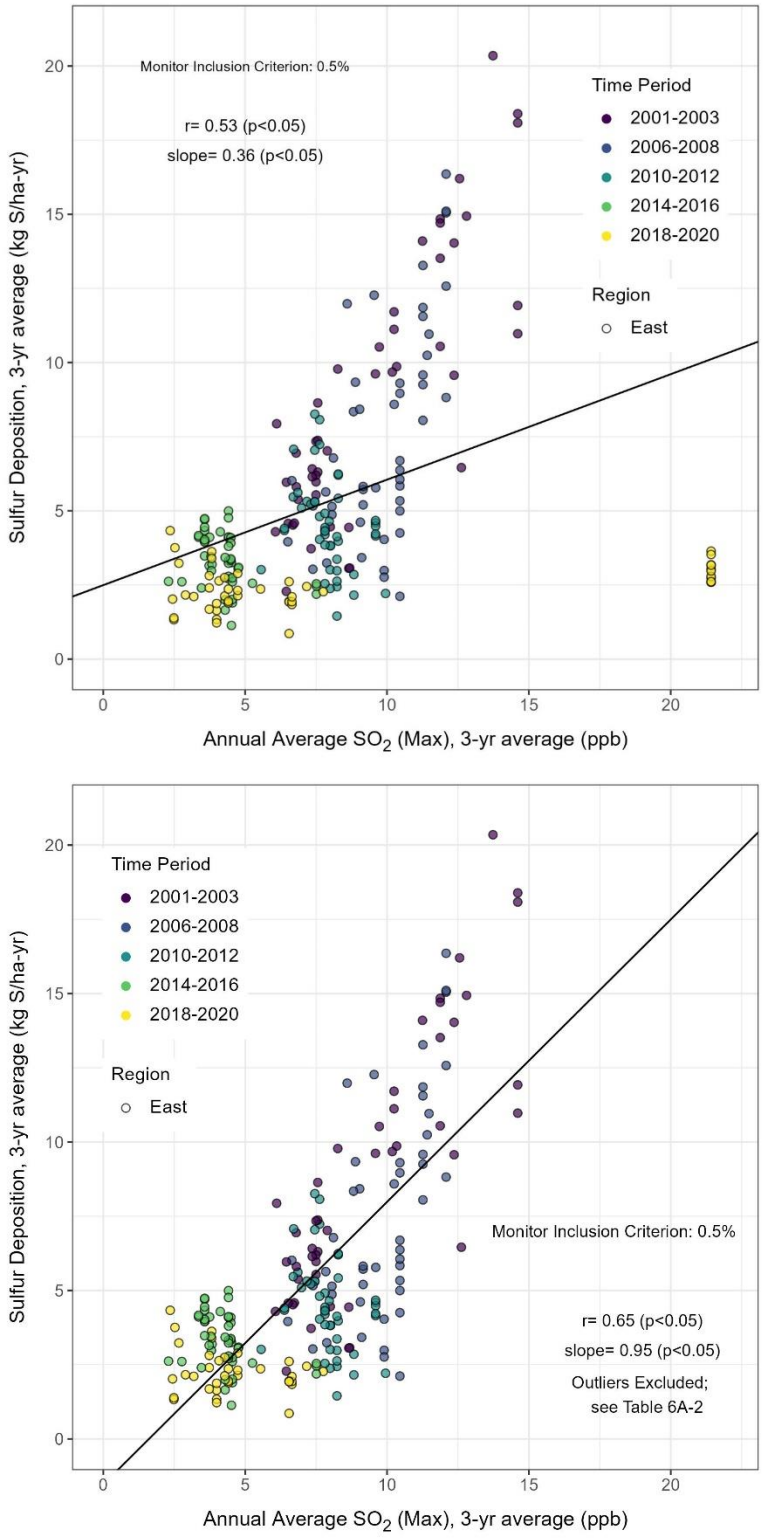

Figure 6A-53. Annual SO_2 EAQM-max values and TDep S deposition in eastern ecoregions (120-hr trajectories, NAM-12, 0.5% monitor inclusion criteria): all values (upper), outliers excluded (lower).

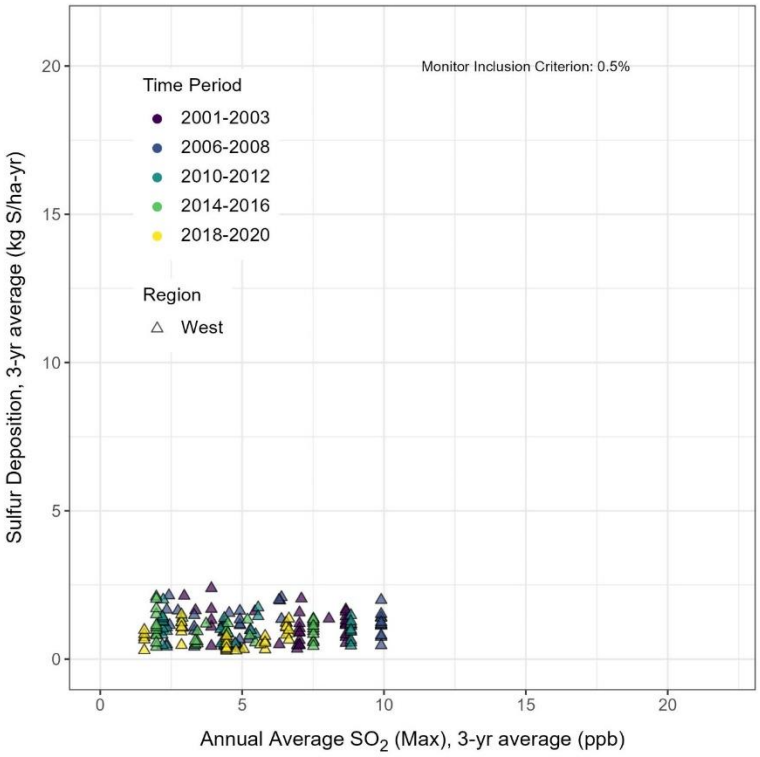

Figure 6A-54. Annual SO_2 EAQM-max values and TDep S deposition in western ecoregions (120-hr trajectories, NAM-12, 0.5% monitor inclusion criteria).

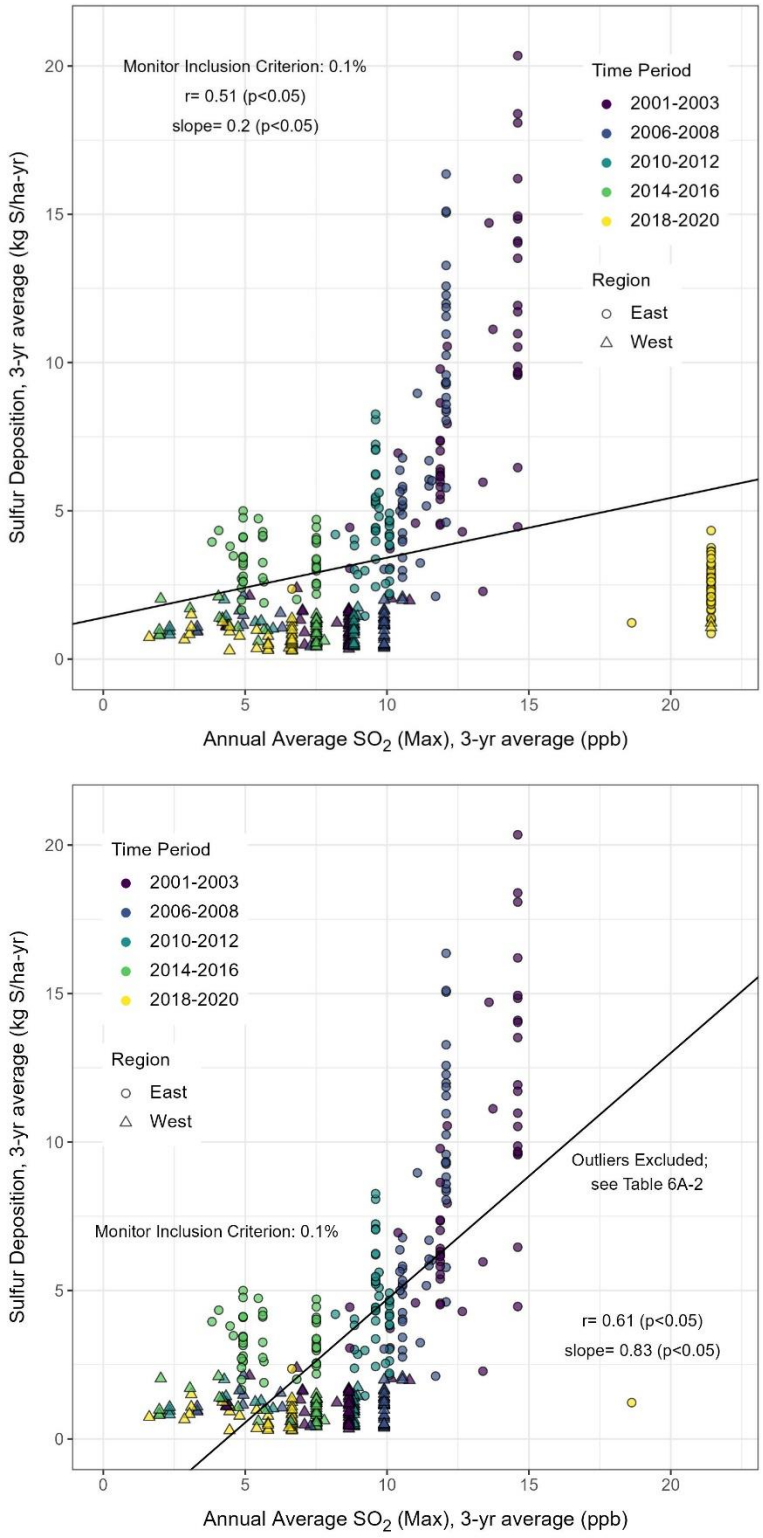

Figure 6A-55. Annual SO_2 EAQM-max values and TDep S deposition in 84 ecoregions (120-hr trajectories, NAM-12, 0.1% monitor inclusion criteria): all values (upper), outliers excluded (lower).

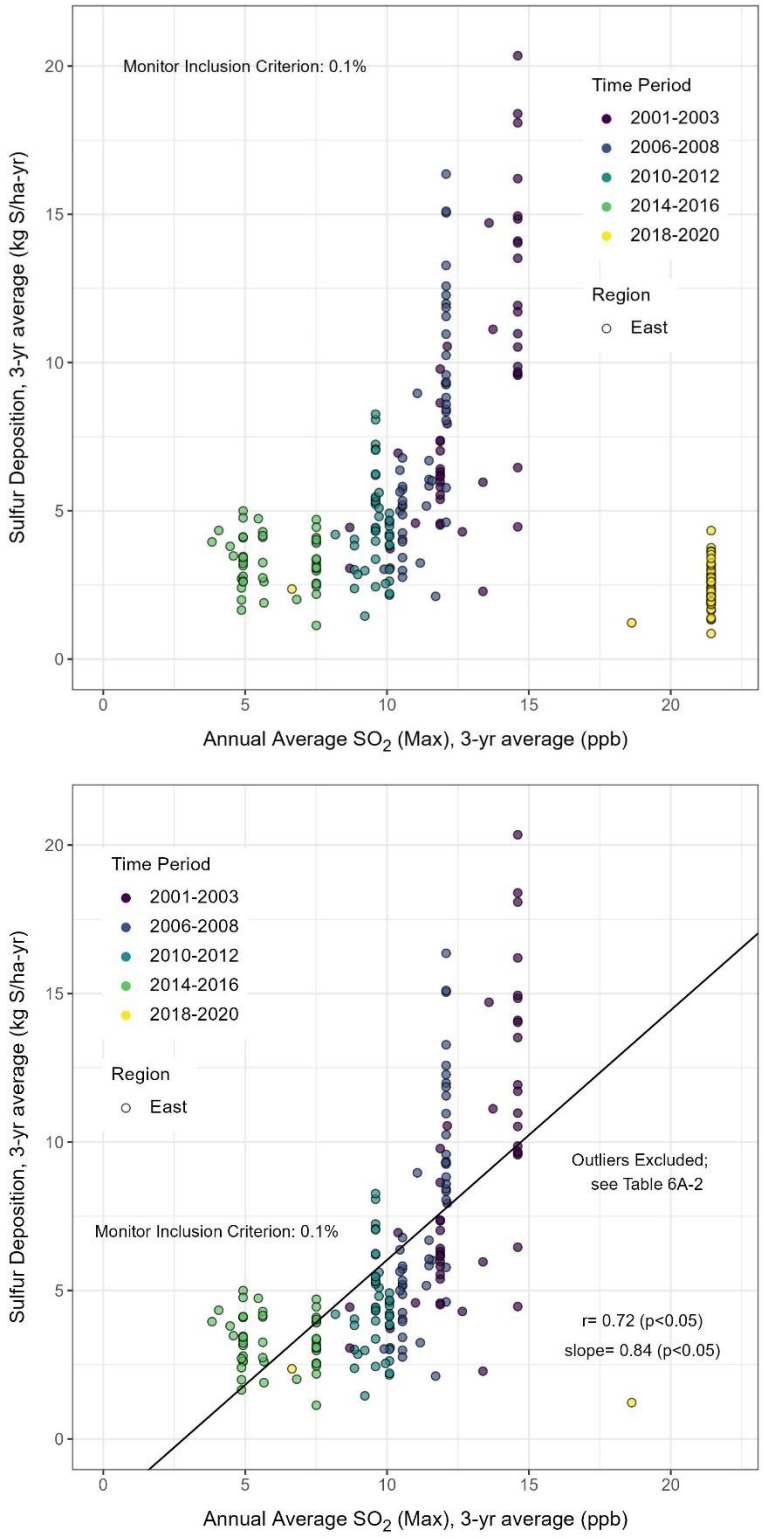

Figure 6A-56. Annual SO_2 EAQM-max values and TDep S deposition in eastern ecoregions (120-hr trajectories, NAM-12, 0.1% monitor inclusion criteria): all values (upper), outliers excluded (lower).

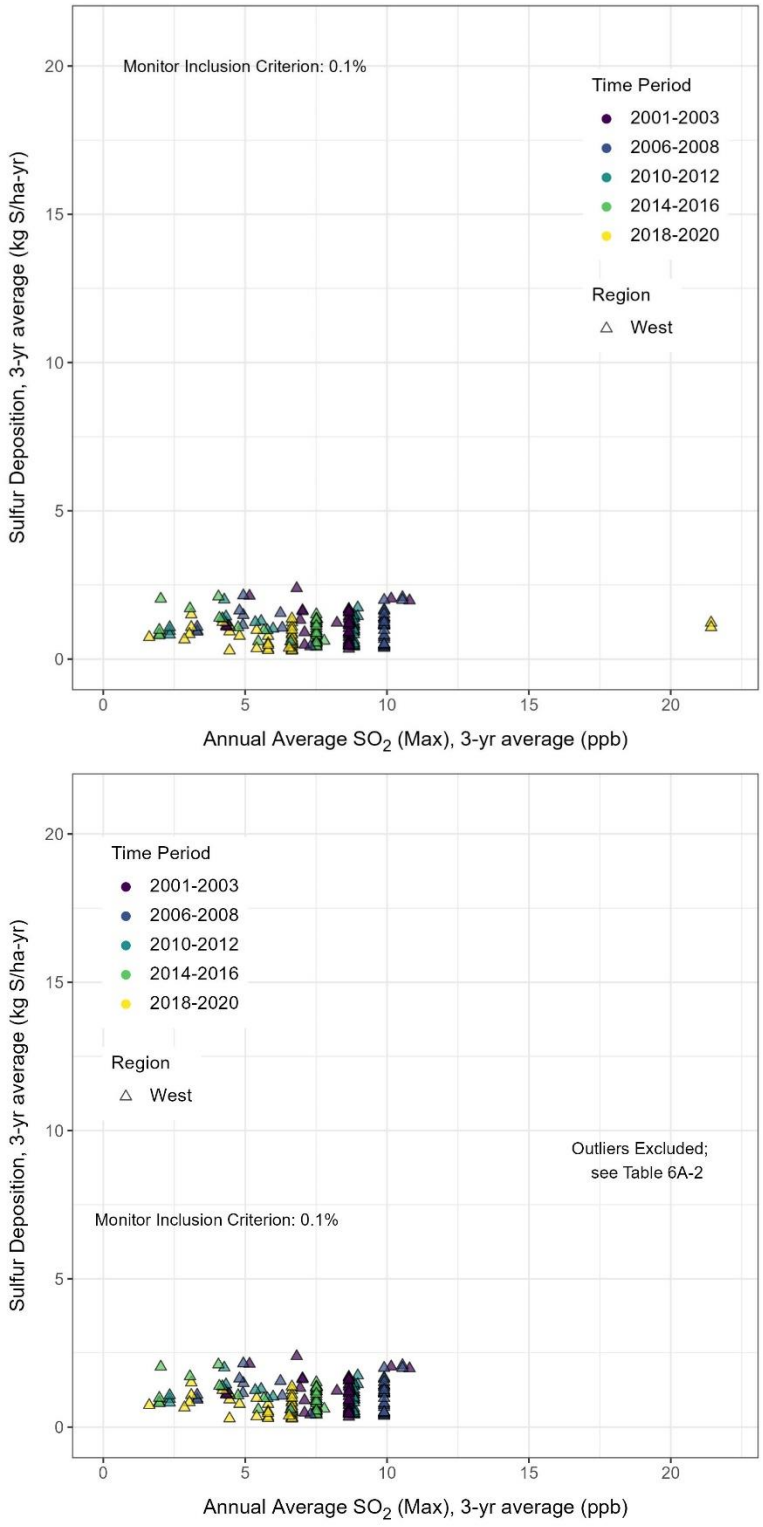

Figure 6A-57. Annual SO_2 EAQM-max values and TDep S deposition in western ecoregions (120-hr trajectories, NAM-12, 0.1% monitor inclusion criteria): all values (upper), outliers excluded (lower).

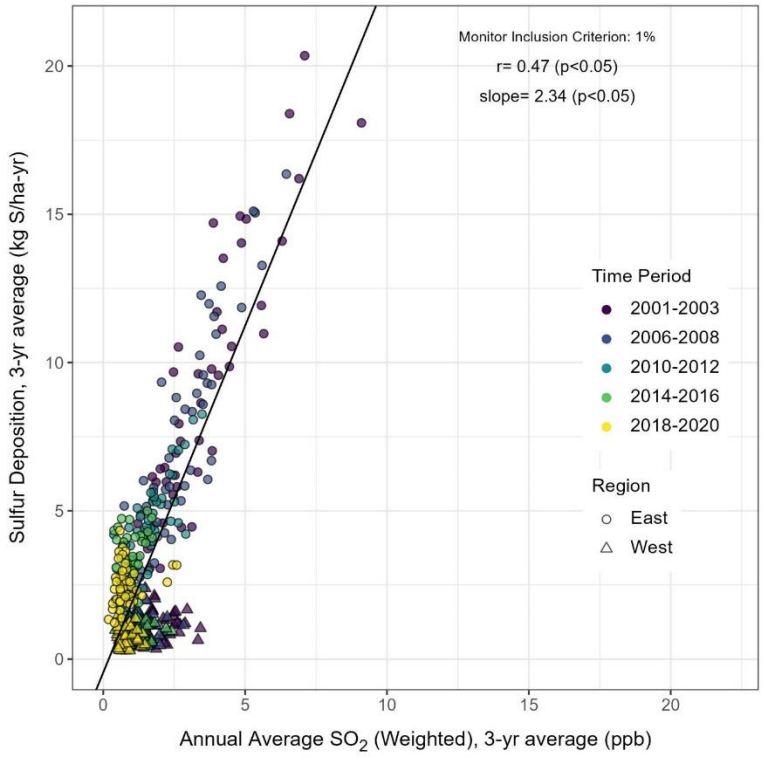

Figure 6A-58. Annual SO_2 EAQM-weighted values and TDep S deposition in 84 ecoregions (120-hr trajectories, NAM-12, 1% monitor inclusion criteria).

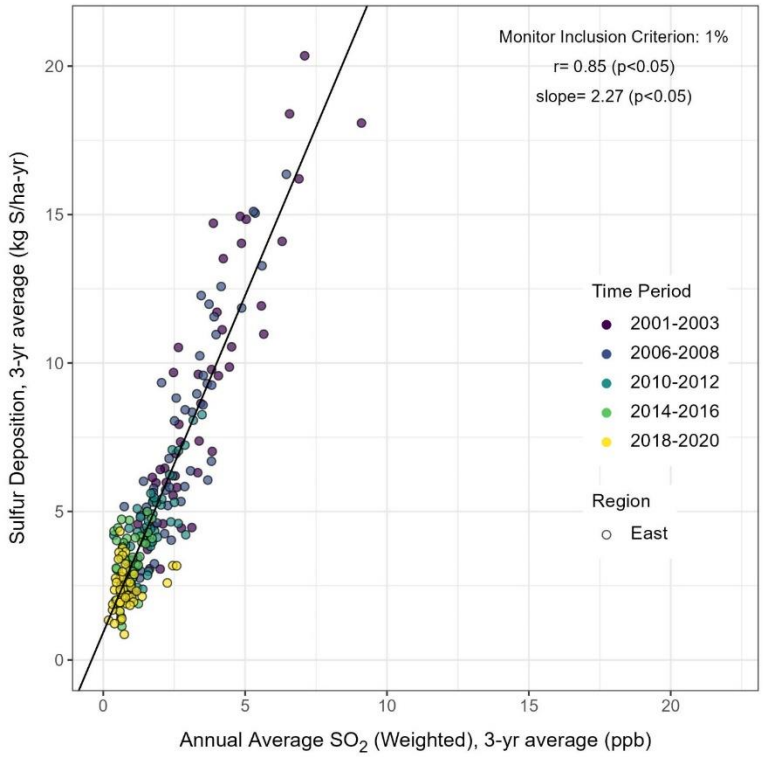

Figure 6A-59. Annual SO_2 EAQM-weighted values and TDep S deposition in eastern ecoregions (120-hr trajectories, NAM-12, 1% monitor inclusion criteria).

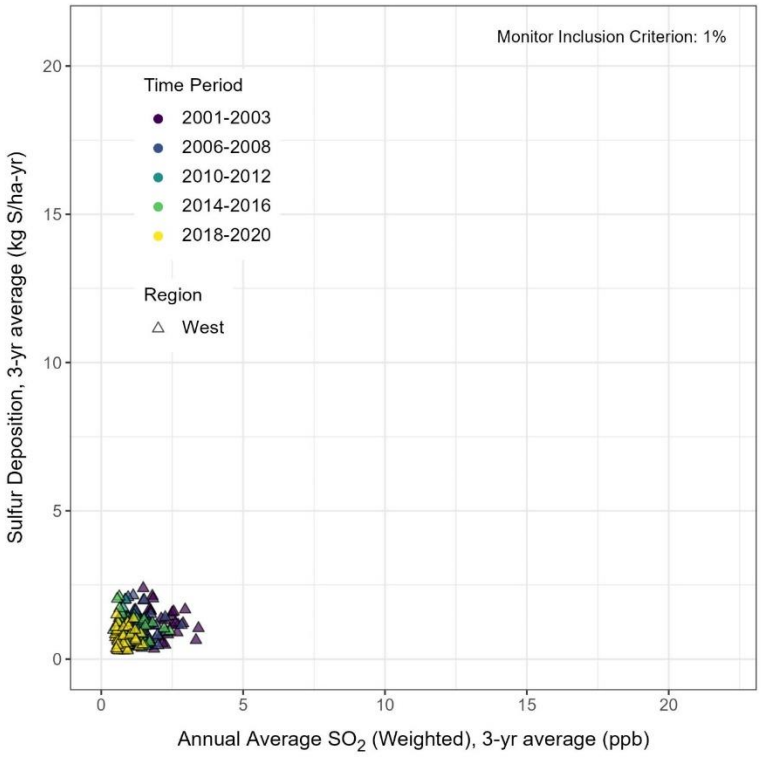

Figure 6A-60. Annual SO_2 EAQM-weighted values and TDep S deposition in western ecoregions (120-hr trajectories, NAM-12, 1% monitor inclusion criteria).

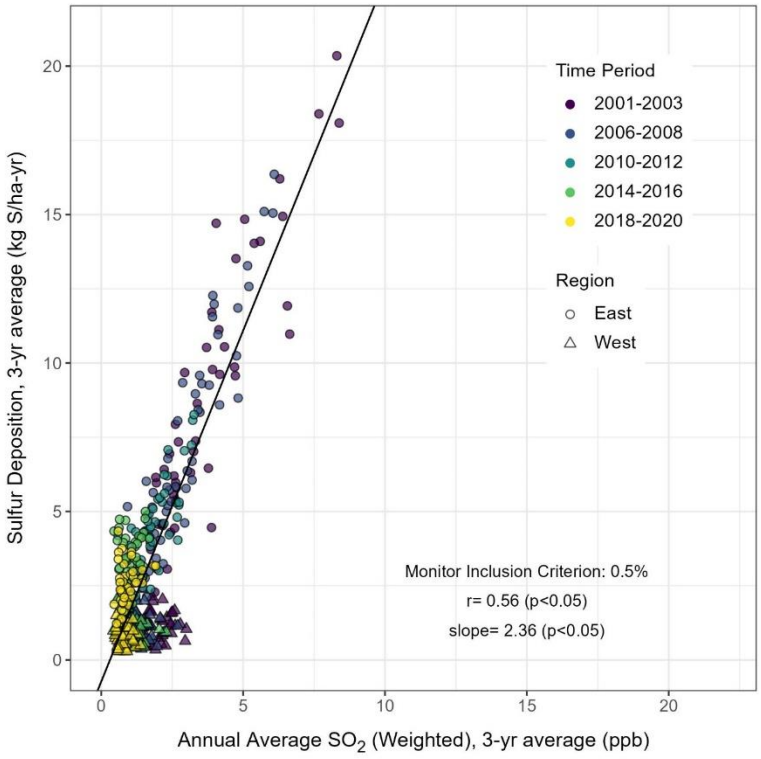

Figure 6A-61. Annual SO_2 EAQM-weighted values and TDep S deposition in 84 ecoregions (120-hr trajectories, NAM-12, 0.5% monitor inclusion criteria).

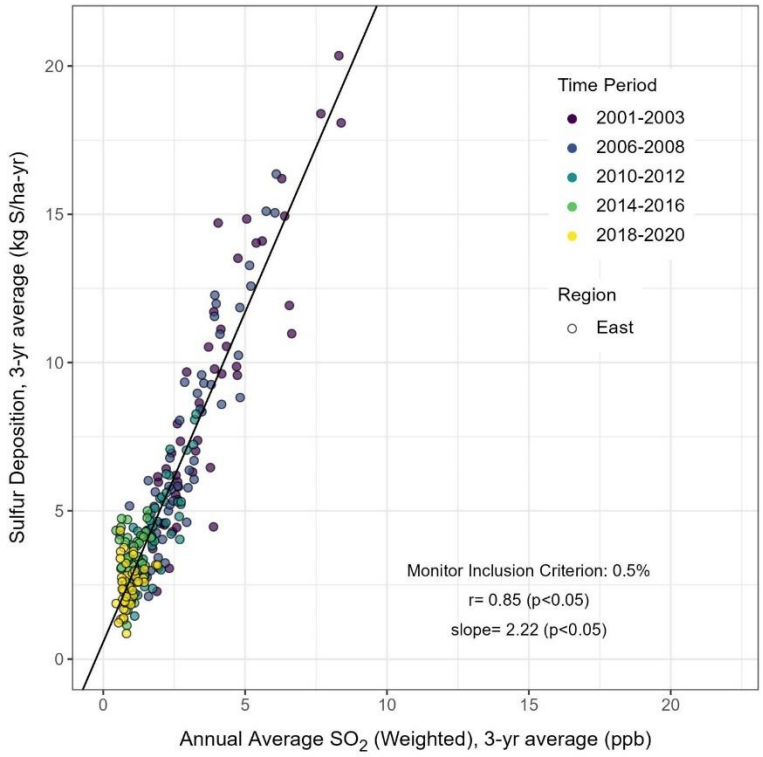

Figure 6A-62. Annual SO_2 EAQM-weighted values and TDep S deposition in eastern ecoregions (120-hr trajectories, NAM-12, 0.5% monitor inclusion criteria).

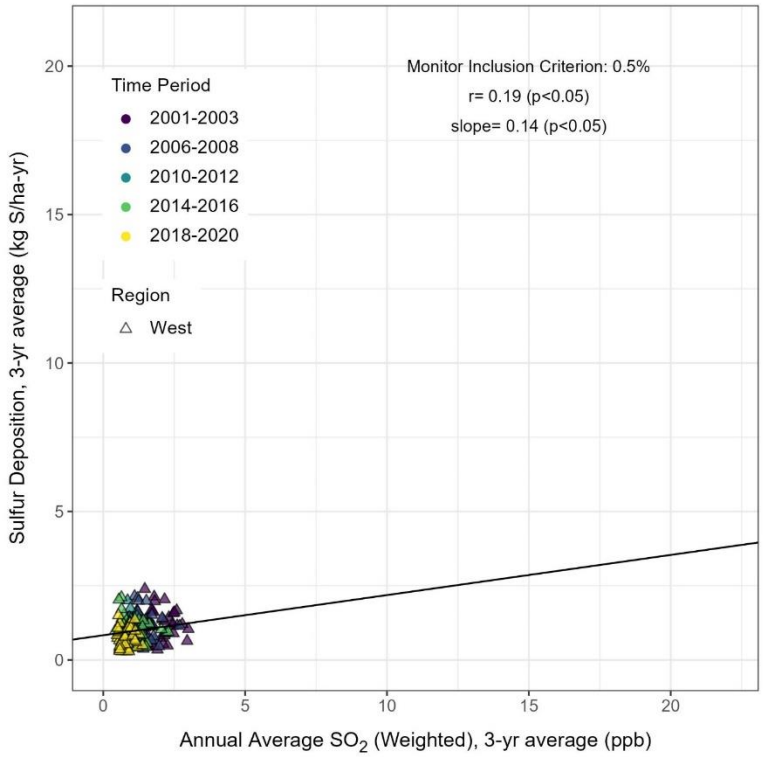

Figure 6A-63. Annual SO_2 EAQM-weighted values and TDep S deposition in western ecoregions (120-hr trajectories, NAM-12, 0.5% monitor inclusion criteria).

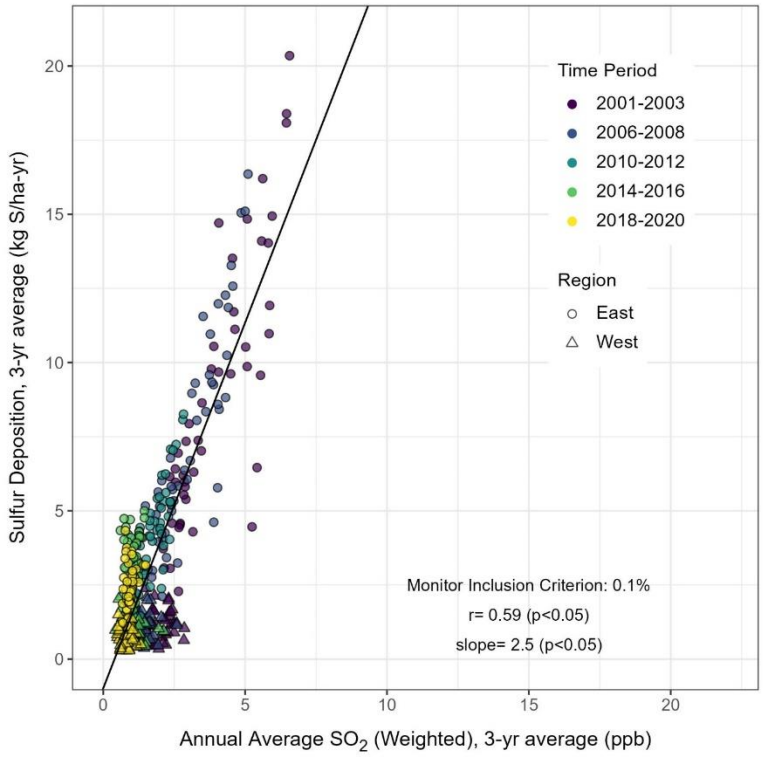

Figure 6A-64. Annual SO_2 EAQM-weighted values and TDep S deposition in 84 ecoregions (120-hr trajectories, NAM-12, 0.1% monitor inclusion criteria).

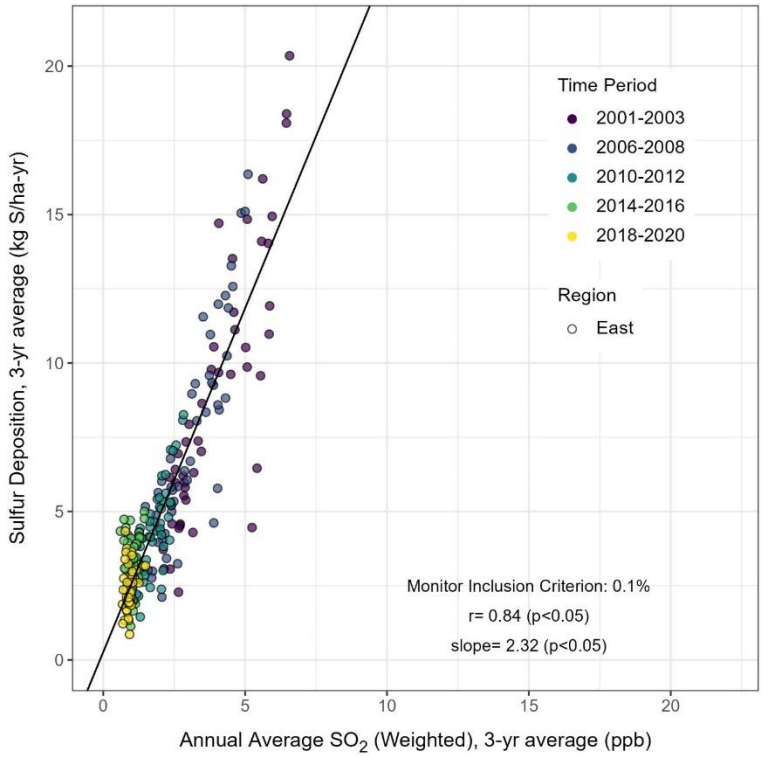

Figure 6A-65. Annual SO_2 EAQM-weighted values and TDep S deposition in eastern ecoregions (120-hr trajectories, NAM-12, 0.1% monitor inclusion criteria).

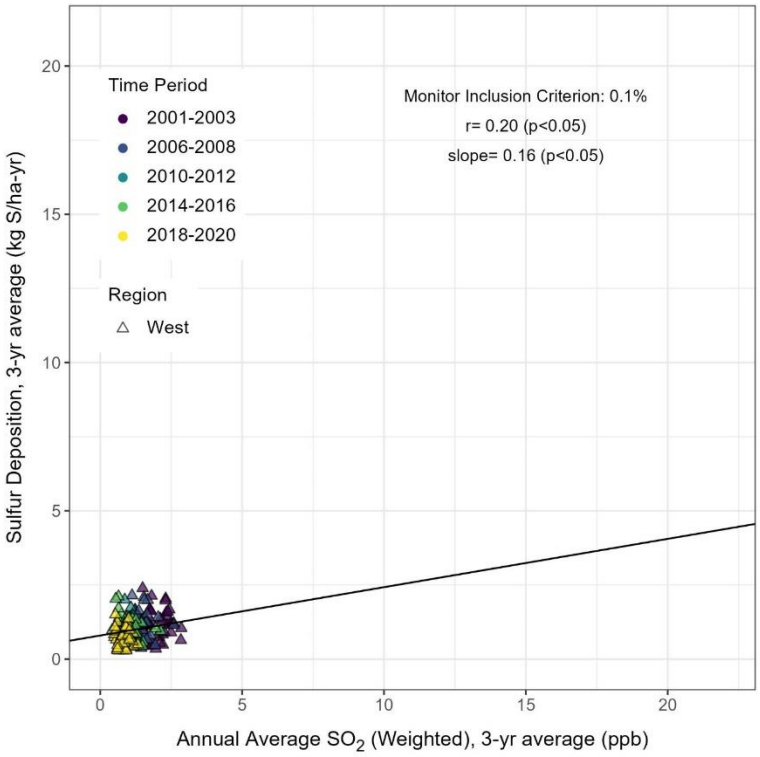

Figure 6A-66. Annual SO_2 EAQM-weighted values and TDep S deposition in western ecoregions (120-hr trajectories, NAM-12, 0.1% monitor inclusion criteria).

6A.6.4. SO_2 Annual Metric – 48-hr

Table 6A-5. Correlation coefficients of TDep estimates of sulfur deposition and annual SO_2 EAQMs generated by HYSPLIT analysis. Data are also split by year and by region (East/West), 48-hr trajectories.

Sulfur Deposition and SO_2					
Annual Max-All Ecoregions- Monitor Inclusion Criteria: 1%	Correlation Coefficient (r) = 0.32*	*Annual Max-All Ecoregions- Monitor Inclusion Criteria: 1%* [a]	Correlation Coefficient (r) = 0.32*	*Weighted Annual Average-All Ecoregions- Monitor Inclusion Criteria: 1%*	Correlation Coefficient (r) = 0.45*
Year	r	Year	r	Year	r
2001 - 2003	0.55*	2001 - 2003	0.55*	2001 - 2003	0.63*
2006 - 2008	0.59*	2006 - 2008	0.59*	2006 - 2008	0.71*
2010 - 2012	0.24*	2010 - 2012	0.24*	2010 - 2012	0.56*
2014 - 2016	-0.21	2014 - 2016	-0.21	2014 - 2016	0.04
2018 - 2020	-0.22*	2018 - 2020	-0.39*	2018 - 2020	-0.01
Annual Max-East Ecoregions- Monitor Inclusion Criteria: 1%	r = 0.67*	*Annual Max-East Ecoregions- Monitor Inclusion Criteria: 1%* [a]	r = 0.73*	*Weighted Annual Average-East Ecoregions- Monitor Inclusion Criteria: 1%*	r = 0.85*
Year	r	Year	r	Year	r
2001 - 2003	0.75*	2001 - 2003	0.75*	2001 - 2003	0.88*
2006 - 2008	0.69*	2006 - 2008	0.69*	2006 - 2008	0.89*
2010 - 2012	0.23	2010 - 2012	0.23	2010 - 2012	0.75*
2014 - 2016	-0.02	2014 - 2016	-0.02	2014 - 2016	0.33*
2018 - 2020	0.09	2018 - 2020	-0.05	2018 - 2020	0.23
Annual Max-West Ecoregions- Monitor Inclusion Criteria: 1%	r = -0.13	*Annual Max-West Ecoregions- Monitor Inclusion Criteria: 1%*	No outliers in West dataset	*Weighted Annual Average-West Ecoregions- Monitor Inclusion Criteria: 1%*	r = 0.07
Year	r	Year	r	Year	r
2001 - 2003	-0.25	2001 - 2003		2001 - 2003	-0.14
2006 - 2008	-0.22	2006 - 2008		2006 - 2008	-0.23
2010 - 2012	-0.27	2010 - 2012		2010 - 2012	-0.27
2014 - 2016	-0.21	2014 - 2016		2014 - 2016	-0.29
2018 - 2020	-0.25	2018 - 2020		2018 - 2020	-0.06

*p< 0.05

[a] Note: There are several outlier points in this comparison where the EAQM-max annual average SO_2 value exceeds 20 ppb in the 2018-2020 period. These points have been removed from these analyses. These data are driven by a monitor in southeastern MO where annual average SO_2 has exceeded 20 ppb in recent years. Any downwind ecoregion that is linked to this upwind monitor will have an EAQM-max with this value. A preliminary analysis suggests that these observed SO_2 data are due to a new source that was not modeled in the CMAQ simulation that informed the TDep estimates of deposition. As there is no deposition monitor in the immediate vicinity of the source it is unlikely that the TDep estimates are capturing the impacts of this source. For that reason, we concluded it was appropriate not to consider these data in our evaluation of the concentration-deposition relationship.

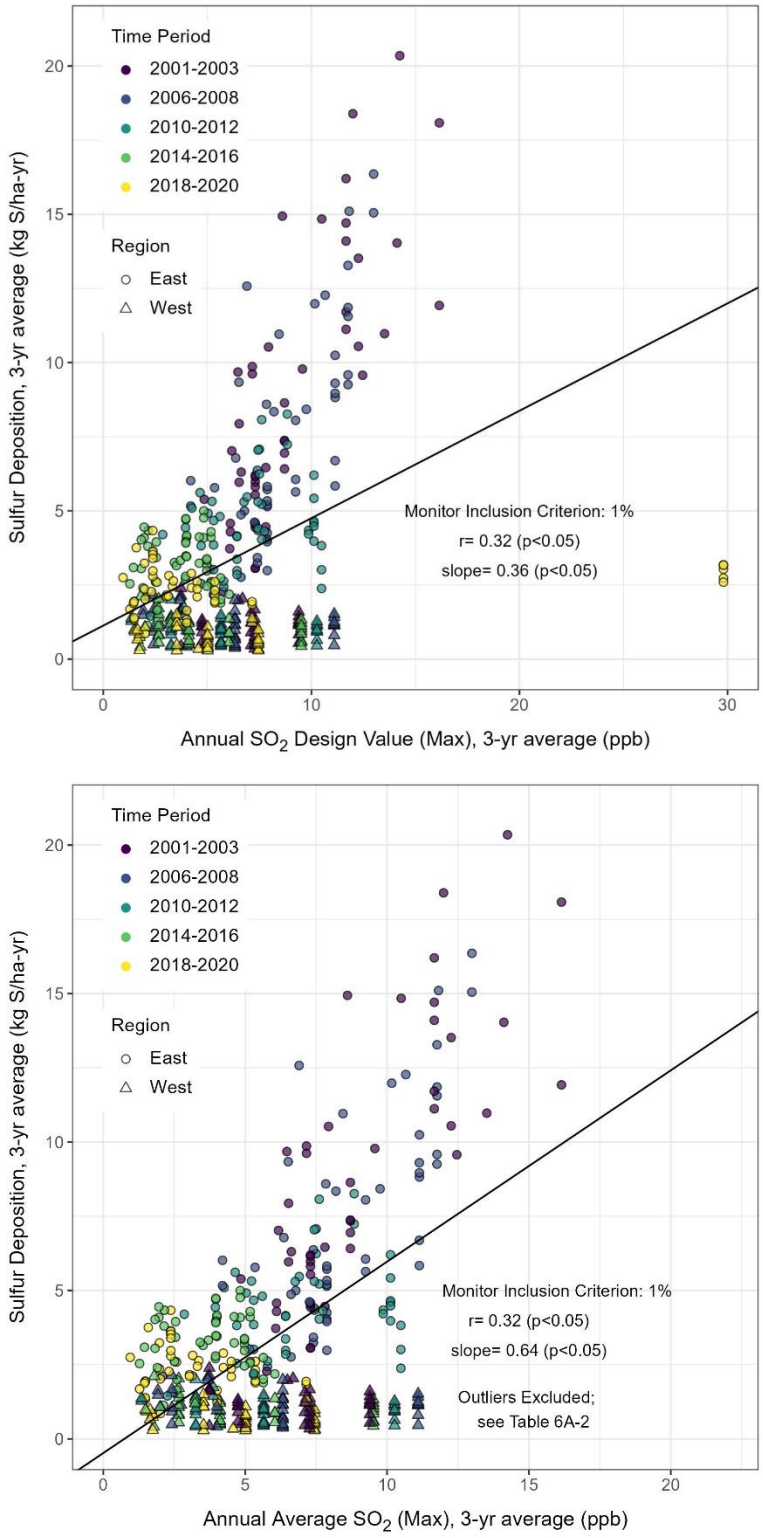

Figure 6A-67. Annual SO_2 EAQM-max values and TDep S deposition in 84 ecoregions (48-hr trajectories, NARR-32, 1% monitor inclusion criteria): all values (upper), outliers excluded (lower).

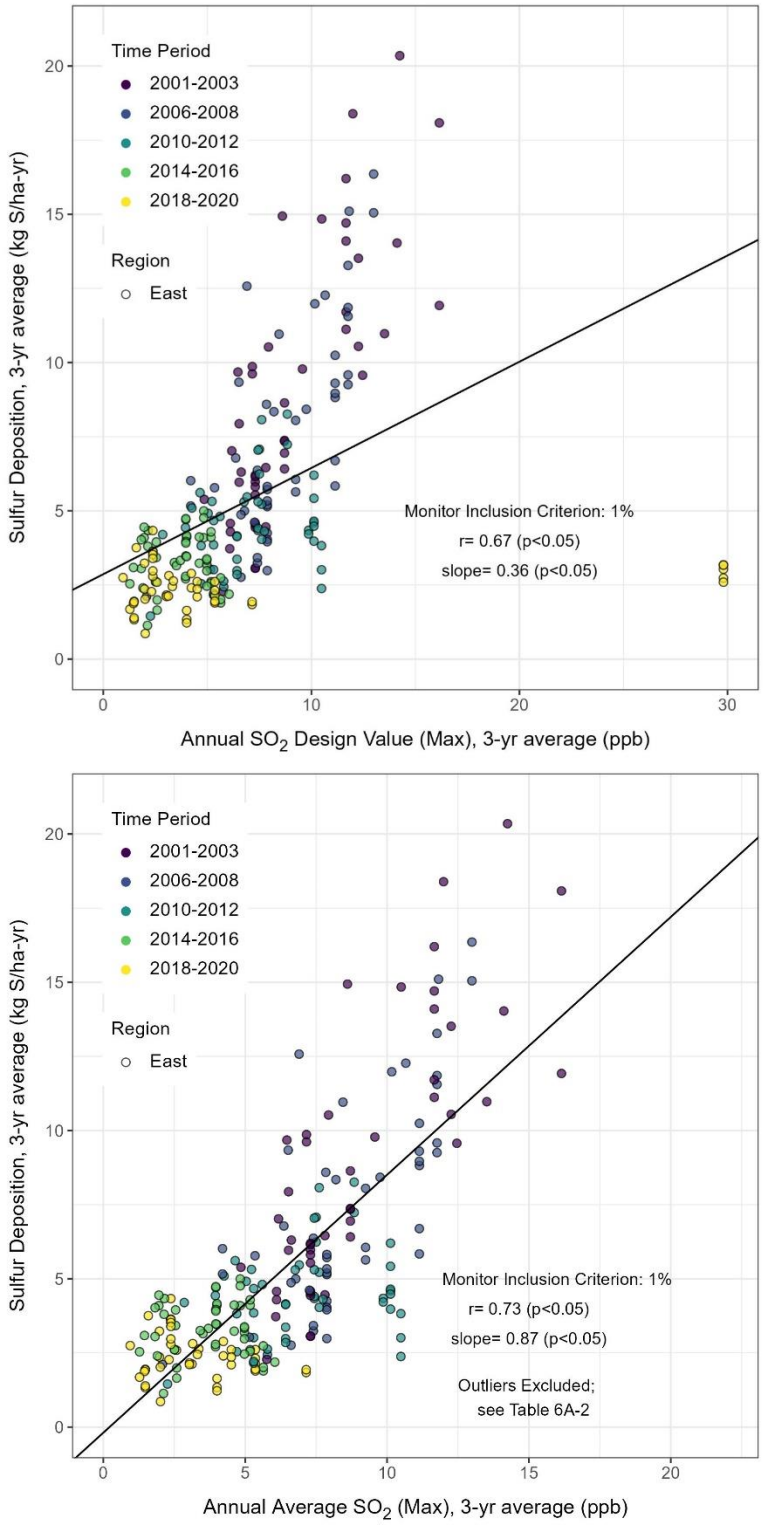

Figure 6A-68. Annual SO_2 EAQM-max values and TDep S deposition in eastern ecoregions (48-hr trajectories, NARR-32, 1% monitor inclusion criteria): all values (upper), outliers excluded (lower).

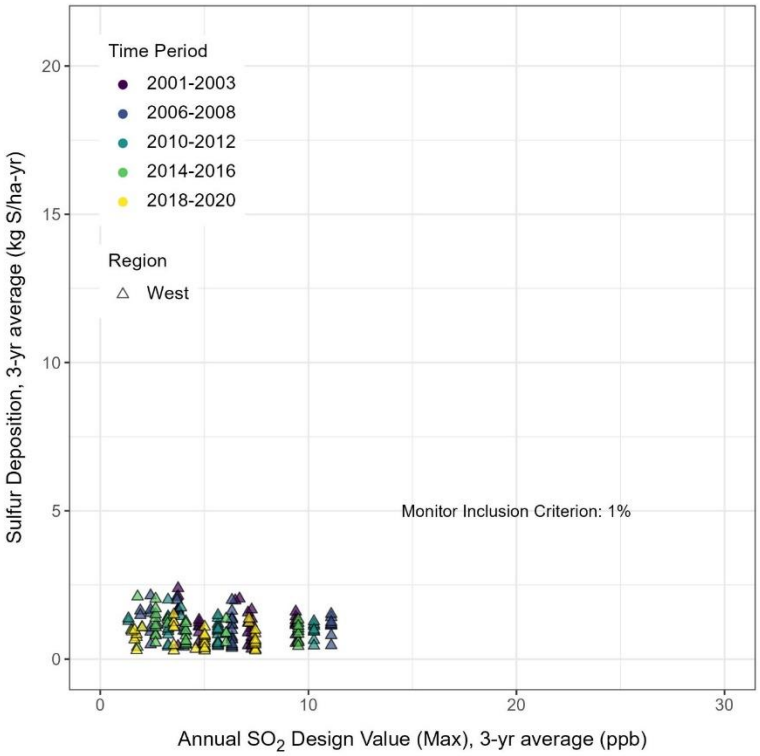

Figure 6A-69. Annual SO_2 EAQM-max values and TDep S deposition in western ecoregions (48-hr trajectories, NARR-32, 1% monitor inclusion criteria).

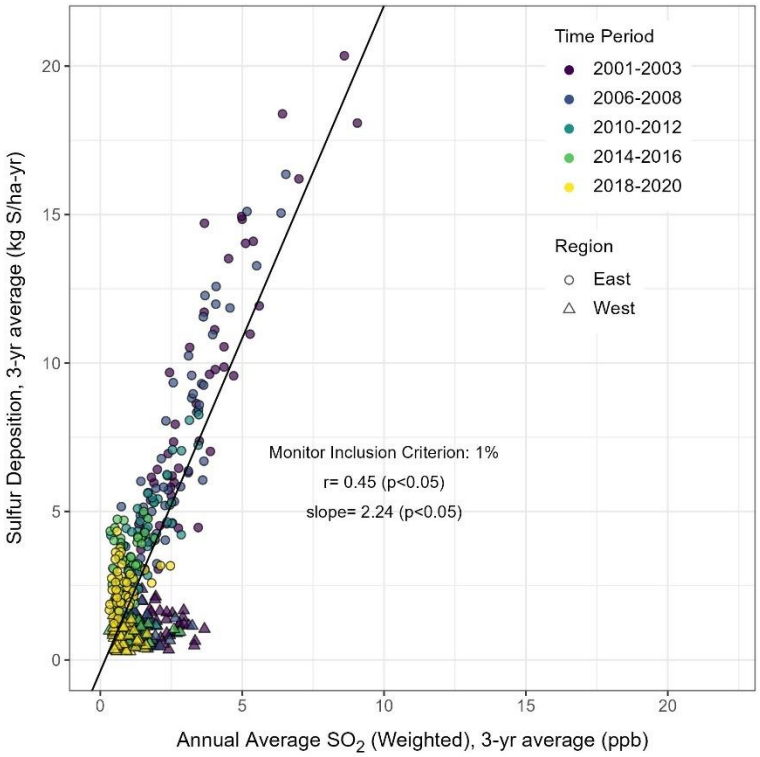

Figure 6A-70. Annual SO_2 EAQM-weighted values and TDep S deposition in 84 ecoregions (48-hr trajectories, NARR-32, 1% monitor inclusion criteria).

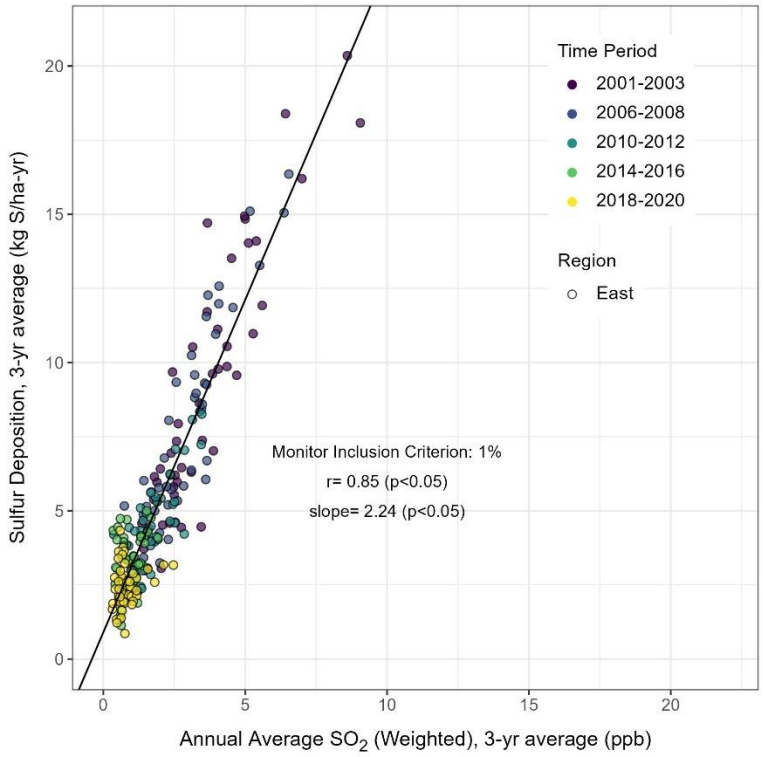

Figure 6A-71. Annual SO_2 EAQM-weighted values and TDep S deposition in eastern ecoregions (48-hr trajectories, NARR-32, 1% monitor inclusion criteria).

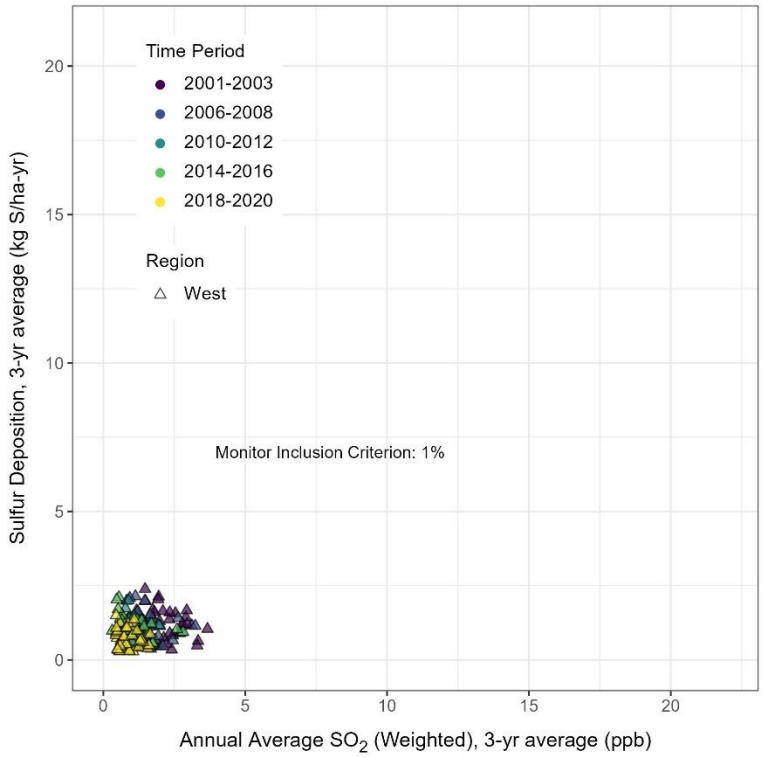

Figure 6A-72. Annual SO_2 EAQM-weighted values and TDep S deposition in western ecoregions (48-hr trajectories, NARR-32, 1% monitor inclusion criteria).

6A.6.5. NO_2 Annual Metric – 120-hr

Table 6A-6. Correlation coefficients of TDep estimates of nitrogen deposition and annual NO_2 EAQMs generated by HYSPLIT analysis, 120-hr trajectories. Data are also split by year and by region (East/West).

Nitrogen Deposition and NO_2					
Annual Max-All Ecoregions- Monitor Inclusion Criteria: 1%	Correlation Coefficient (r) = -0.12*	*Annual Max-All Ecoregions- Monitor Inclusion Criteria: 0.5%*	Correlation Coefficient (r) = -0.17*	*Annual Max-All Ecoregions- Monitor Inclusion Criteria: 0.1%*	Correlation Coefficient (r) = 0.02
Year	r	Year	r	Year	r
2001 - 2003	-0.14	2001 - 2003	-0.31*	2001 - 2003	-0.67*
2006 - 2008	-0.06	2006 - 2008	0.05	2006 - 2008	0.65*
2010 - 2012	-0.26*	2010 - 2012	-0.26*	2010 - 2012	-0.06
2014 - 2016	-0.28*	2014 - 2016	-0.41*	2014 - 2016	-0.65*
2018 - 2020	-0.37*	2018 - 2020	-0.58*	2018 - 2020	-0.58*
Annual Max-East Ecoregions- Monitor Inclusion Criteria: 1%	r = 0.42*	*Annual Max-East Ecoregions- Monitor Inclusion Criteria: 0.5%*	r = 0.35*	*Annual Max-East Ecoregions- Monitor Inclusion Criteria: 0.1%*	r = 0.44*
Year	r	Year	r	Year	r
2001 - 2003	0.41*	2001 - 2003	0.24*	2001 - 2003	-0.12
2006 - 2008	0.47*	2006 - 2008	0.35*	2006 - 2008	0.38*
2010 - 2012	0.29*	2010 - 2012	0.15	2010 - 2012	0.06
2014 - 2016	0.20	2014 - 2016	0.03	2014 - 2016	0.16
2018 - 2020	0.11	2018 - 2020	0.02	2018 - 2020	0.29*
Annual Max-West Ecoregions- Monitor Inclusion Criteria: 1%	r = 0.02	*Annual Max-West Ecoregions- Monitor Inclusion Criteria: 0.5%*	r = -0.04	*Annual Max-West Ecoregions- Monitor Inclusion Criteria: 0.1%*	r = 0.07
Year	r	Year	r	Year	r
2001 - 2003	-0.00	2001 - 2003	-0.12	2001 - 2003	-0.32
2006 - 2008	-0.02	2006 - 2008	-0.05	2006 - 2008	0.24
2010 - 2012	-0.16	2010 - 2012	0.02	2010 - 2012	-0.16
2014 - 2016	-0.00	2014 - 2016	-0.19	2014 - 2016	-0.15
2018 - 2020	-0.00	2018 - 2020	-0.25	2018 - 2020	0.0
Weighted Annual Average-All Ecoregions- Monitor Inclusion Criteria: 1%	r = 0.03	*Weighted Annual Average-All Ecoregions- Monitor Inclusion Criteria: 0.5%*	r = -0.06	*Weighted Annual Average-All Ecoregions- Monitor Inclusion Criteria: 0.1%*	r = -0.10*
Year	r	Year	r	Year	r
2001 - 2003	0.0	2001 - 2003	-0.1	2001 - 2003	-0.22*
2006 - 2008	-0.08	2006 - 2008	-0.21	2006 - 2008	-0.35*
2010 - 2012	-0.03	2010 - 2012	-0.14	2010 - 2012	-0.26*
2014 - 2016	-0.07	2014 - 2016	-0.20	2014 - 2016	-0.34*
2018 - 2020	-0.2	2018 - 2020	-0.37*	2018 - 2020	-0.52*
Weighted Annual Average-East Ecoregions- Monitor Inclusion Criteria: 1%	r = 0.55*	*Weighted Annual Average-East Ecoregions- Monitor Inclusion Criteria: 0.5%*	r = 0.48*	*Weighted Annual Average-East Ecoregions- Monitor Inclusion Criteria: 0.1%*	r = 0.45*
Year	r	Year	r	Year	r
2001 - 2003	0.62*	2001 - 2003	0.61*	2001 - 2003	0.62*
2006 - 2008	0.54*	2006 - 2008	0.39*	2006 - 2008	0.45*
2010 - 2012	0.42*	2010 - 2012	0.32*	2010 - 2012	0.26

2014 - 2016	0.38*	2014 - 2016	0.21	2014 - 2016	0.07
2018 - 2020	0.09	2018 - 2020	-0.03	2018 - 2020	-0.14
Weighted Annual Average-West Ecoregions- Monitor Inclusion Criteria: 1%	r = -0.12	*Weighted Annual Average-West Ecoregions- Monitor Inclusion Criteria: 0.5%*	r = -0.17*	*Weighted Annual Average-West Ecoregions- Monitor Inclusion Criteria: 0.1%*	r = -0.11
Year	r	Year	r	Year	r
2001 - 2003	-0.01	2001 - 2003	-0.22	2001 - 2003	-0.23
2006 - 2008	-0.21	2006 - 2008	-0.23	2006 - 2008	-0.27
2010 - 2012	-0.28	2010 - 2012	-0.34*	2010 - 2012	-0.34*
2014 - 2016	-0.16	2014 - 2016	-0.28	2014 - 2016	-0.27
2018 - 2020	-0.10	2018 - 2020	-0.26	2018 - 2020	-0.25
*p< 0.05					

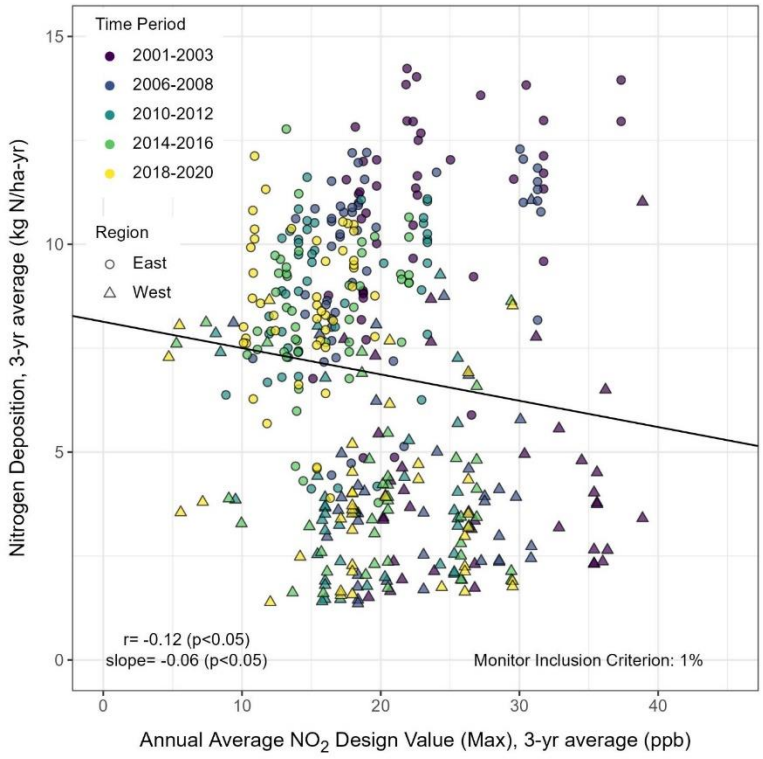

Figure 6A-73. Annual NO_2 EAQM-max values and TDep N deposition in 84 ecoregions (120-hr trajectories, NAM-12, 1% monitor inclusion criteria).

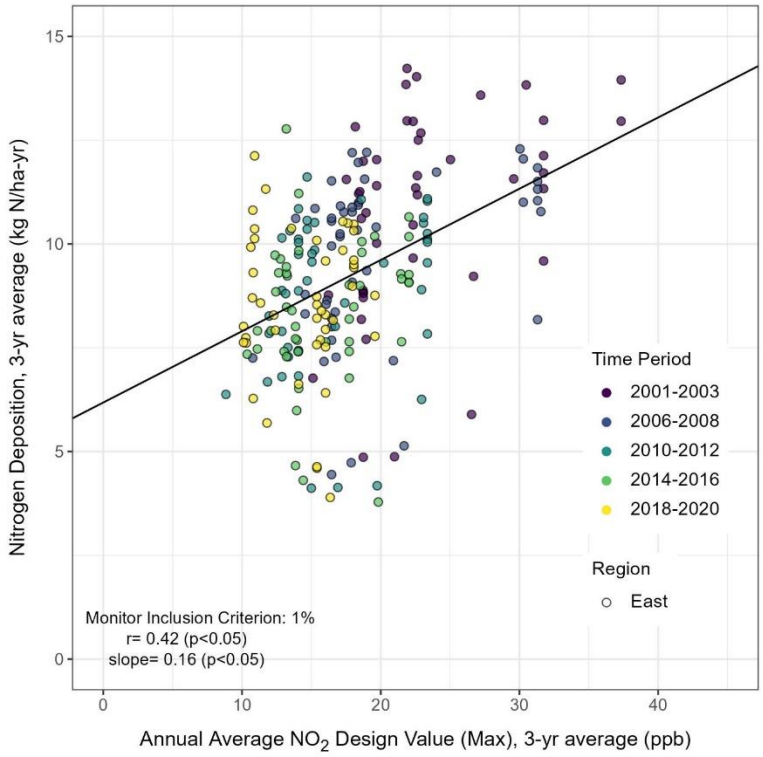

Figure 6A-74. Annual NO_2 EAQM-max values and TDep N deposition in eastern ecoregions (120-hr trajectories, NAM-12, 1% monitor inclusion criteria).

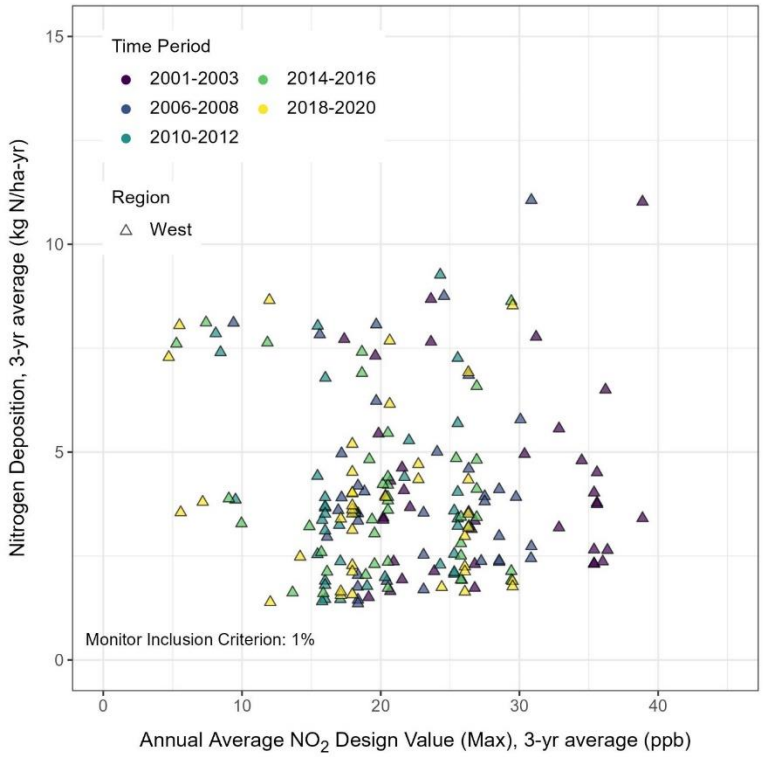

Figure 6A-75. Annual NO_2 EAQM-max values and TDep N deposition in western ecoregions (120-hr trajectories, NAM-12, 1% monitor inclusion criteria).

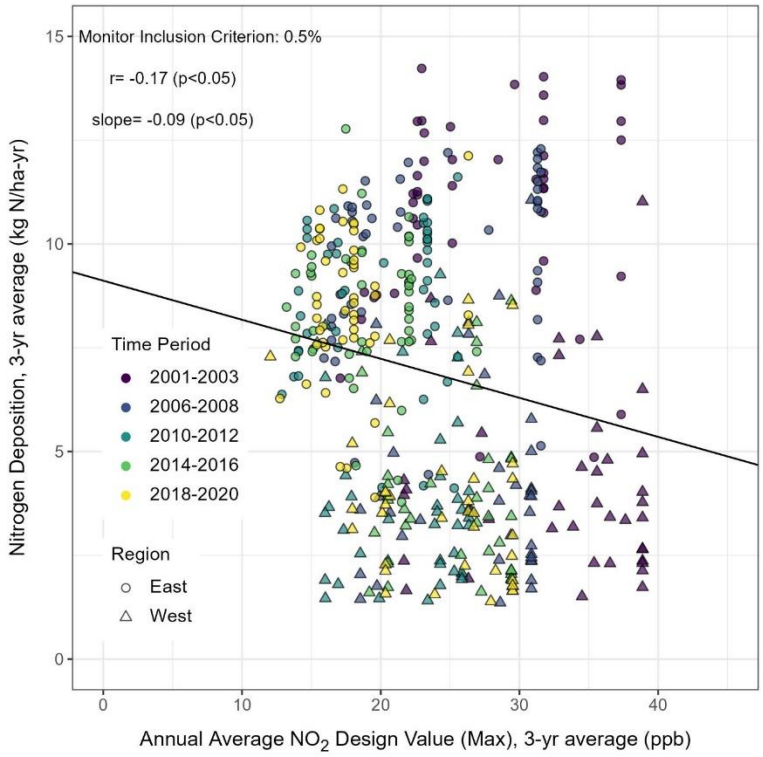

Figure 6A-76. Annual NO_2 EAQM-max values and TDep N deposition in 84 ecoregions (120-hr trajectories, NAM-12, 0.5% monitor inclusion criteria).

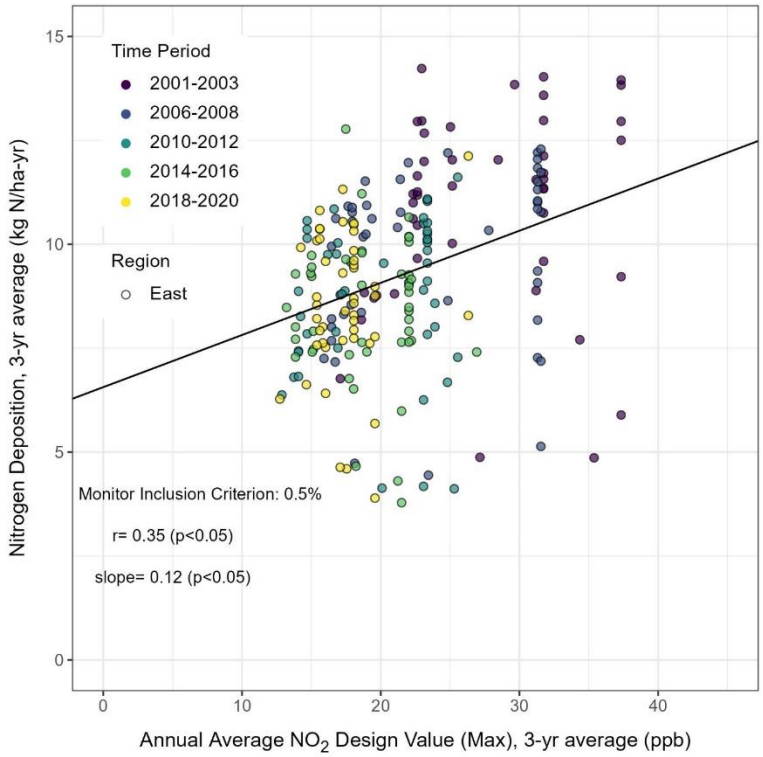

Figure 6A-77. Annual NO_2 EAQM-max values and TDep N deposition in eastern ecoregions (120-hr trajectories, NAM-12, 0.5% monitor inclusion criteria).

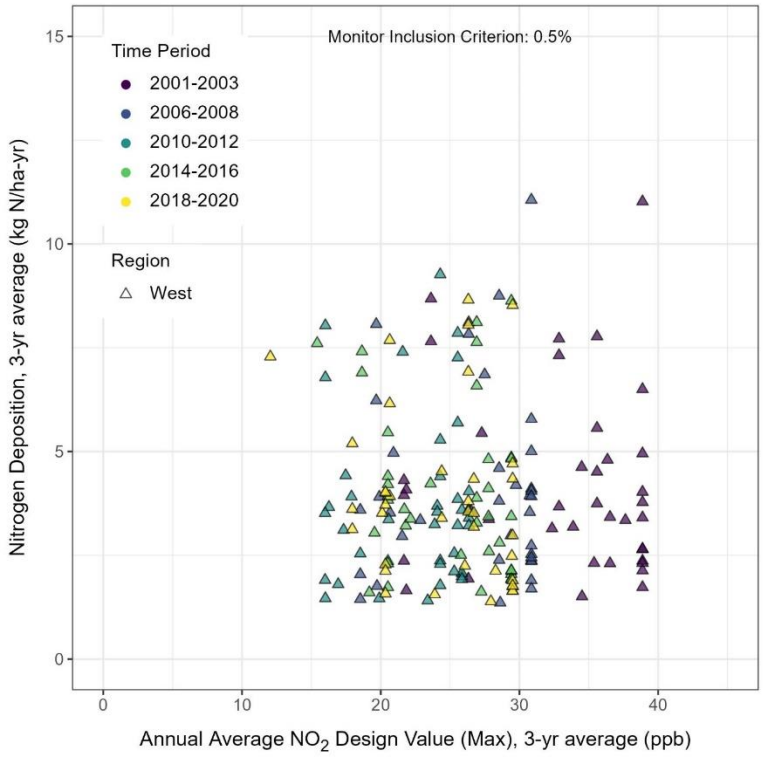

Figure 6A-78. Annual NO_2 EAQM-max values and TDep N deposition in western ecoregions (120-hr trajectories, NAM-12, 0.5% monitor inclusion criteria).

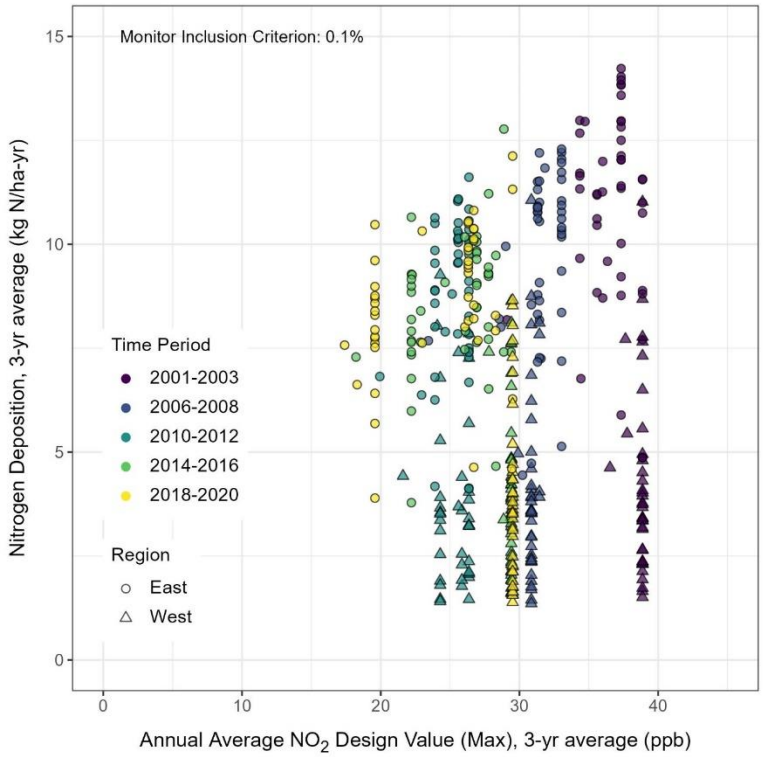

Figure 6A-79. Annual NO_2 EAQM-max values and TDep N deposition in 84 ecoregions (120-hr trajectories, NAM-12, 0.1% monitor inclusion criteria).

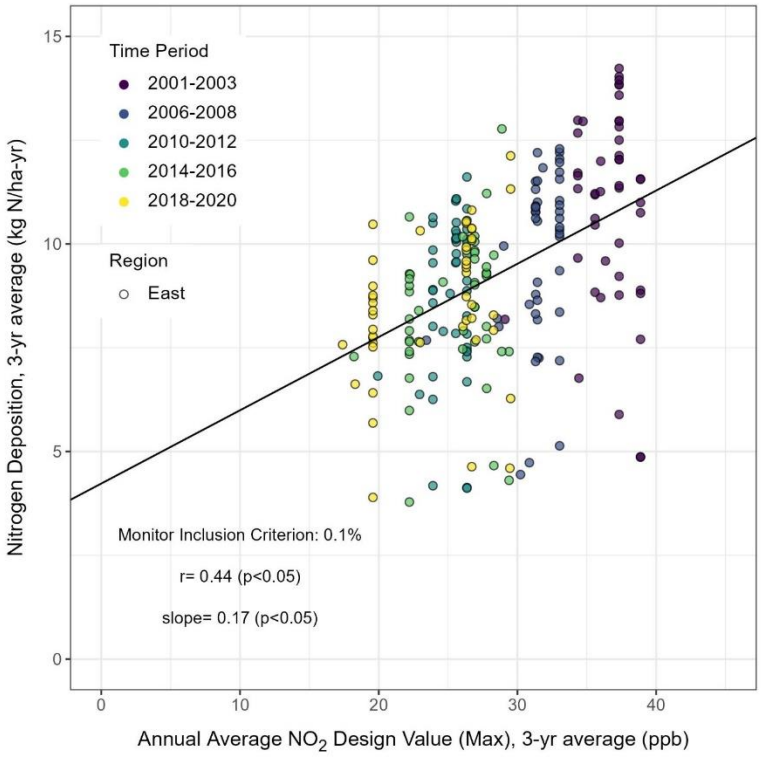

Figure 6A-80. Annual NO_2 EAQM-max values and TDep N deposition in eastern ecoregions (120-hr trajectories, NAM-12, 0.1% monitor inclusion criteria).

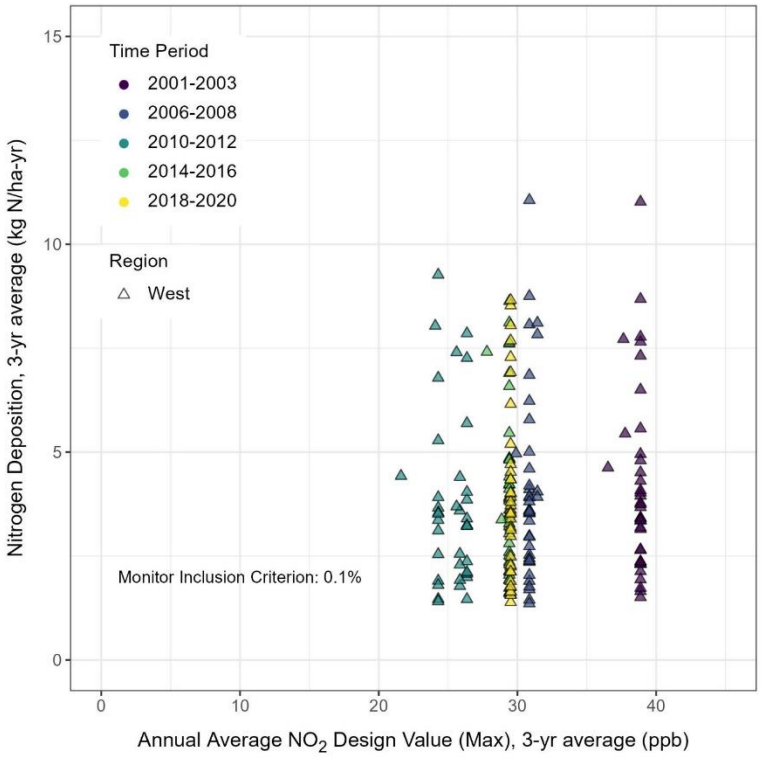

Figure 6A-81. Annual NO_2 EAQM-max values and TDep N deposition in western ecoregions (120-hr trajectories, NAM-12, 0.1% monitor inclusion criteria).

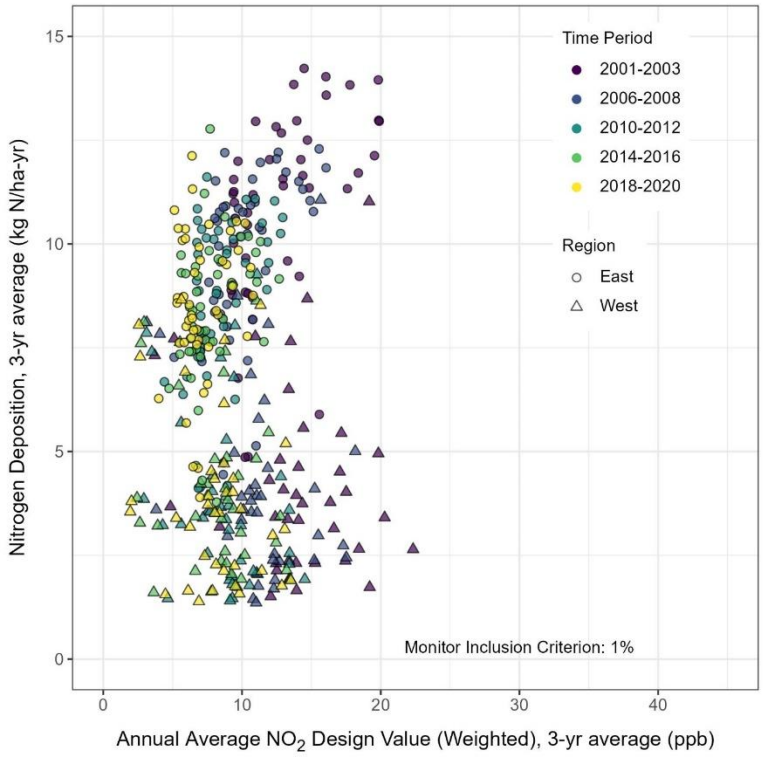

Figure 6A-82. Annual NO_2 EAQM-weighted values and TDep N deposition in 84 ecoregions (120-hr trajectories, NAM-12, 1% monitor inclusion criteria).

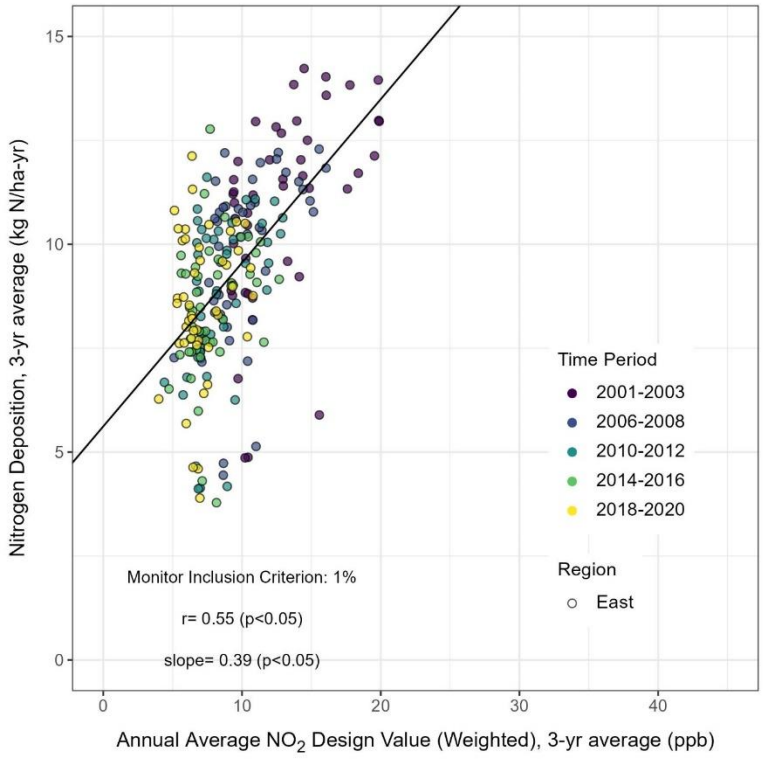

Figure 6A-83. Annual NO_2 EAQM-weighted values and TDep N deposition in eastern ecoregions (120-hr trajectories, NAM-12, 1% monitor inclusion criteria).

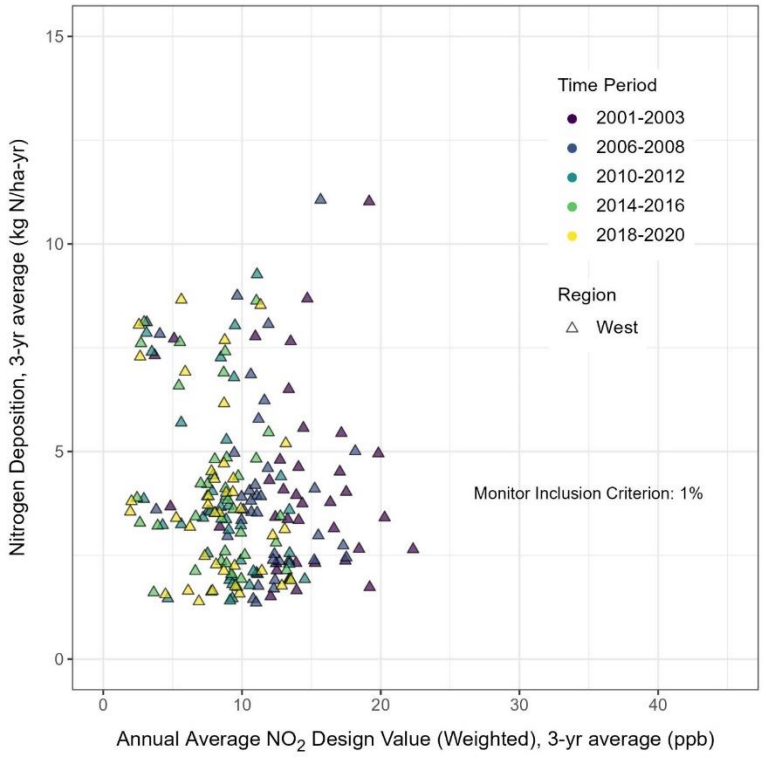

Figure 6A-84. Annual NO_2 EAQM-weighted values and TDep N deposition in western ecoregions (120-hr trajectories, NAM-12, 1% monitor inclusion criteria).

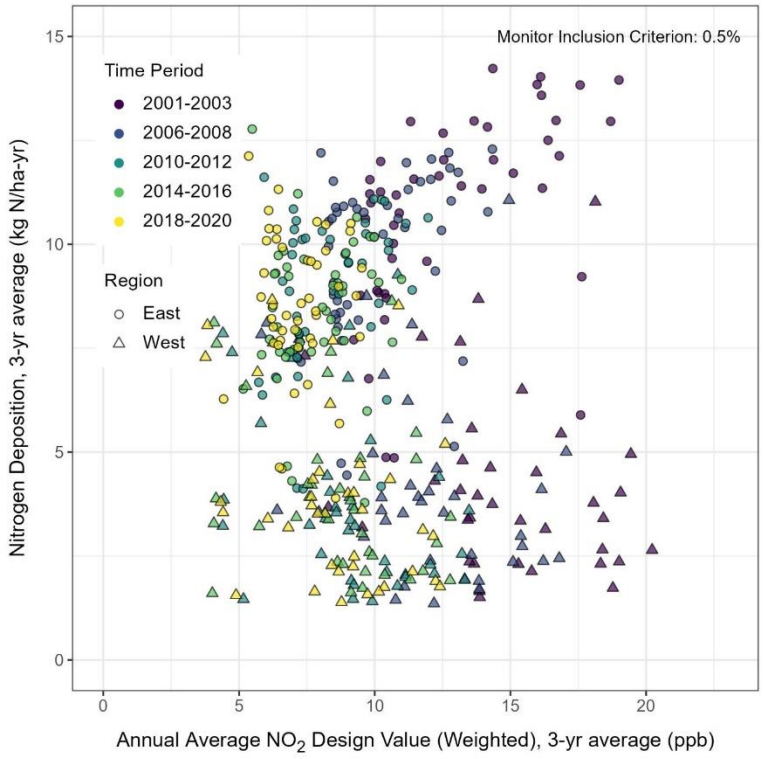

Figure 6A-85. Annual NO_2 EAQM-weighted values and TDep N deposition in 84 ecoregions (120-hr trajectories, NAM-12, 0.5% monitor inclusion criteria).

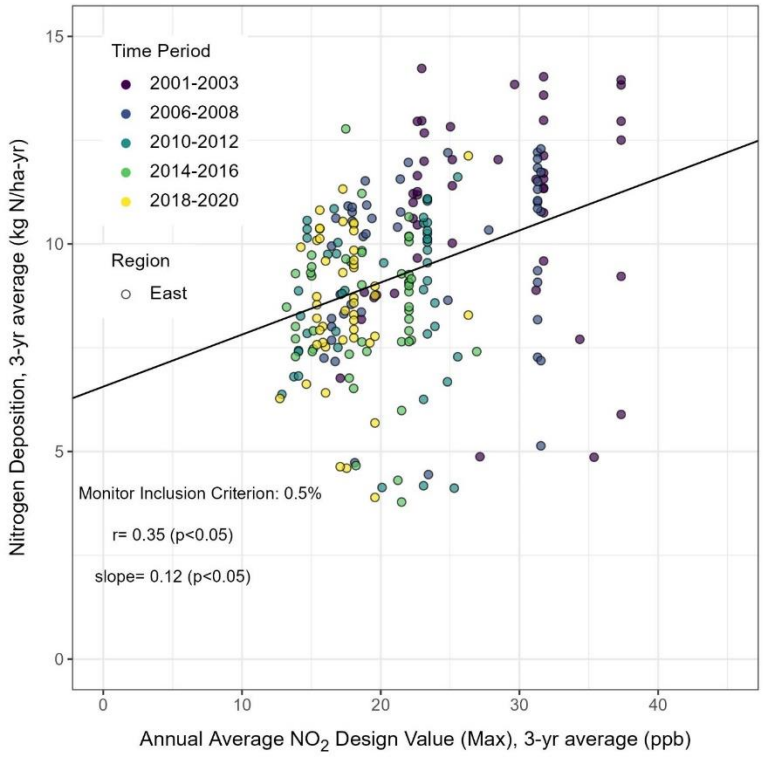

Figure 6A-86. Annual NO_2 EAQM-max values and TDep N deposition in eastern ecoregions (120-hr trajectories, NAM-12, 0.5% monitor inclusion criteria).

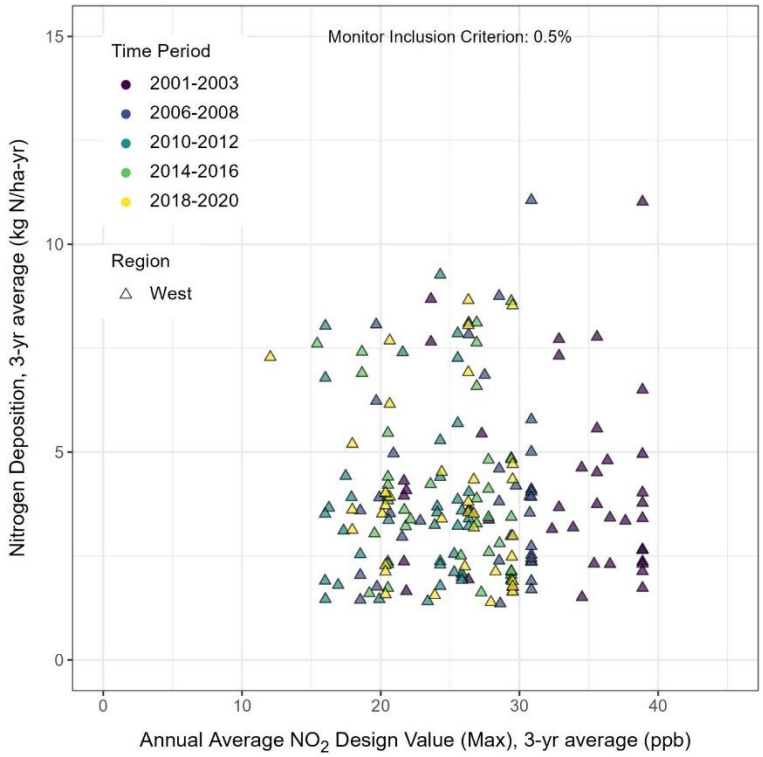

Figure 6A-87. Annual NO_2 EAQM-max values and TDep N deposition in western ecoregions (120-hr trajectories, NAM-12, 0.5% monitor inclusion criteria).

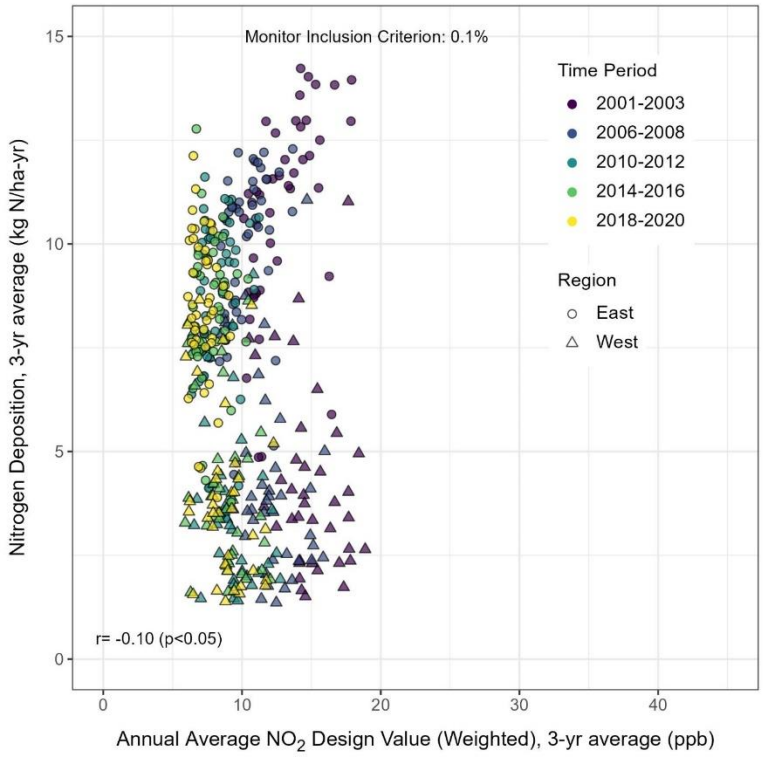

Figure 6A-88. Annual NO_2 EAQM-weighted values and TDep N deposition in 84 ecoregions (120-hr trajectories, NAM-12, 0.1% monitor inclusion criteria).

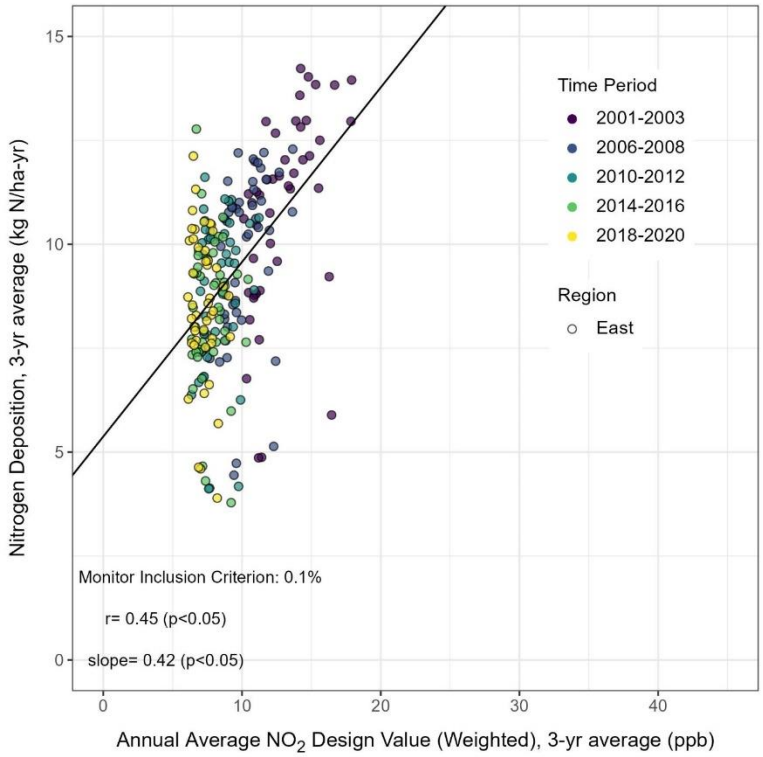

Figure 6A-89. Annual NO_2 EAQM-weighted values and TDep N deposition in eastern ecoregions (120-hr trajectories, NAM-12, 0.1% monitor inclusion criteria).

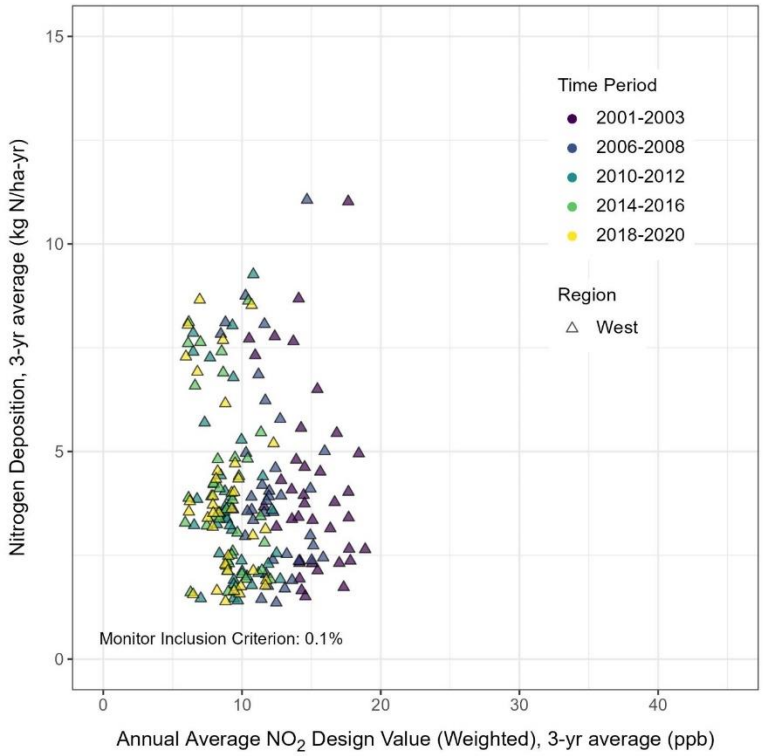

Figure 6A-90. Annual NO_2 EAQM-weighted values and TDep N deposition in western ecoregions (120-hr trajectories, NAM-12, 0.1% monitor inclusion criteria).

6A.6.6. NO_2 Annual Metric – 48-hr

Table 6A-7. Correlation coefficients of TDep estimates of nitrogen deposition and annual NO_2 EAQMs generated by HYSPLIT analysis, 48-hr trajectories. Data are also split by year and by region (East/West).

Nitrogen Deposition and NO_2			
Annual Max-All Ecoregions- Monitor Inclusion Criteria: 1%	Correlation Coefficient (r) = -0.06	*Weighted Annual Average-All Ecoregions- Monitor Inclusion Criteria: 1%*	Correlation Coefficient (r) = 0.06
Year	r	Year	r
2001 - 2003	-0.05	2001 - 2003	0.01
2006 - 2008	0.01	2006 - 2008	-0.03
2010 - 2012	-0.16	2010 - 2012	0.01
2014 - 2016	-0.25*	2014 - 2016	-0.05
2018 - 2020	-0.36*	2018 - 2020	-0.15
Annual Max-East Ecoregions- Monitor Inclusion Criteria: 1%	r = 0.42*	*Weighted Annual Average-East Ecoregions- Monitor Inclusion Criteria: 1%*	r = 0.56*
Year	r	Year	r
2001 - 2003	0.38*	2001 - 2003	0.63*
2006 - 2008	0.35*	2006 - 2008	0.55*

2010 - 2012	0.34*	2010 - 2012	0.48*
2014 - 2016	0.29*	2014 - 2016	0.40*
2018 - 2020	0.13	2018 - 2020	0.13
Annual Max-West Ecoregions- Monitor Inclusion Criteria: 1%	r = -0.06	*Weighted Annual Average-West Ecoregions- Monitor Inclusion Criteria: 1%*	r = -0.13
Year	r	Year	r
2001 - 2003	-0.20	2001 - 2003	-0.17
2006 - 2008	-0.11	2006 - 2008	-0.26
2010 - 2012	-0.18	2010 - 2012	-0.24
2014 - 2016	-0.08	2014 - 2016	-0.15
2018 - 2020	-0.01	2018 - 2020	-0.06
*p< 0.05			

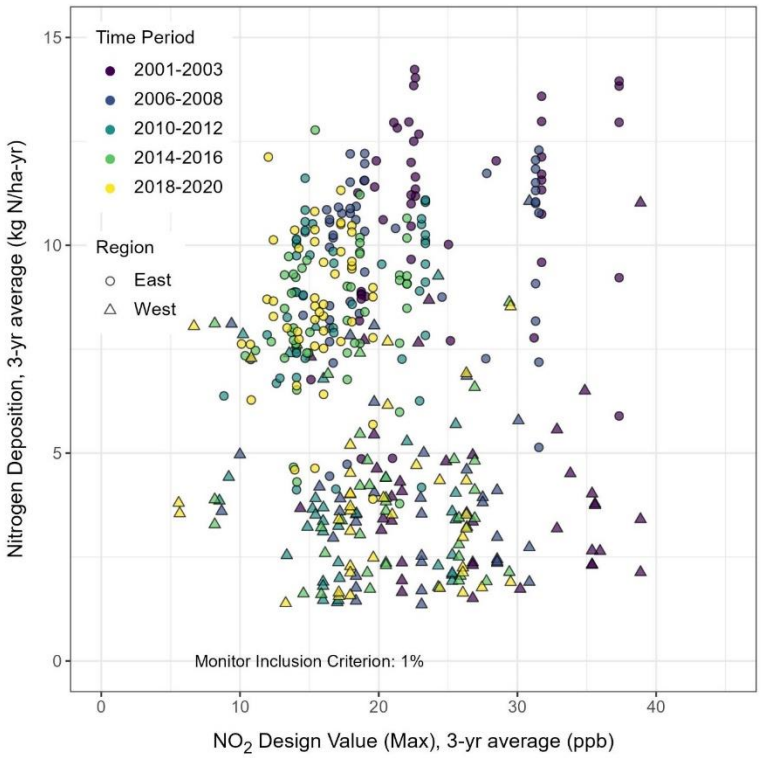

Figure 6A-91. Annual NO_2 EAQM-max values and TDep N deposition in 84 ecoregions (48-hr trajectories, NARR-32, 1% monitor inclusion criteria).

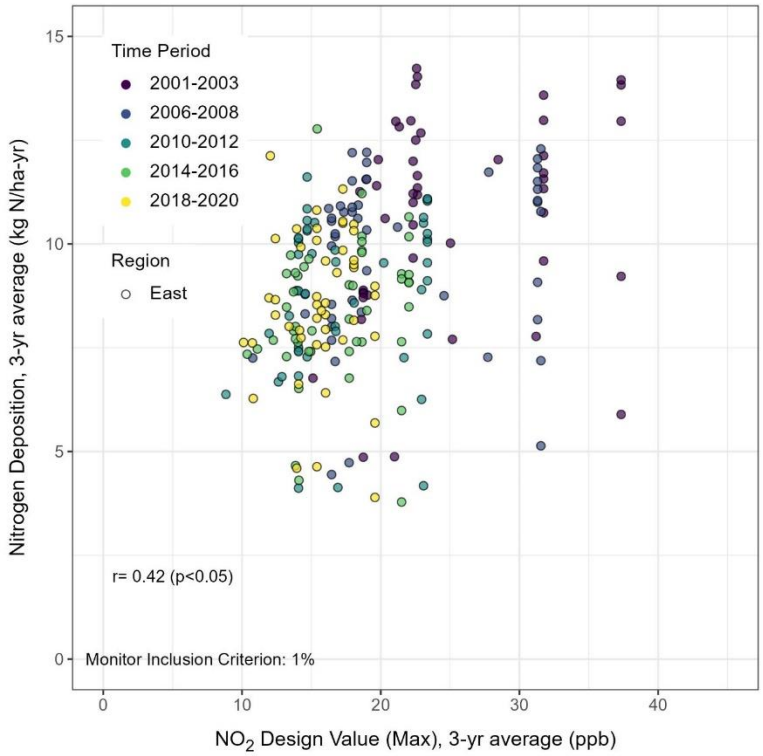

Figure 6A-92. Annual NO_2 EAQM-max values and TDep N deposition in eastern ecoregions (48-hr trajectories, NARR-32, 1% monitor inclusion criteria).

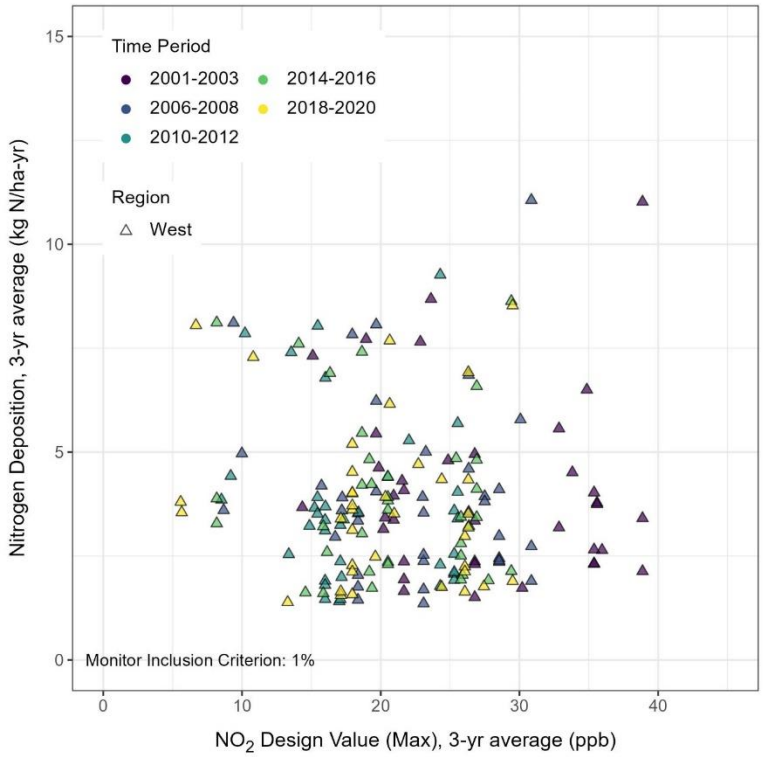

Figure 6A-93. Annual NO_2 EAQM-max values and TDep N deposition in western ecoregions (48-hr trajectories, NARR-32, 1% monitor inclusion criteria).

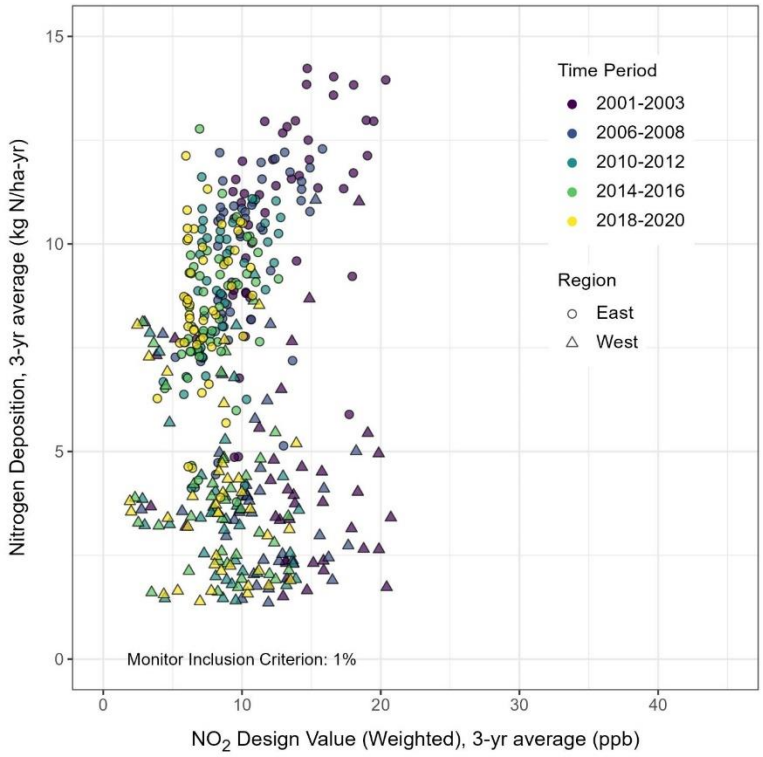

Figure 6A-94. Annual NO_2 EAQM-weighted values and TDep N deposition in 84 ecoregions (48-hr trajectories, NARR-32, 1% monitor inclusion criteria).

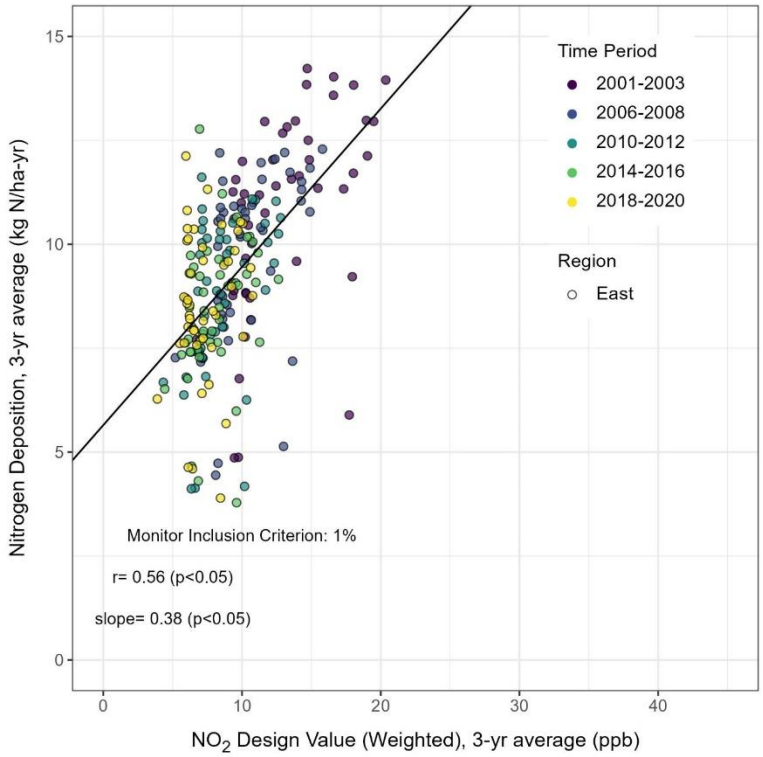

Figure 6A-95. Annual NO_2 EAQM-weighted values and TDep N deposition in eastern ecoregions (48-hr trajectories, NARR-32, 1% monitor inclusion criteria).

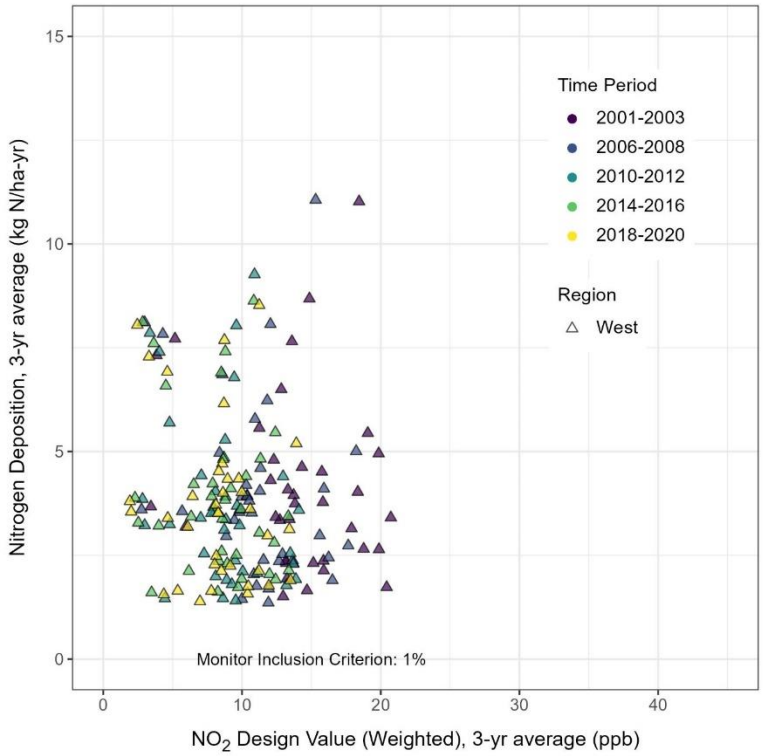

Figure 6A-96. Annual NO_2 EAQM-weighted values and TDep N deposition in western ecoregions (48-hr trajectories, NARR-32, 1% monitor inclusion criteria).

6A.6.7. $PM_{2.5}$ Annual Metric – 120-hr

6A.6.7.1. Nitrogen

Table 6A-8. Correlation coefficients of TDep estimates of nitrogen deposition and annual $PM_{2.5}$ EAQMs generated by HYSPLIT analysis, 120-hr trajectories. Data are also split by year and by region (East/West).

Nitrogen Deposition and $PM_{2.5}$					
Annual Max-All Ecoregions- Monitor Inclusion Criteria: 1%	Correlation Coefficient (r) = -0.13*	*Annual Max-All Ecoregions- Monitor Inclusion Criteria: 0.5%*	Correlation Coefficient (r) = -0.22*	*Annual Max-All Ecoregions- Monitor Inclusion Criteria: 0.1%*	Correlation Coefficient (r) = -0.38*
Year	r	Year	r	Year	r
2001 - 2003	-0.01	2001 - 2003	-0.12	2001 - 2003	-0.67*
2006 - 2008	0.03	2006 - 2008	-0.30*	2006 - 2008	-0.73*
2010 - 2012	0.16	2010 - 2012	-0.14	2010 - 2012	-0.75*
2014 - 2016	-0.37*	2014 - 2016	-0.46*	2014 - 2016	-0.76*
2018 - 2020	-0.47*	2018 - 2020	-0.49*	2018 - 2020	-0.72*
Annual Max-East Ecoregions- Monitor Inclusion Criteria: 1%	r = 0.47*	*Annual Max-East Ecoregions- Monitor Inclusion Criteria: 0.5%*	r = 0.53*	*Annual Max-East Ecoregions- Monitor Inclusion Criteria: 0.1%*	r = 0.29*
Year	r	Year	r	Year	r
2001 - 2003	0.53*	2001 - 2003	0.64*	2001 - 2003	0.08
2006 - 2008	0.29	2006 - 2008	0.34*	2006 - 2008	-0.17
2010 - 2012	0.38*	2010 - 2012	0.46*	2010 - 2012	-0.20
2014 - 2016	0.35*	2014 - 2016	0.27	2014 - 2016	-0.26
2018 - 2020	0.18	2018 - 2020	0.26	2018 - 2020	-0.09
Annual Max-West Ecoregions- Monitor Inclusion Criteria: 1%	r = -0.11	*Annual Max-West Ecoregions- Monitor Inclusion Criteria: 0.5%*	r = -0.12	*Annual Max-West Ecoregions- Monitor Inclusion Criteria: 0.1%*	r = -0.01
Year	r	Year	r	Year	r
2001 - 2003	-0.05	2001 - 2003	-0.18	2001 - 2003	0.07
2006 - 2008	-0.1	2006 - 2008	-0.22	2006 - 2008	-0.28
2010 - 2012	-0.21	2010 - 2012	-0.13	2010 - 2012	-0.23
2014 - 2016	-0.36*	2014 - 2016	-0.24	2014 - 2016	-0.37
2018 - 2020	-0.11	2018 - 2020	-0.07	2018 - 2020	-0.37*
Weighted Annual Average-All Ecoregions- Monitor Inclusion Criteria: 1%	r = 0.42*	*Weighted Annual Average-All Ecoregions- Monitor Inclusion Criteria: 0.5%*	r = 0.45*	*Weighted Annual Average-All Ecoregions- Monitor Inclusion Criteria: 0.1%*	r = 0.39*
Year	r	Year	r	Year	r
2001 - 2003	0.64*	2001 - 2003	0.65*	2001 - 2003	0.71*
2006 - 2008	0.60*	2006 - 2008	0.64*	2006 - 2008	0.67*
2010 - 2012	0.69*	2010 - 2012	0.75*	2010 - 2012	0.77*
2014 - 2016	0.39*	2014 - 2016	0.45*	2014 - 2016	0.40*
2018 - 2020	0.23	2018 - 2020	-0.09	2018 - 2020	-0.29*
Weighted Annual Average-East Ecoregions- Monitor Inclusion Criteria: 1%	r = 0.57*	*Weighted Annual Average-East Ecoregions- Monitor Inclusion Criteria: 0.5%*	r = 0.62*	*Weighted Annual Average-East Ecoregions- Monitor Inclusion Criteria: 0.1%*	r = 0.53*
Year	r	Year	r	Year	r
2001 - 2003	0.80*	2001 - 2003	0.85*	2001 - 2003	0.81*
2006 - 2008	0.70*	2006 - 2008	0.67*	2006 - 2008	0.53*

2010 - 2012	0.57*	2010 - 2012	0.60*	2010 - 2012	0.52*
2014 - 2016	0.37*	2014 - 2016	0.42*	2014 - 2016	0.29
2018 - 2020	0.26	2018 - 2020	0.27	2018 - 2020	0.17
Weighted Annual Average-West Ecoregions- Monitor Inclusion Criteria: 1%	r = 0.06	*Weighted Annual Average-West Ecoregions- Monitor Inclusion Criteria: 0.5%*	r = 0.02	*Weighted Annual Average-West Ecoregions- Monitor Inclusion Criteria: 0.1%*	r = 0.05
Year	r	Year	r	Year	r
2001 - 2003	0.08	2001 - 2003	-0.03	2001 - 2003	0.06
2006 - 2008	-0.08	2006 - 2008	-0.14	2006 - 2008	-0.09
2010 - 2012	0.11	2010 - 2012	0.09	2010 - 2012	0.15
2014 - 2016	-0.13	2014 - 2016	-0.16	2014 - 2016	-0.16
2018 - 2020	-0.03	2018 - 2020	-0.02	2018 - 2020	-0.00
*p< 0.05					

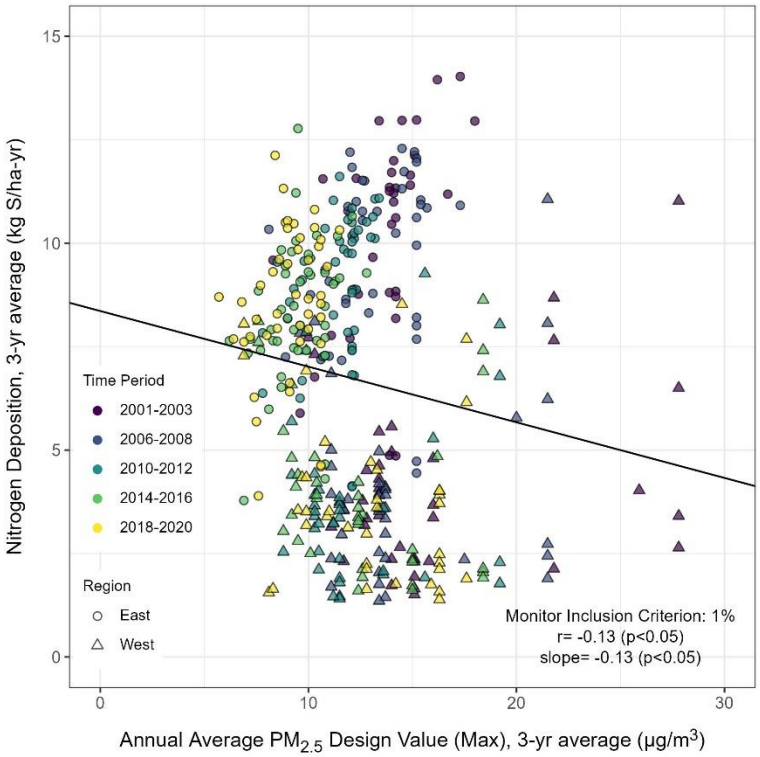

Figure 6A-97. Annual $PM_{2.5}$ EAQM-max values and TDep N deposition in 84 ecoregions (120-hr trajectories, NAM-12, 1% monitor inclusion criteria).

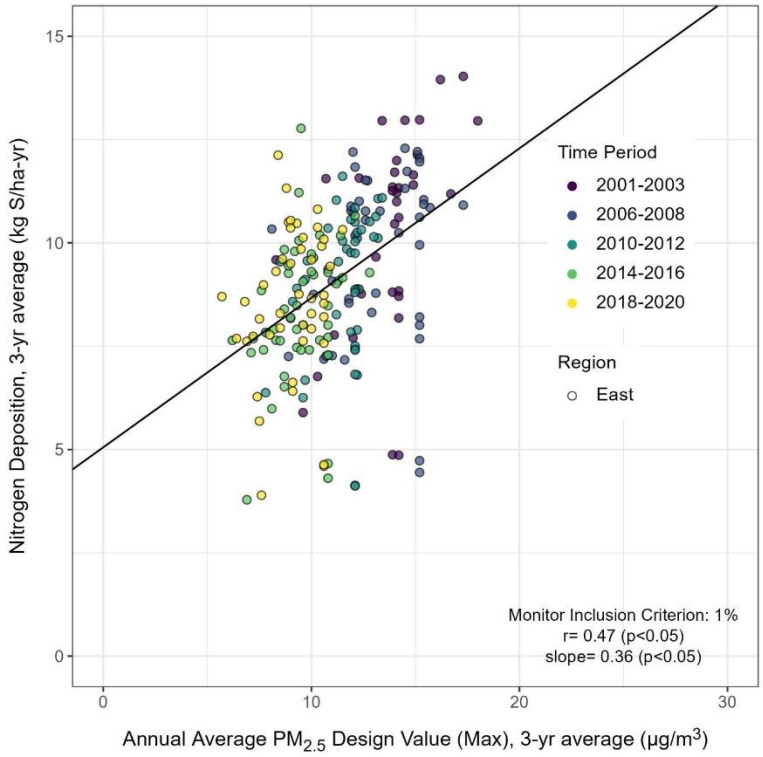

Figure 6A-98. Annual $PM_{2.5}$ EAQM-max values and TDep N deposition in eastern ecoregions (120-hr trajectories, NAM-12, 1% monitor inclusion criteria).

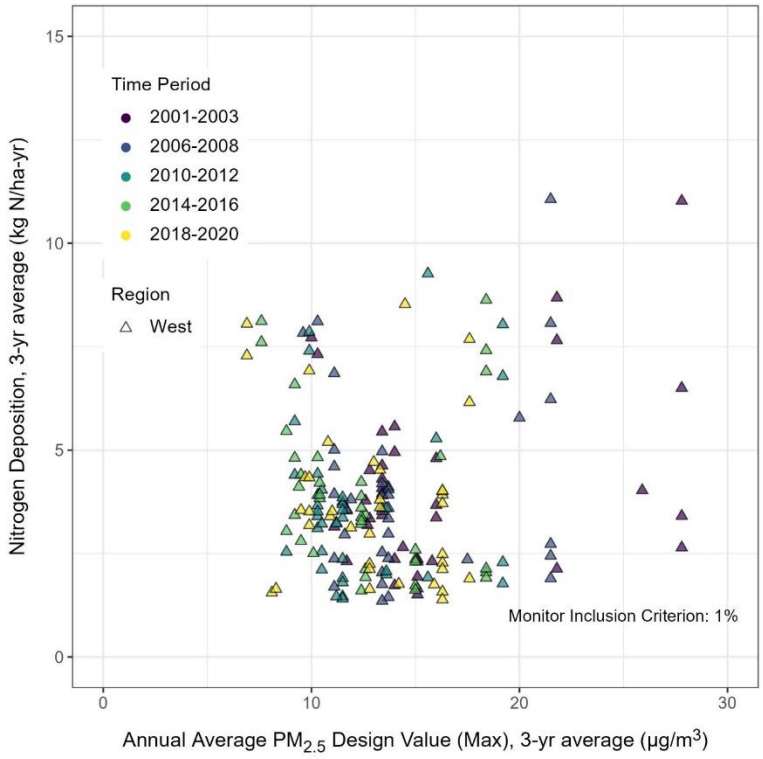

Figure 6A-99. Annual $PM_{2.5}$ EAQM-max values and TDep N deposition in western ecoregions (120-hr trajectories, NAM-12, 1% monitor inclusion criteria).

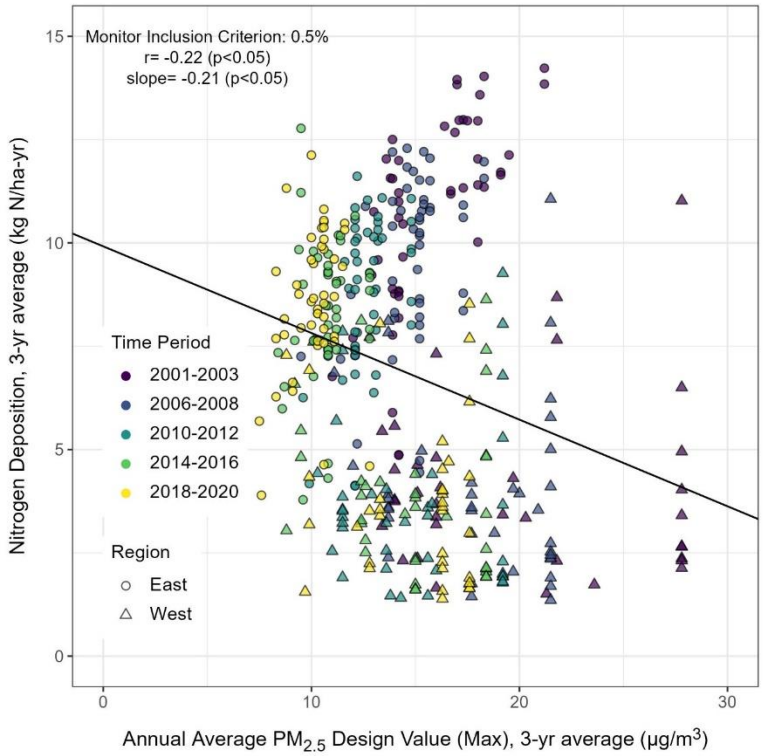

Figure 6A-100. Annual $PM_{2.5}$ EAQM-max values and TDep N deposition in 84 ecoregions (120-hr trajectories, NAM-12, 0.5% monitor inclusion criteria).

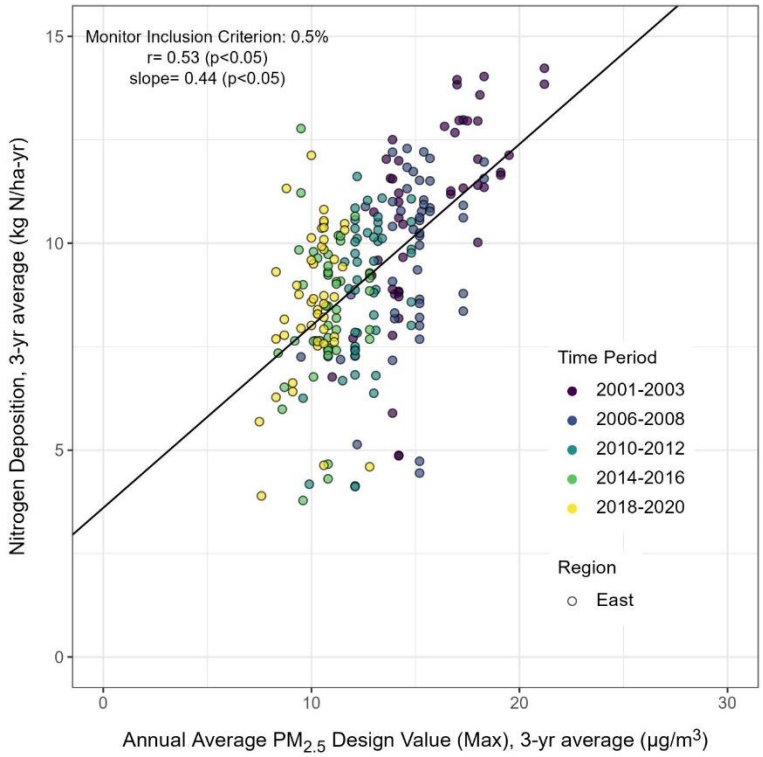

Figure 6A-101. Annual $PM_{2.5}$ EAQM-max values and TDep N deposition in eastern ecoregions (120-hr trajectories, NAM-12, 0.5% monitor inclusion criteria).

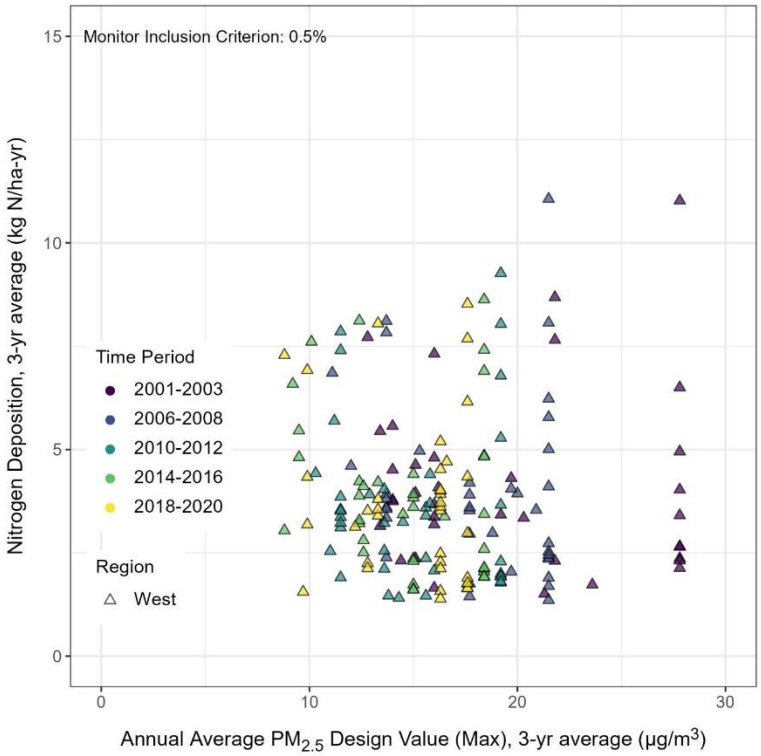

Figure 6A-102. Annual $PM_{2.5}$ EAQM-max values and TDep N deposition in western ecoregions (120-hr trajectories, NAM-12, 0.5% monitor inclusion criteria).

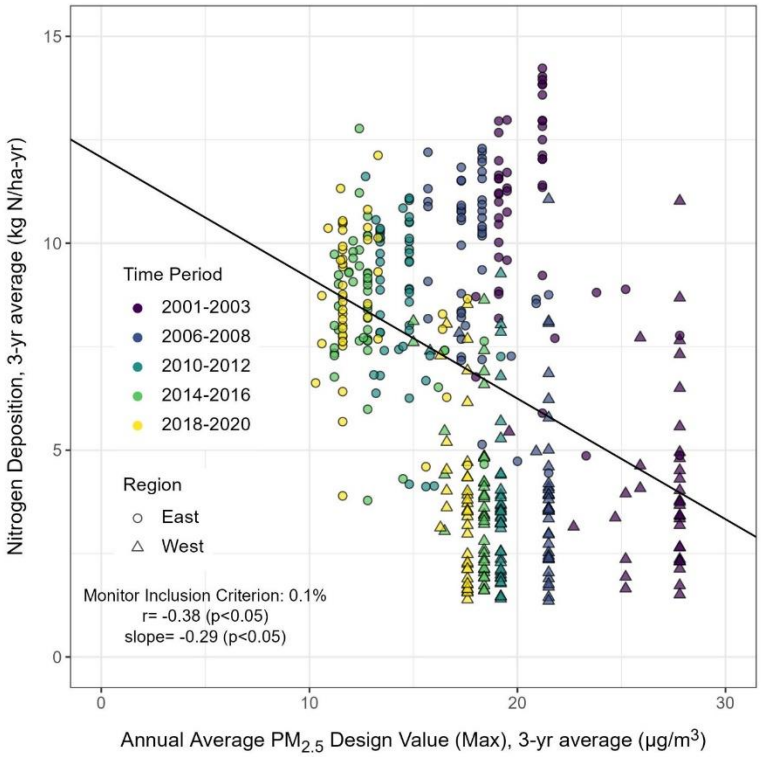

Figure 6A-103. Annual $PM_{2.5}$ EAQM-max values and TDep N deposition in 84 ecoregions (120-hr trajectories, NAM-12, 0.1% monitor inclusion criteria).

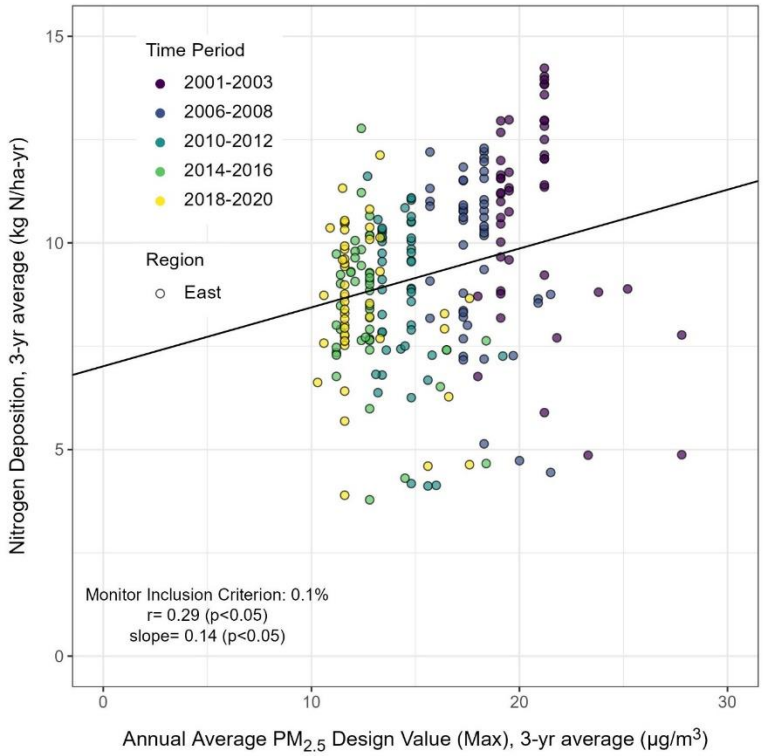

Figure 6A-104. Annual $PM_{2.5}$ EAQM-max values and TDep N deposition in eastern ecoregions (120-hr trajectories, NAM-12, 0.1% monitor inclusion criteria).

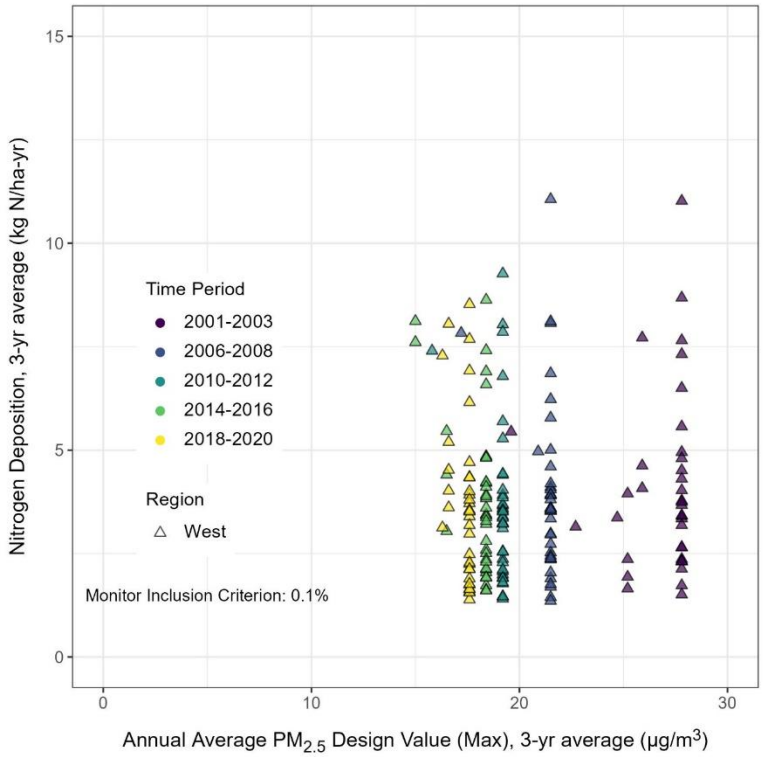

Figure 6A-105. Annual $PM_{2.5}$ EAQM-max values and TDep N deposition in western ecoregions (120-hr trajectories, NAM-12, 0.1% monitor inclusion criteria).

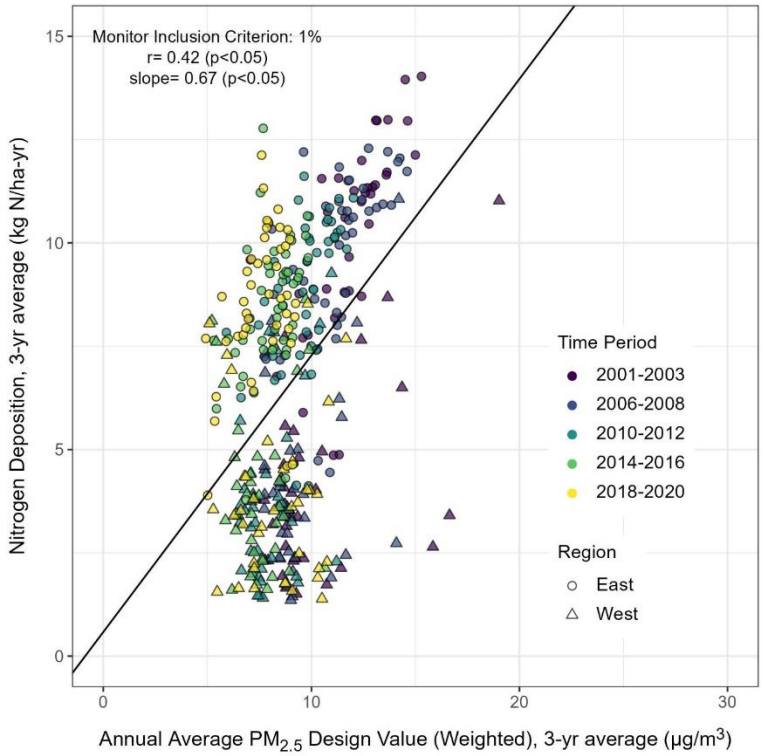

Figure 6A-106. Annual $PM_{2.5}$ EAQM-weighted values and TDep N deposition in 84 ecoregions (120-hr trajectories, NAM-12, 1% monitor inclusion criteria).

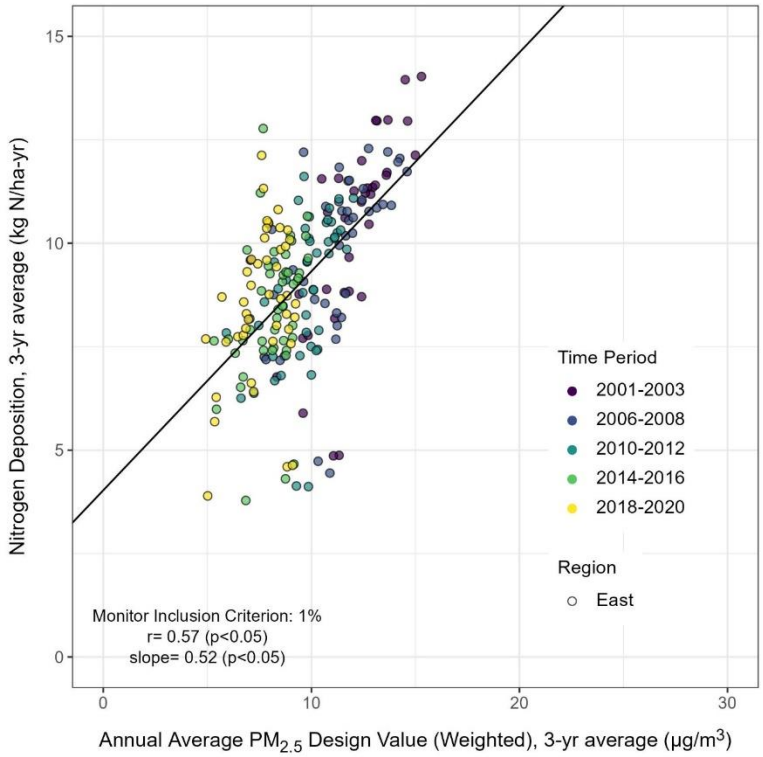

Figure 6A-107. Annual $PM_{2.5}$ EAQM-weighted values and TDep N deposition in eastern ecoregions (120-hr trajectories, NAM-12, 1% monitor inclusion criteria).

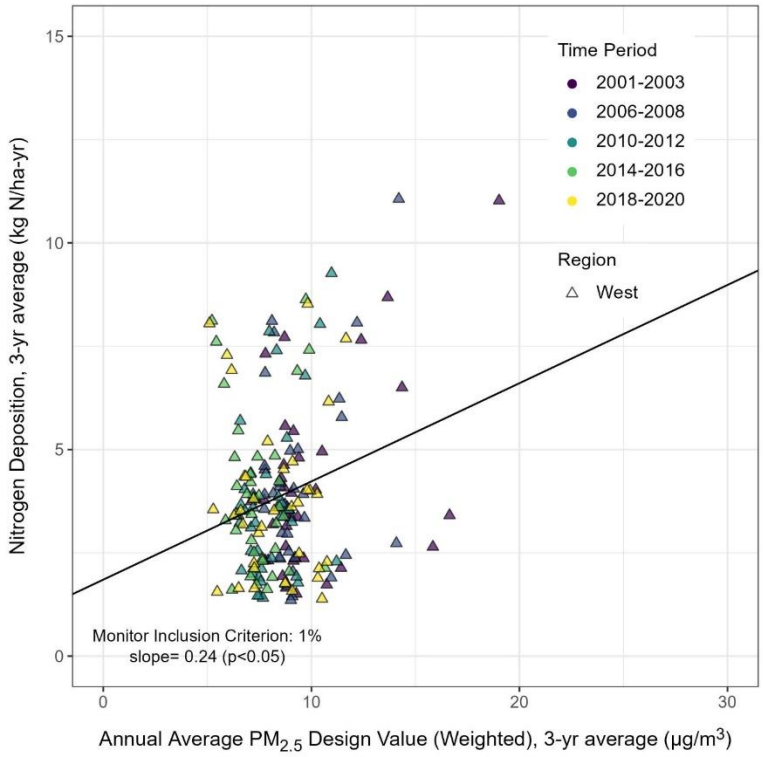

Figure 6A-108. Annual $PM_{2.5}$ EAQM-weighted values and TDep N deposition in west ecoregions (120-hr trajectories, NAM-12, 1% monitor inclusion criteria).

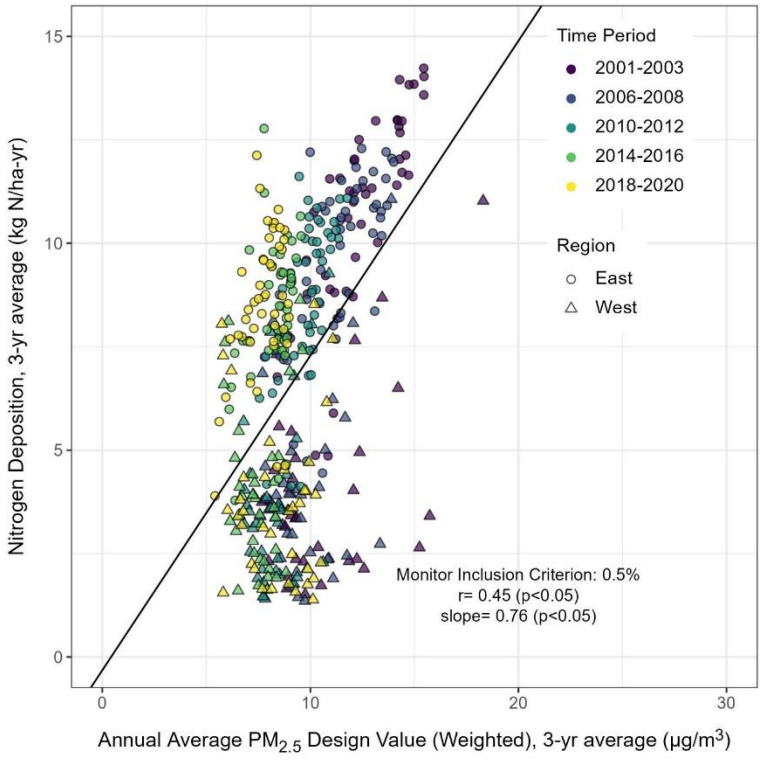

Figure 6A-109. Annual $PM_{2.5}$ EAQM-weighted values and TDep N deposition in 84 ecoregions (120-hr trajectories, NAM-12, 0.5% monitor inclusion criteria).

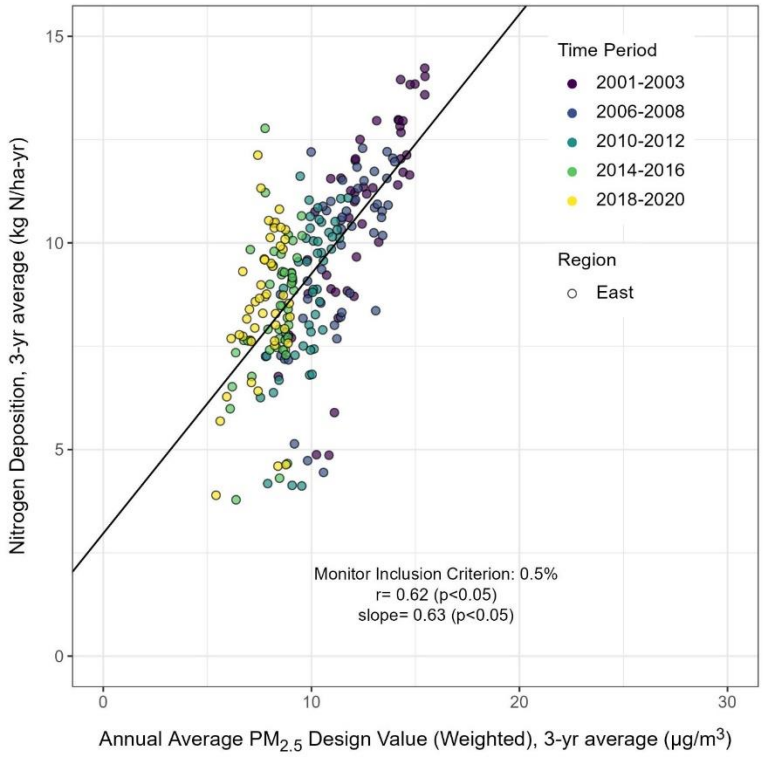

Figure 6A-110. Annual $PM_{2.5}$ EAQM-weighted values and TDep N deposition in eastern ecoregions (120-hr trajectories, NAM-12, 0.5% monitor inclusion criteria).

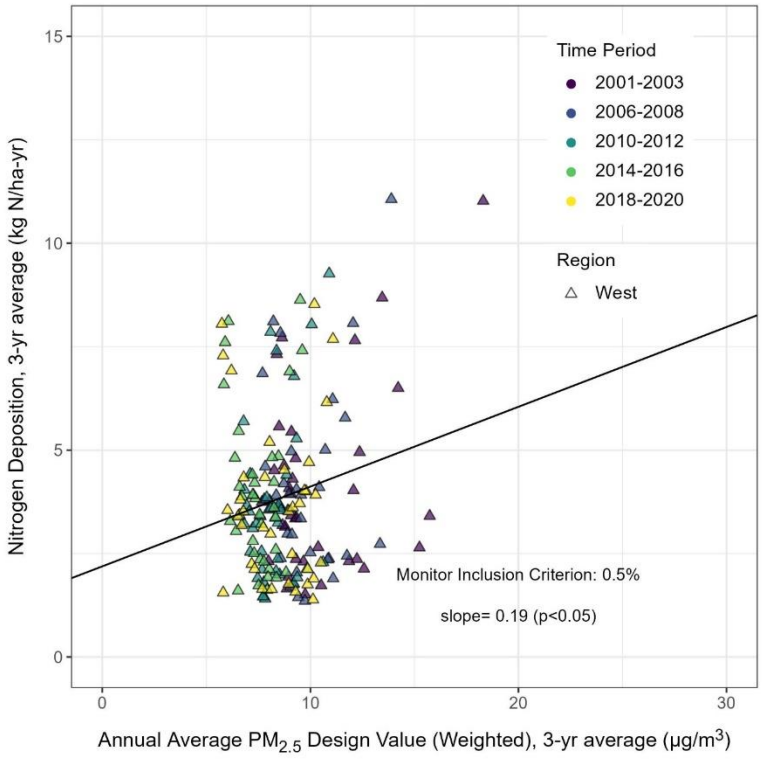

Figure 6A-111. Annual $PM_{2.5}$ EAQM-weighted values and TDep N deposition in western ecoregions (120-hr trajectories, NAM-12, 0.5% monitor inclusion criteria).

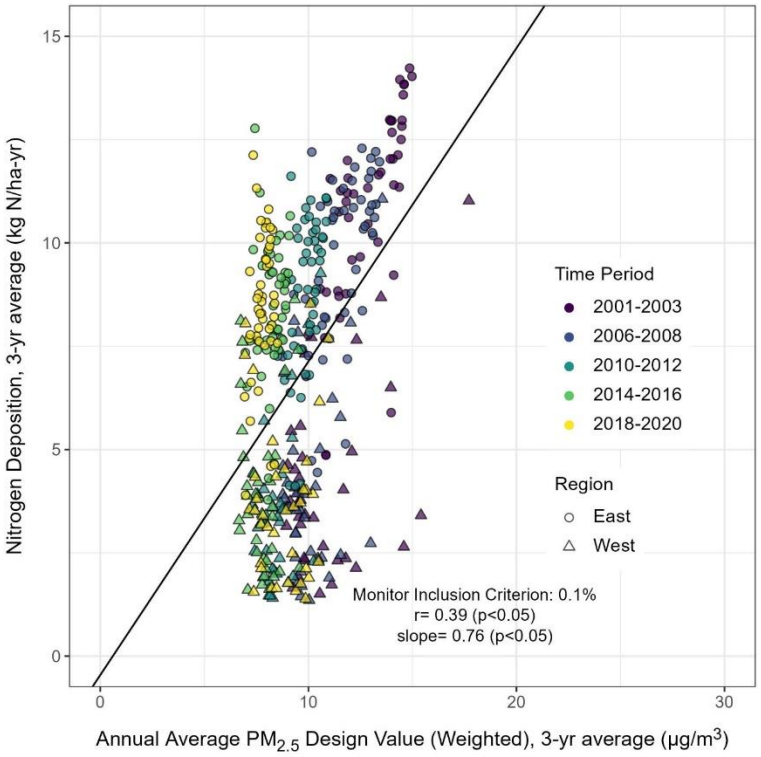

Figure 6A-112. Annual $PM_{2.5}$ EAQM-weighted values and TDep N deposition in 84 ecoregions (120-hr trajectories, NAM-12, 0.1% monitor inclusion criteria).

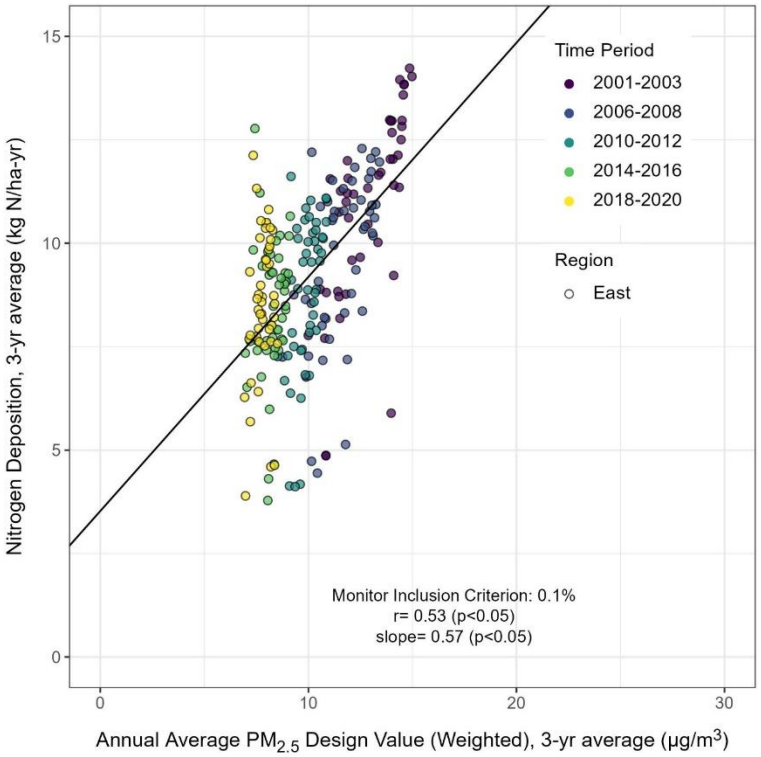

Figure 6A-113. Annual $PM_{2.5}$ EAQM-weighted values and TDep N deposition in eastern ecoregions (120-hr trajectories, NAM-12, 0.1% monitor inclusion criteria).

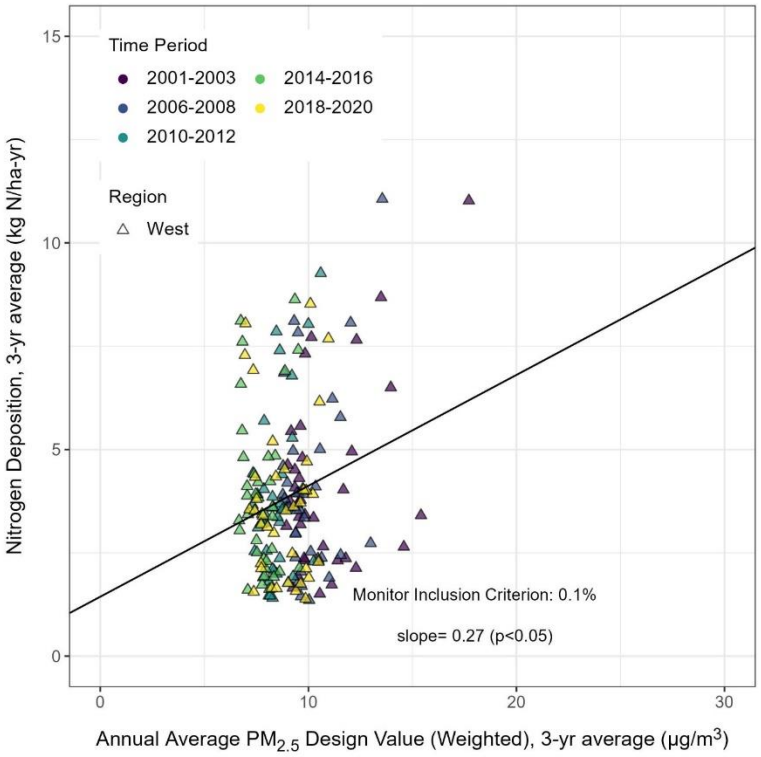

Figure 6A-114. Annual $PM_{2.5}$ EAQM-weighted values and TDep N deposition in western ecoregions (120-hr trajectories, NAM-12, 0.1% monitor inclusion criteria).

6A.6.7.2. Sulfur

Table 6A-9. Correlation coefficients of Tdep estimates of sulfur deposition and annual $PM_{2.5}$ EAQMs generated by HYSPLIT analysis, 120-hr trajectories. Data are also split by year and by region (East/West).

Sulfur Deposition and $PM_{2.5}$					
Annual Max-All Ecoregions- Monitor Inclusion Criteria: 1%	Correlation Coefficient (r) = -0.14*	*Annual Max-All Ecoregions- Monitor Inclusion Criteria: 0.5%*	Correlation Coefficient (r) = -0.22*	*Annual Max-All Ecoregions- Monitor Inclusion Criteria: 0.1%*	Correlation Coefficient (r) = -0.31*
Year	r	Year	r	Year	r
2001 - 2003	-0.13	2001 - 2003	-0.21	2001 - 2003	-0.73*
2006 - 2008	-0.05	2006 - 2008	-0.37*	2006 - 2008	-0.73*
2010 - 2012	0.07	2010 - 2012	-0.22*	2010 - 2012	-0.78*
2014 - 2016	-0.44*	2014 - 2016	-0.53*	2014 - 2016	-0.84*
2018 - 2020	-0.40*	2018 - 2020	-0.54*	2018 - 2020	-0.80*
Annual Max-East Ecoregions- Monitor Inclusion Criteria: 1%	r = 0.74*	*Annual Max-East Ecoregions- Monitor Inclusion Criteria: 0.5%*	r = 0.83*	*Annual Max-East Ecoregions- Monitor Inclusion Criteria: 0.1%*	r = 0.58*
Year	r	Year	r	Year	r
2001 - 2003	0.69*	2001 - 2003	0.73*	2001 - 2003	0.02
2006 - 2008	0.39*	2006 - 2008	0.53*	2006 - 2008	0.10
2010 - 2012	0.47*	2010 - 2012	0.70*	2010 - 2012	-0.04
2014 - 2016	0.41*	2014 - 2016	0.43*	2014 - 2016	-0.47*

2018 - 2020	0.61*	2018 - 2020	0.53*	2018 - 2020	-0.30*
Annual Max-West Ecoregions- Monitor Inclusion Criteria: 1%	r = -0.36*	*Annual Max-West Ecoregions- Monitor Inclusion Criteria: 0.5%*	r = -0.33*	*Annual Max-West Ecoregions- Monitor Inclusion Criteria: 0.1%*	r = 0.19*
Year	r	Year	r	Year	r
2001 - 2003	-0.45*	2001 - 2003	-0.48*	2001 - 2003	-0.17
2006 - 2008	-0.47*	2006 - 2008	-0.61*	2006 - 2008	-0.31
2010 - 2012	-0.64*	2010 - 2012	-0.56*	2010 - 2012	-0.23
2014 - 2016	-0.73*	2014 - 2016	-0.53*	2014 - 2016	-0.48*
2018 - 2020	-0.20	2018 - 2020	-0.37*	2018 - 2020	-0.39*
Weighted Annual Average-All Ecoregions- Monitor Inclusion Criteria: 1%	r = 0.43*	*Weighted Annual Average-All Ecoregions- Monitor Inclusion Criteria: 0.5%*	r = 0.48*	*Weighted Annual Average-All Ecoregions- Monitor Inclusion Criteria: 0.1%*	r = 0.46*
Year	r	Year	r	Year	r
2001 - 2003	0.51*	2001 - 2003	0.55*	2001 - 2003	0.62*
2006 - 2008	0.46*	2006 - 2008	0.56*	2006 - 2008	0.64*
2010 - 2012	0.60*	2010 - 2012	0.70*	2010 - 2012	0.76*
2014 - 2016	0.33*	2014 - 2016	0.43*	2014 - 2016	0.38*
2018 - 2020	-0.00	2018 - 2020	-0.07	2018 - 2020	-0.22*
Weighted Annual Average-East Ecoregions- Monitor Inclusion Criteria: 1%	r = 0.80*	*Weighted Annual Average-East Ecoregions- Monitor Inclusion Criteria: 0.5%*	r = 0.90*	*Weighted Annual Average-East Ecoregions- Monitor Inclusion Criteria: 0.1%*	r = 0.89*
Year	r	Year	r	Year	r
2001 - 2003	0.83*	2001 - 2003	0.88*	2001 - 2003	0.88*
2006 - 2008	0.73*	2006 - 2008	0.86*	2006 - 2008	0.93*
2010 - 2012	0.64*	2010 - 2012	0.84*	2010 - 2012	0.89*
2014 - 2016	0.52*	2014 - 2016	0.65*	2014 - 2016	0.53*
2018 - 2020	0.63*	2018 - 2020	0.69*	2018 - 2020	0.72*
Weighted Annual Average-West Ecoregions- Monitor Inclusion Criteria: 1%	r = -0.19*	*Weighted Annual Average-West Ecoregions- Monitor Inclusion Criteria: 0.5%*	r = -0.22*	*Weighted Annual Average-West Ecoregions- Monitor Inclusion Criteria: 0.1%*	r = -0.13
Year	r	Year	r	Year	r
2001 - 2003	-0.30	2001 - 2003	-0.44*	2001 - 2003	-0.37*
2006 - 2008	-0.52*	2006 - 2008	-0.62*	2006 - 2008	-0.60*
2010 - 2012	-0.30	2010 - 2012	-0.40*	2010 - 2012	-0.35*
2014 - 2016	-0.48*	2014 - 2016	-0.55*	2014 - 2016	-0.58*
2018 - 2020	-0.20	2018 - 2020	-0.25	2018 - 2020	-0.24
*p< 0.05					

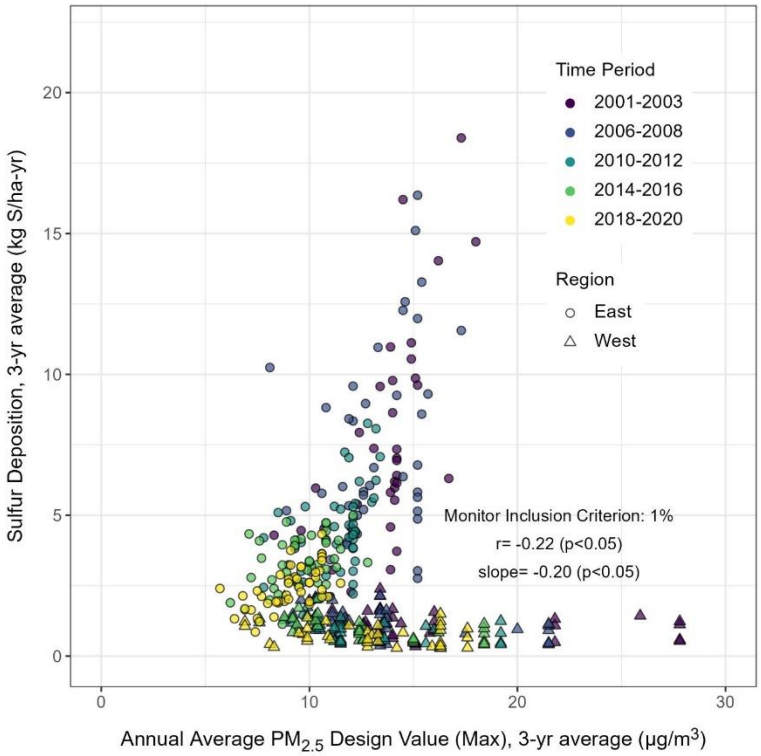

Figure 6A-115. Annual $PM_{2.5}$ EAQM-max values and TDep S deposition in 84 ecoregions (120-hr trajectories, NAM-12, 1% monitor inclusion criteria).

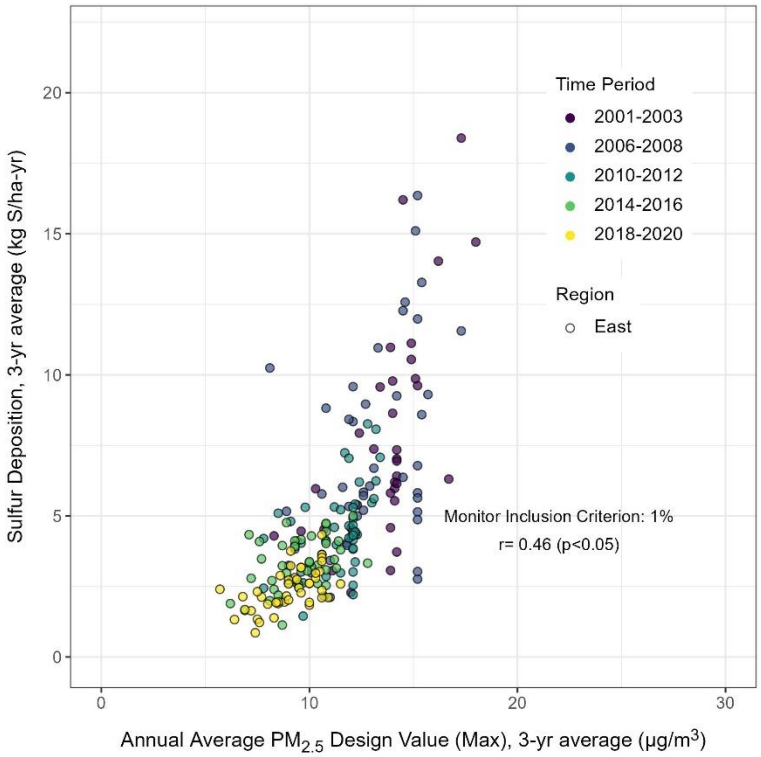

Figure 6A-116. Annual $PM_{2.5}$ EAQM-max values and TDep S deposition in eastern ecoregions (120-hr trajectories, NAM-12, 1% monitor inclusion criteria).

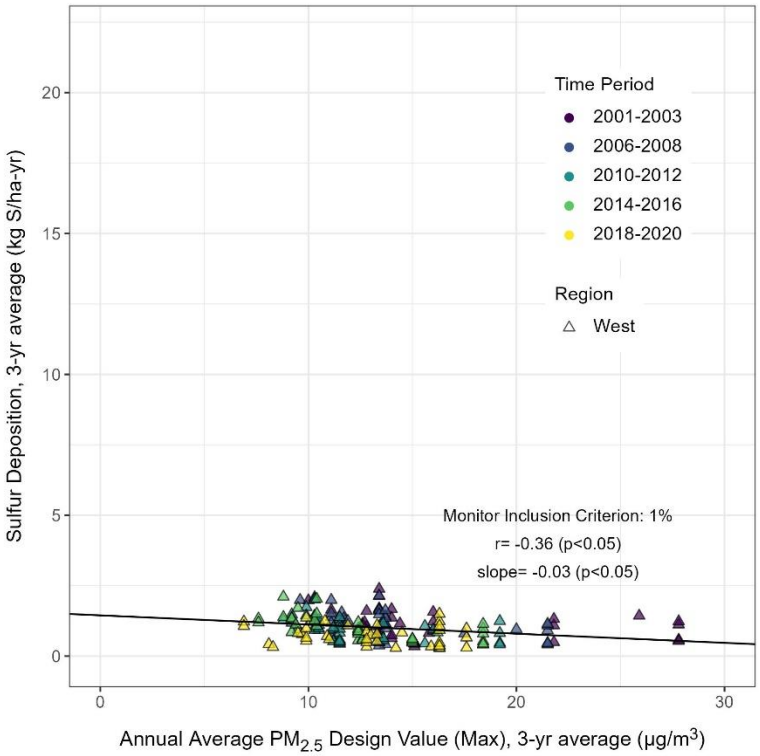

Figure 6A-117. Annual $PM_{2.5}$ EAQM-max values and TDep S deposition in western ecoregions (120-hr trajectories, NAM-12, 1% monitor inclusion criteria)

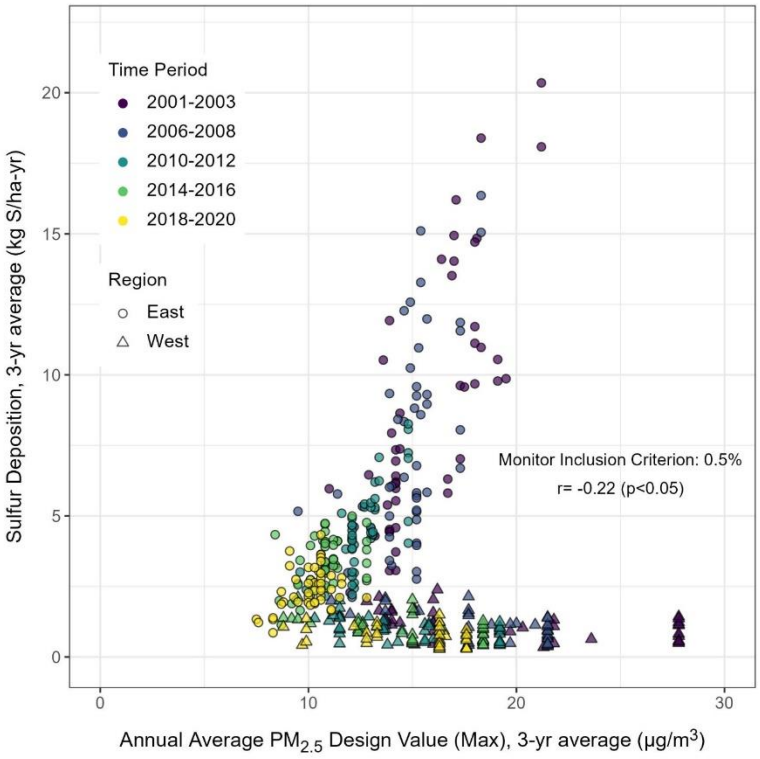

Figure 6A-118. Annual $PM_{2.5}$ EAQM-max values and TDep S deposition in 84 ecoregions (120-hr trajectories, NAM-12, 0.5% monitor inclusion criteria).

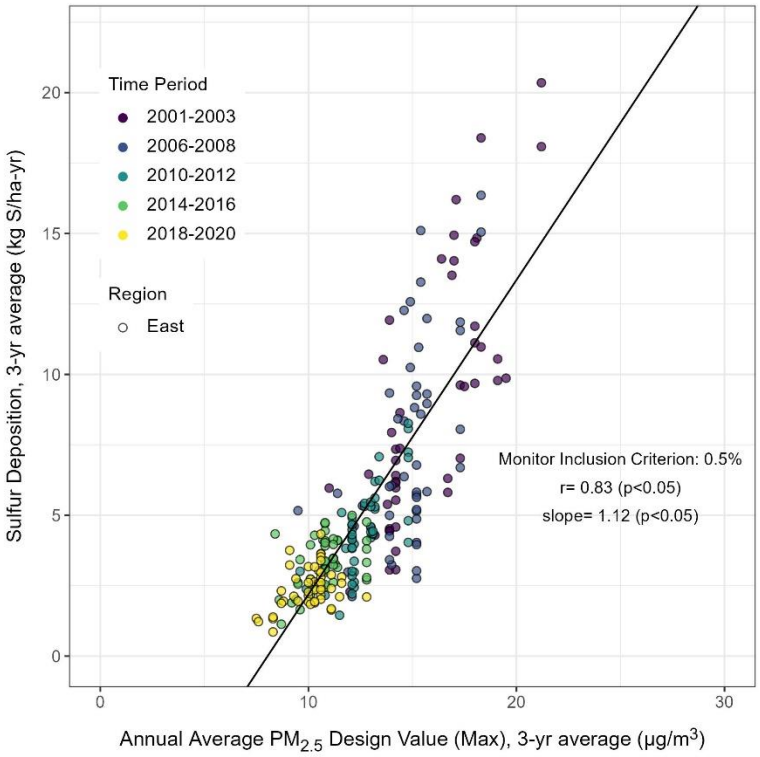

Figure 6A-119. Annual $PM_{2.5}$ EAQM-max values and TDep S deposition in eastern ecoregions (120-hr trajectories, NAM-12, 0.5% monitor inclusion criteria).

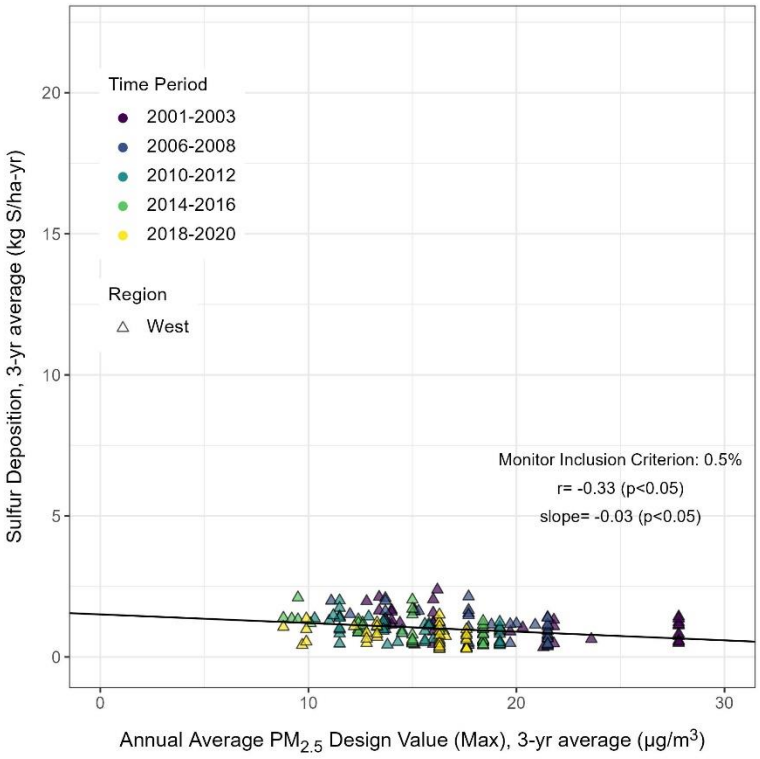

Figure 6A-120. Annual $PM_{2.5}$ EAQM-max values and TDep S deposition in western ecoregions (120-hr trajectories, NAM-12, 0.5% monitor inclusion criteria).

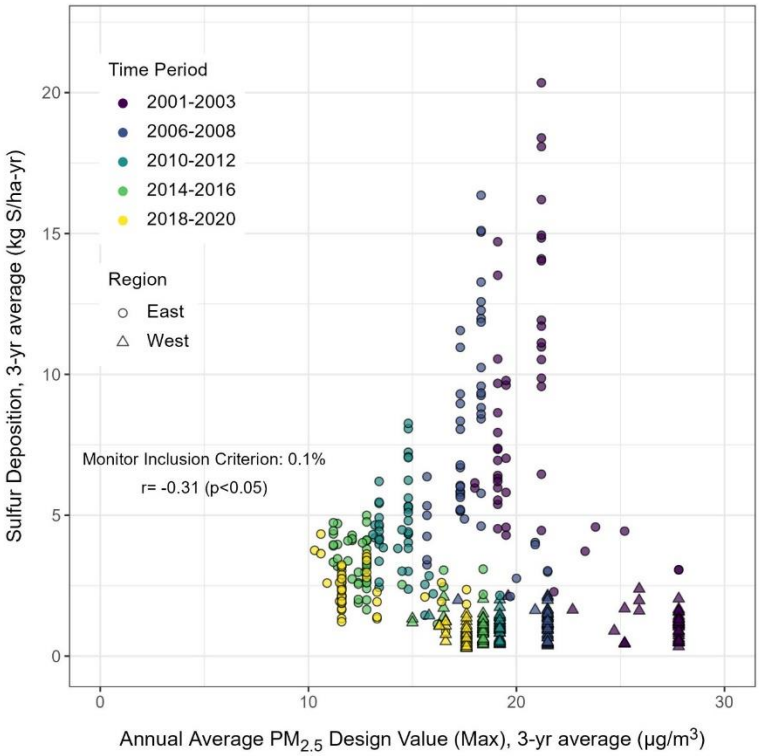

Figure 6A-121. Annual $PM_{2.5}$ EAQM-max values and Tdep S deposition in 84 ecoregions (120-hr trajectories, NAM-12, 0.1% monitor inclusion criteria).

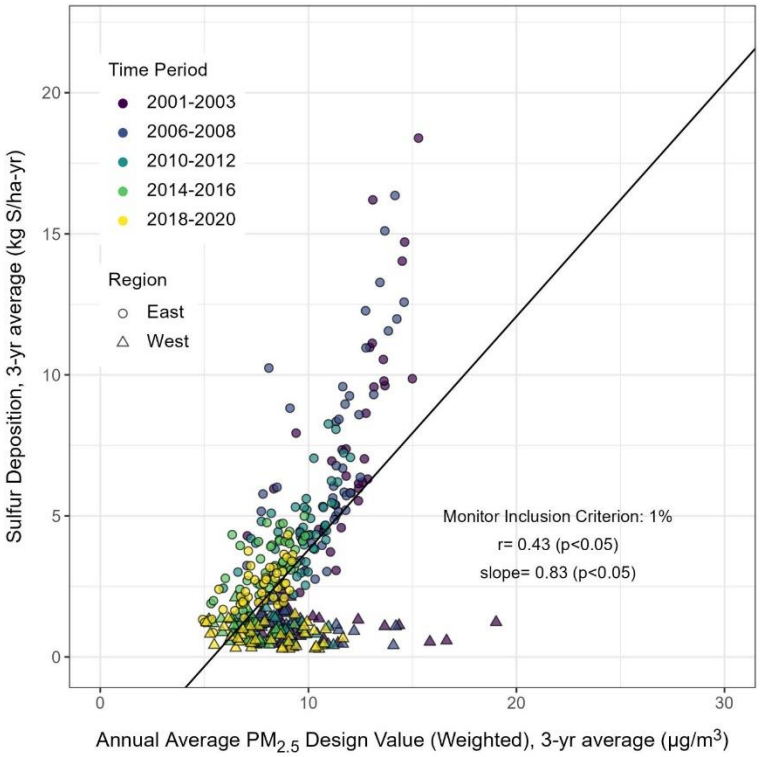

Figure 6A-122. Annual $PM_{2.5}$ EAQM-weighted values and TDep S deposition in 84 ecoregions (120-hr trajectories, NAM-12, 1% monitor inclusion criteria).

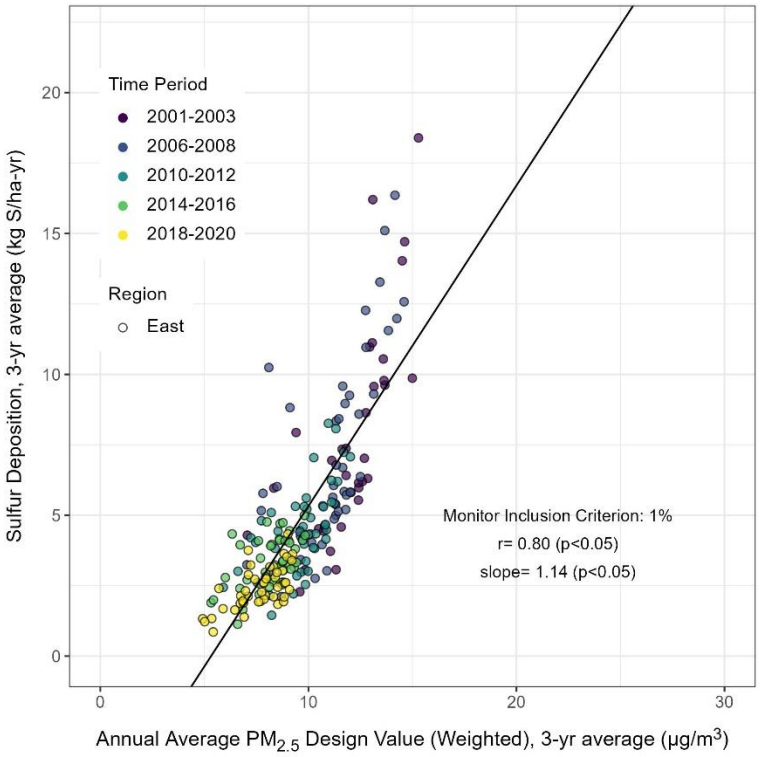

Figure 6A-123. Annual $PM_{2.5}$ EAQM-weighted values and TDep S deposition in eastern ecoregions (120-hr trajectories, NAM-12, 1% monitor inclusion criteria).

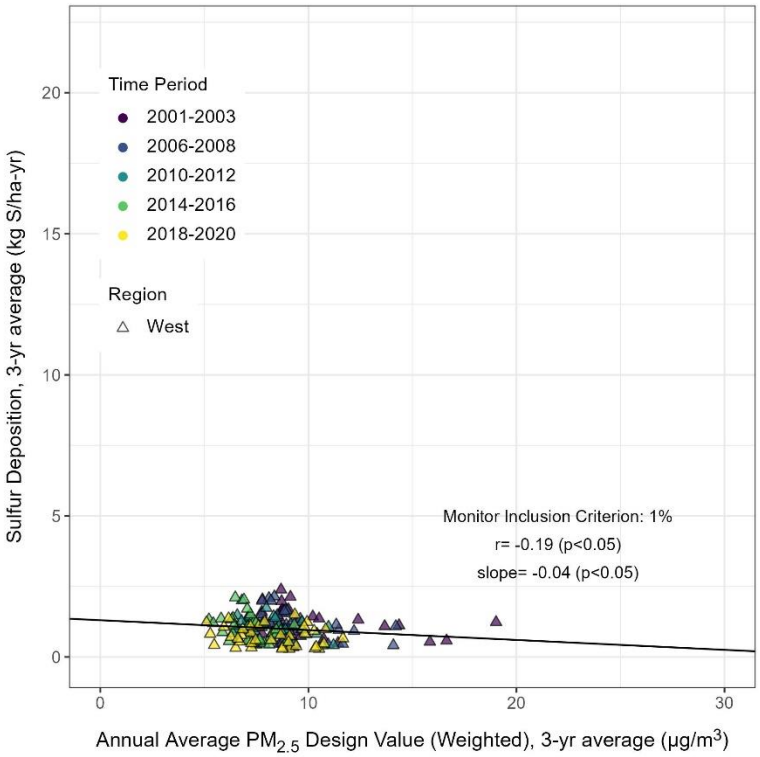

Figure 6A-124. Annual $PM_{2.5}$ EAQM-weighted values and TDep S deposition in western ecoregions (120-hr trajectories, NAM-12, 1% monitor inclusion criteria).

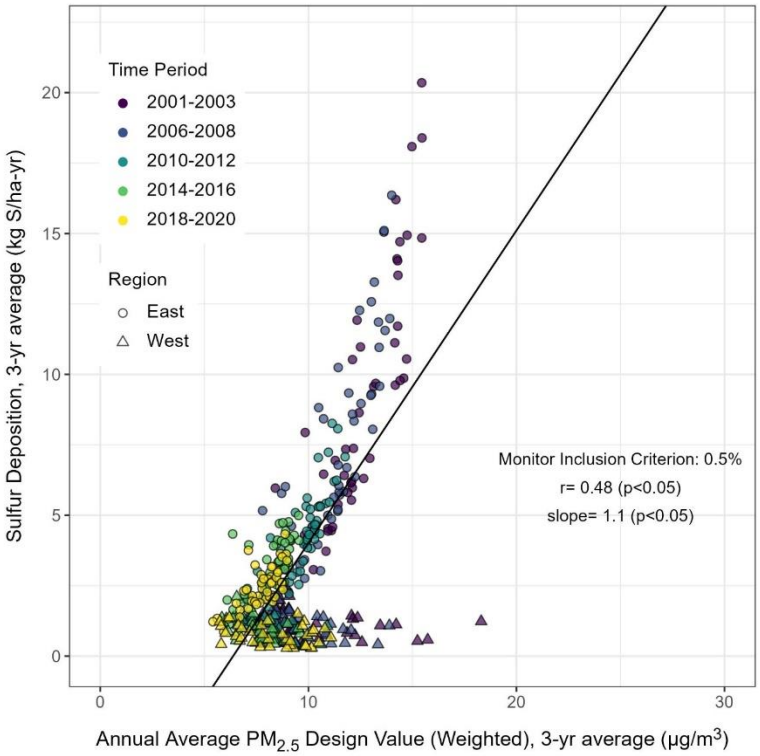

Figure 6A-125. Annual $PM_{2.5}$ EAQM-weighted values and TDep S deposition in 84 ecoregions (120-hr trajectories, NAM-12, 0.5% monitor inclusion criteria).

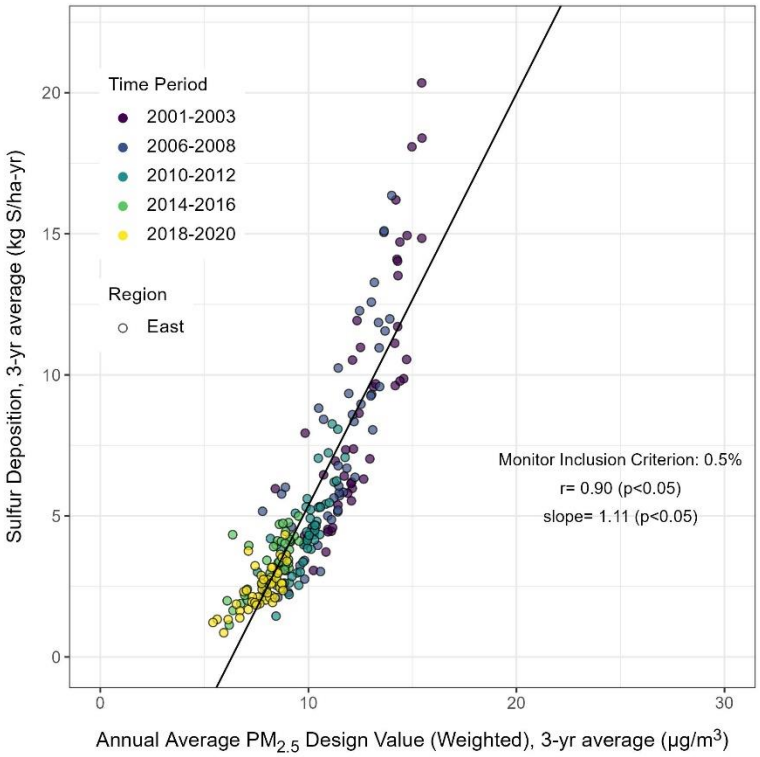

Figure 6A-126. Annual $PM_{2.5}$ EAQM-weighted values and TDep S deposition in eastern ecoregions (120-hr trajectories, NAM-12, 0.5% monitor inclusion criteria).

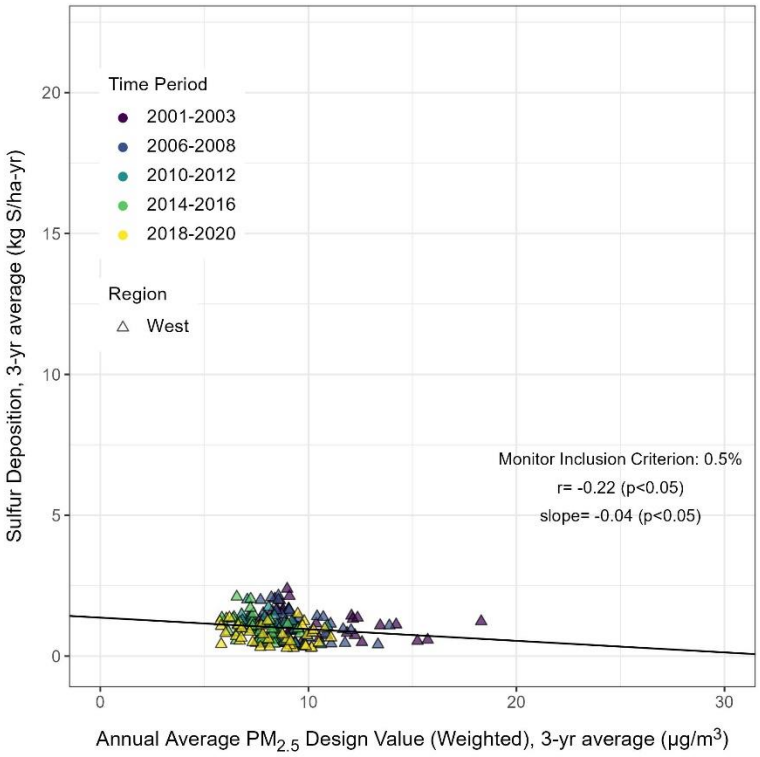

Figure 6A-127. Annual $PM_{2.5}$ EAQM-weighted values and TDep S deposition in western ecoregions (120-hr trajectories, NAM-12, 0.5% monitor inclusion criteria).

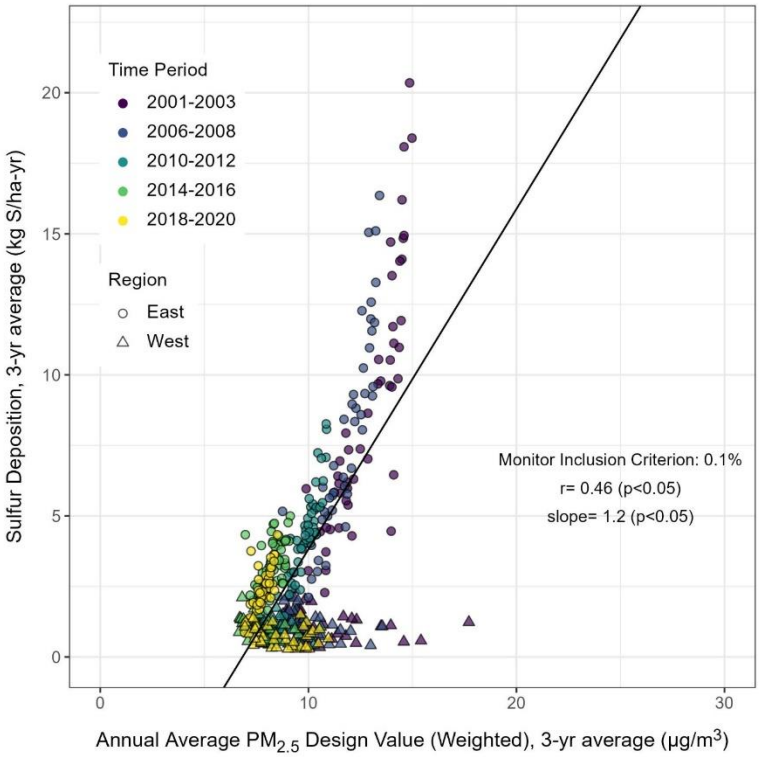

Figure 6A-128. Annual $PM_{2.5}$ EAQM-weighted values and TDep S deposition in 84 ecoregions (120-hr trajectories, NAM-12, 0.1% monitor inclusion criteria).

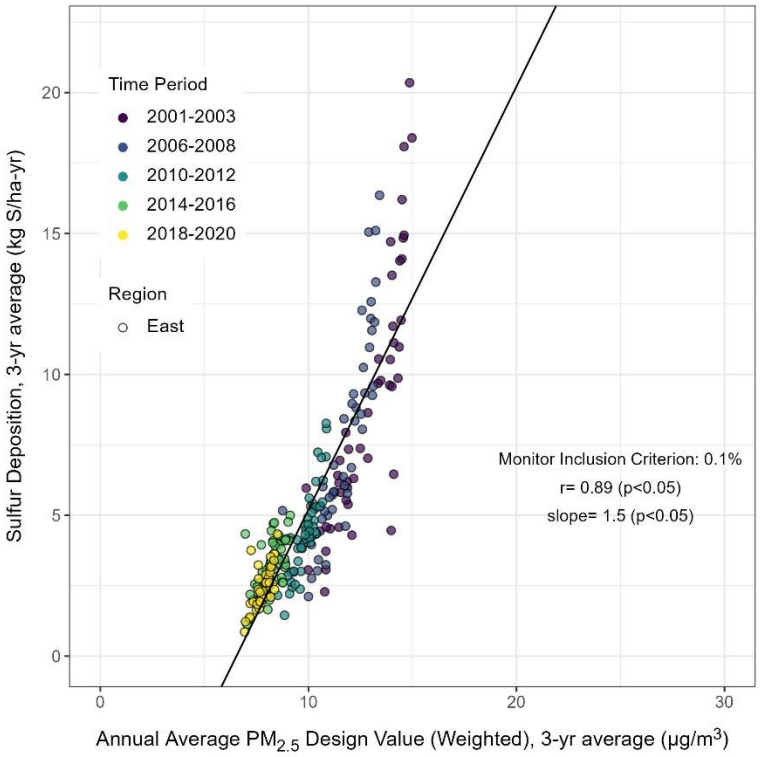

Figure 6A-129. Annual $PM_{2.5}$ EAQM-weighted values and TDep S deposition in eastern ecoregions (120-hr trajectories, NAM-12, 0.1% monitor inclusion criteria).

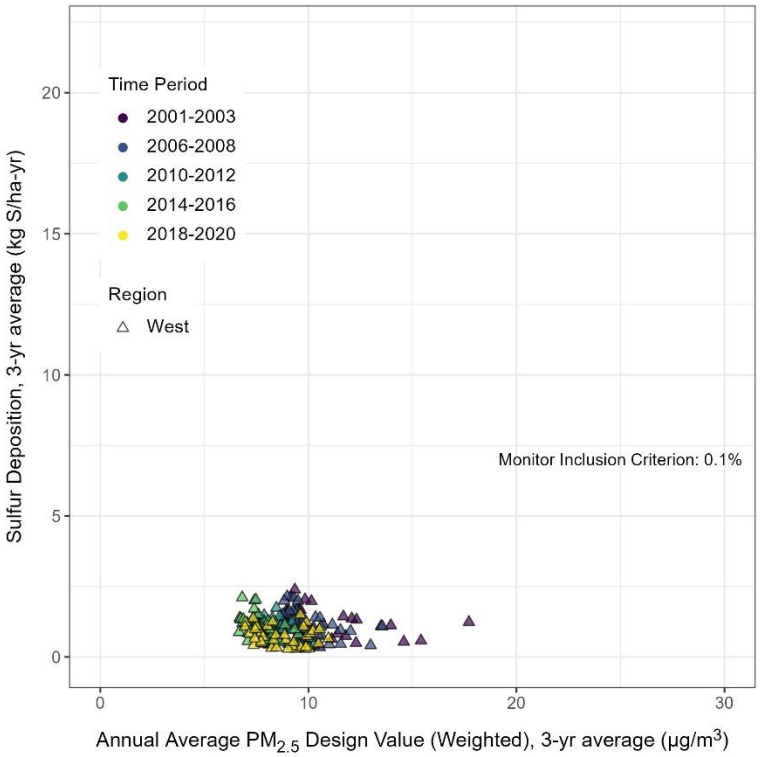

Figure 6A-130. Annual $PM_{2.5}$ EAQM-weighted values and TDep S deposition in western ecoregions (120-hr trajectories, NAM-12, 0.1% monitor inclusion criteria).

6A.6.8. $PM_{2.5}$ Annual Metric – 48-hr

6A.6.8.1. Nitrogen

Table 6A-10. Correlation coefficients of TDep estimates of nitrogen deposition and annual $PM_{2.5}$ EAQMs generated by HYSPLIT analysis, 48-hr trajectories. Data are also split by year and by region (East/West).

Nitrogen Deposition and $PM_{2.5}$			
Annual Max-All Ecoregions- Monitor Inclusion Criteria: 1%	Correlation Coefficient (r) = 0.05	*Weighted Annual Average-All Ecoregions- Monitor Inclusion Criteria: 1%*	Correlation Coefficient (r) = 0.50*
Year	r	Year	r
2001 - 2003	0.28*	2001 - 2003	0.69*
2006 - 2008	0.24*	2006 - 2008	0.64*
2010 - 2012	0.30*	2010 - 2012	0.71*
2014 - 2016	-0.18	2014 - 2016	0.44*
2018 - 2020	-0.39*	2018 - 2020	0.003
Annual Max-East Ecoregions- Monitor Inclusion Criteria: 1%	r = 0.51*	*Weighted Annual Average-East Ecoregions- Monitor Inclusion Criteria: 1%*	r = 0.61*
Year	r	Year	r
2001 - 2003	0.60*	2001 - 2003	0.81*
2006 - 2008	0.30*	2006 - 2008	0.63*
2010 - 2012	0.36*	2010 - 2012	0.57*
2014 - 2016	0.25	2014 - 2016	0.39*
2018 - 2020	0.38*	2018 - 2020	0.21
Annual Max-West Ecoregions- Monitor Inclusion Criteria: 1%	r = -0.08	*Weighted Annual Average-West Ecoregions- Monitor Inclusion Criteria: 1%*	r = 0.04
Year	r	Year	r
2001 - 2003	-0.07	2001 - 2003	0.02
2006 - 2008	-0.11	2006 - 2008	-0.08
2010 - 2012	-0.04	2010 - 2012	0.13
2014 - 2016	-0.23	2014 - 2016	-0.11
2018 - 2020	-0.12	2018 - 2020	-0.007
*p< 0.05			

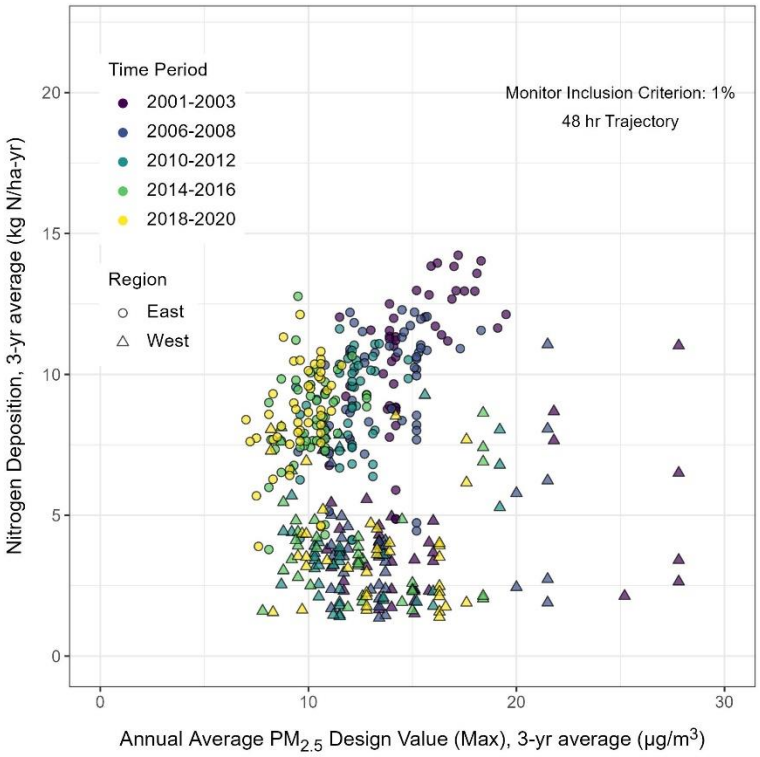

Figure 6A-131. Annual $PM_{2.5}$ EAQM-max values and TDep N deposition in 84 ecoregions (48-hr trajectories, NARR-32, 1% monitor inclusion criteria).

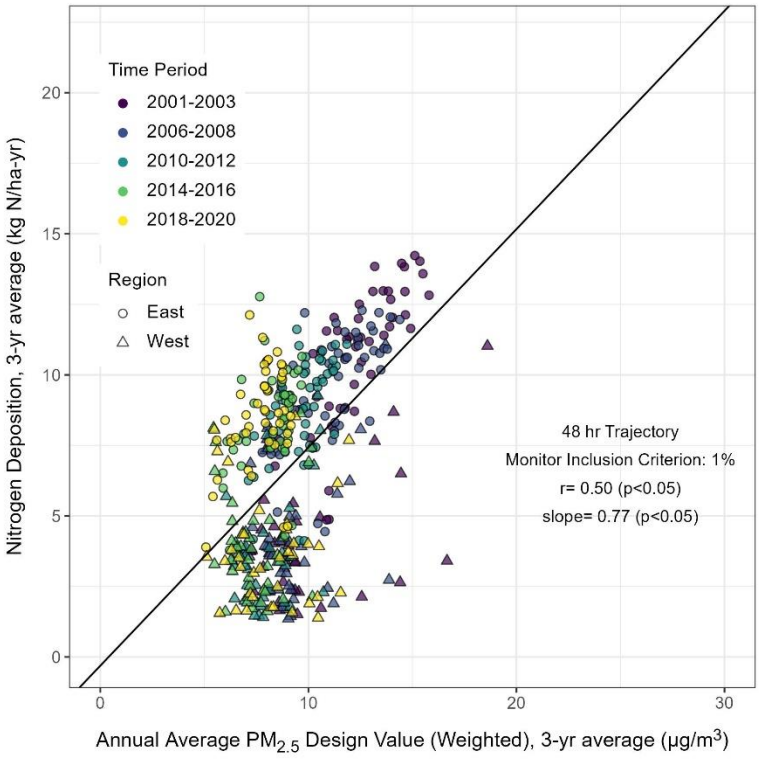

Figure 6A-132. Annual $PM_{2.5}$ EAQM-weighted values and TDep N deposition in 84 ecoregions (48-hr trajectories, NARR-32, 1% monitor inclusion criteria).

6A.6.8.2. Sulfur

Table 6A-11. Correlation coefficients of TDep estimates of sulfur deposition and annual $PM_{2.5}$ EAQMs generated by HYSPLIT analysis, 48-hr trajectories. Data are also split by year and by region (East/West).

Sulfur Deposition and $PM_{2.5}$			
Annual Max-All Ecoregions- Monitor Inclusion Criteria: 1%	Correlation Coefficient (r) = 0.05	*Weighted Annual Average-All Ecoregions- Monitor Inclusion Criteria: 1%*	Correlation Coefficient (r) = 0.51*
Year	r	Year	r
2001 - 2003	0.16	2001 - 2003	0.59*
2006 - 2008	0.15	2006 - 2008	0.53*
2010 - 2012	0.23*	2010 - 2012	0.64*
2014 - 2016	-0.24*	2014 - 2016	0.43*
2018 - 2020	-0.40*	2018 - 2020	0.05
Annual Max-East Ecoregions- Monitor Inclusion Criteria: 1%	r = 0.80*	*Weighted Annual Average-East Ecoregions- Monitor Inclusion Criteria: 1%*	r = 0.85*
Year	r	Year	r
2001 - 2003	0.62*	2001 - 2003	0.83*
2006 - 2008	0.42*	2006 - 2008	0.75*
2010 - 2012	0.61*	2010 - 2012	0.67*
2014 - 2016	0.48*	2014 - 2016	0.62*
2018 - 2020	0.52*	2018 - 2020	0.61*
Annual Max-West Ecoregions- Monitor Inclusion Criteria: 1%	r = -0.37*	*Weighted Annual Average-West Ecoregions- Monitor Inclusion Criteria: 1%*	r = -0.21*
Year	r	Year	r
2001 - 2003	-0.50*	2001 - 2003	-0.38*
2006 - 2008	-0.46*	2006 - 2008	-0.53*
2010 - 2012	-0.50*	2010 - 2012	-0.28*
2014 - 2016	-0.61*	2014 - 2016	-0.47*
2018 - 2020	-0.26	2018 - 2020	-0.19
*p< 0.05			

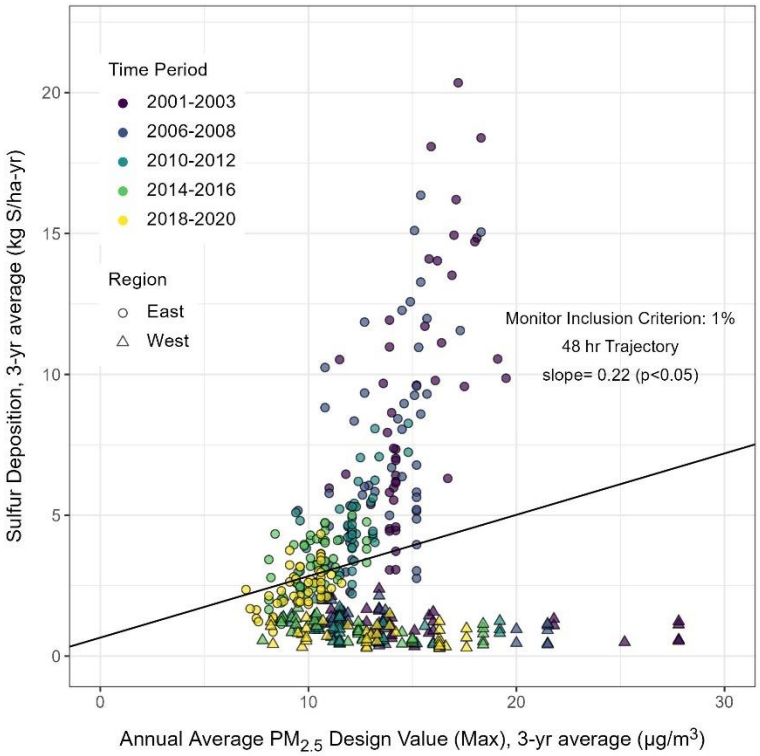

Figure 6A-133. Annual $PM_{2.5}$ EAQM-max values and Tdep S deposition in 84 ecoregions (48-hr trajectories, NARR-32, 1% monitor inclusion criteria).

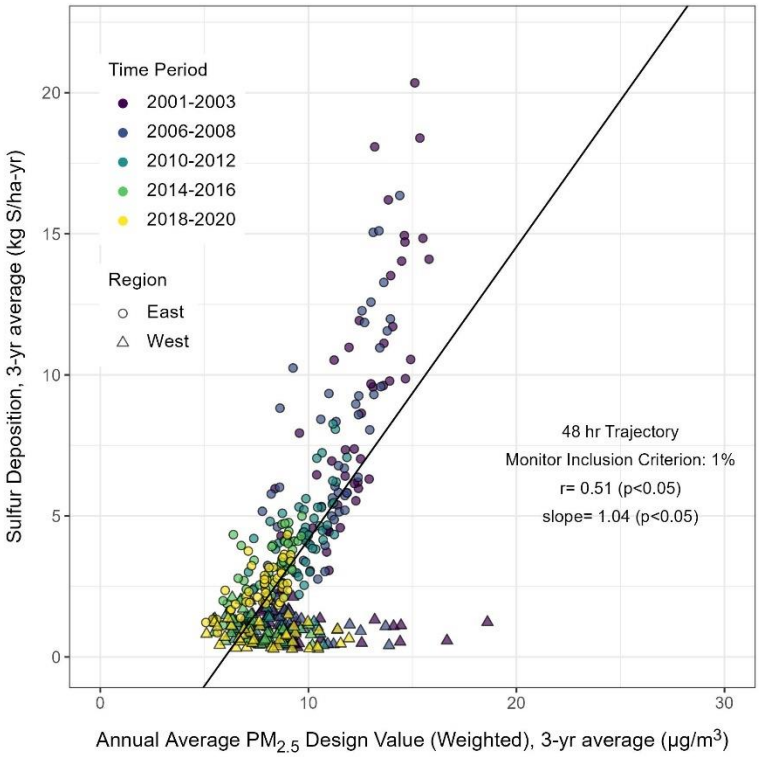

Figure 6A-134. Annual $PM_{2.5}$ EAQM-weighted values and TDep S deposition in 84 ecoregions (48-hr trajectories, NARR-32, 1% monitor inclusion criteria).

Attachment

Attachment

Maps Showing Monitor Sites of Influence for NO_2, $PM_{2.5}$, and SO_2 (3-hour metric) Based on Different Inclusion Criteria for 16 Example Ecoregions

As described in Appendix 6-A, we considered the effect, at 16 representative ecoregions across the U.S., of using different trajectory hit rates as criteria for monitoring site inclusion. Figures 6A-5 through 6A-20 of Appendix 6A show the results of those sensitivity tests for the annual SO_2 metric and 16 ecoregions. The following maps similar show this effect over the same 16 ecoregions for the annual NO_2, annual $PM_{2.5}$, and the 3-hour SO_2 metrics.

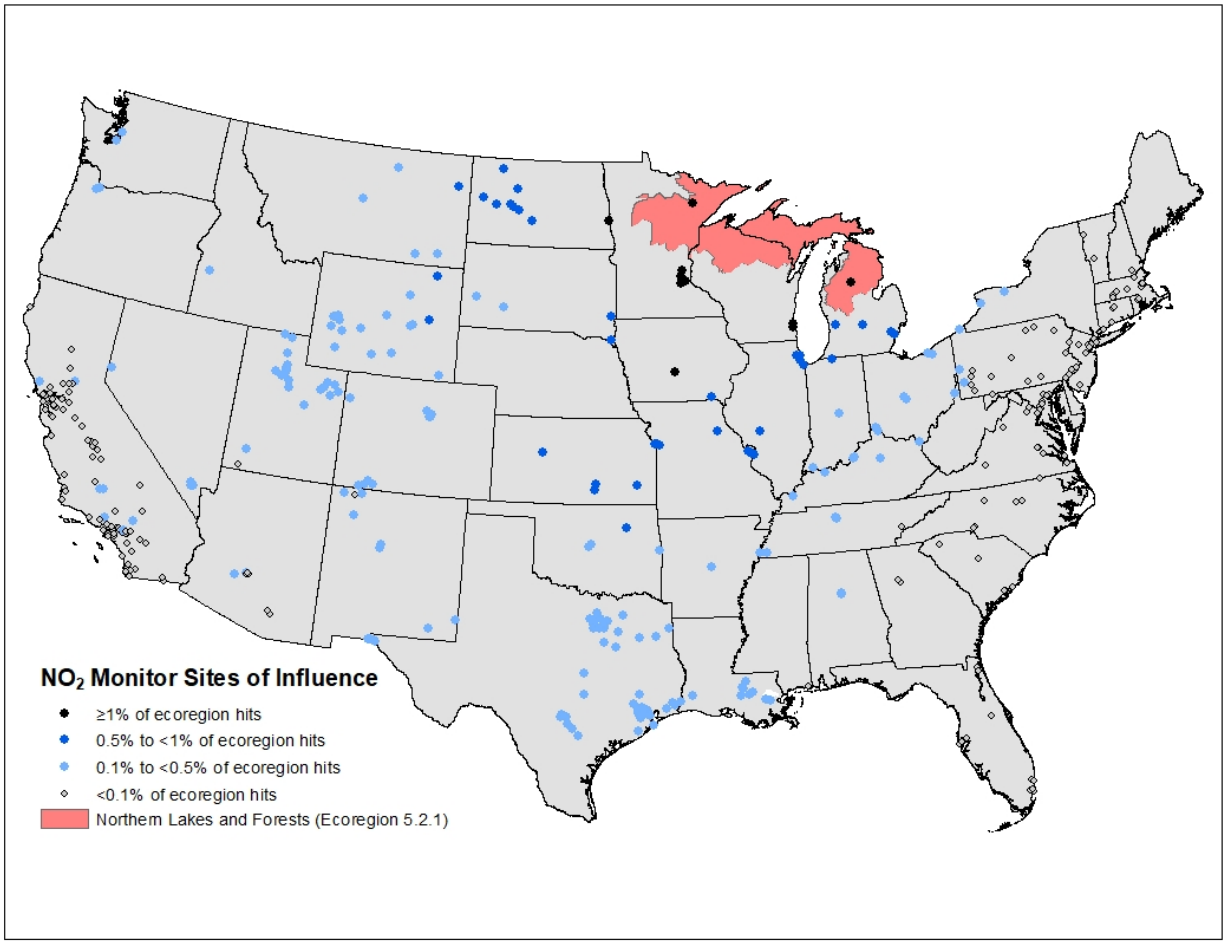

Figure 1. Monitoring sites (annual NO_2 metric) of potential influence for ecoregion 5.2.1 (red shaded region).

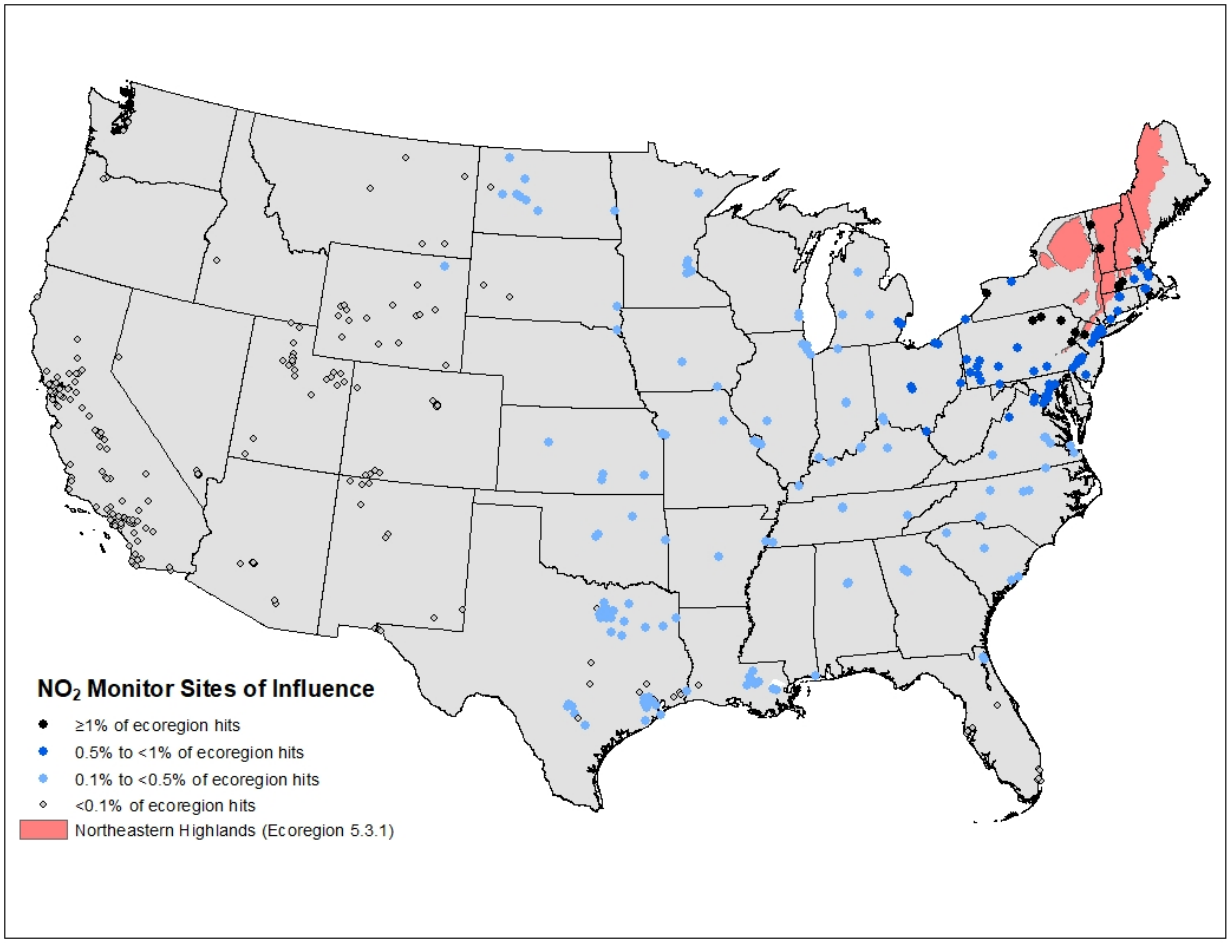

Figure 2. Monitoring sites (annual NO_2 metric) of potential influence for ecoregion 5.3.1 (red shaded region).

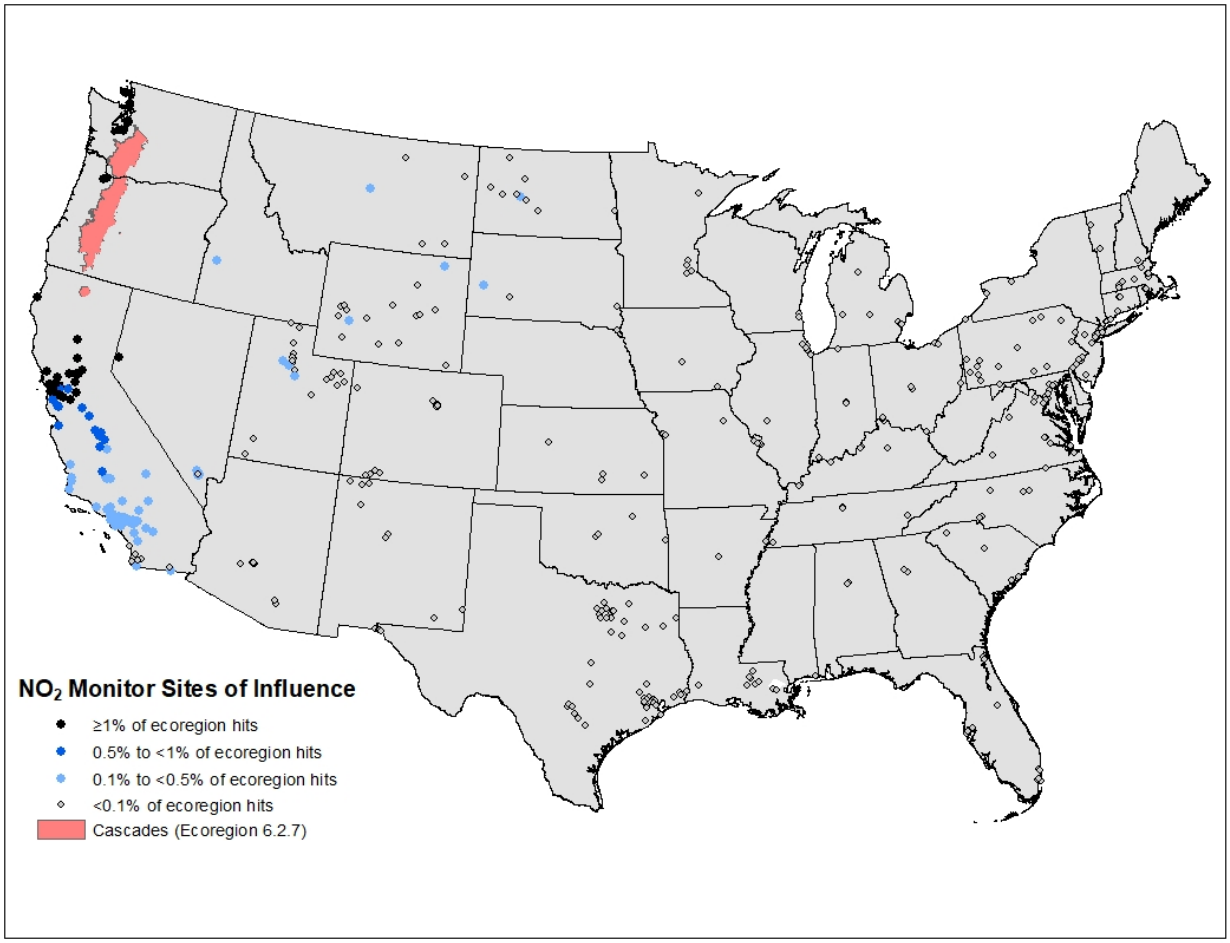

Figure 3. Monitoring sites (annual NO_2 metric) of potential influence for ecoregion 6.2.7 (red shaded region).

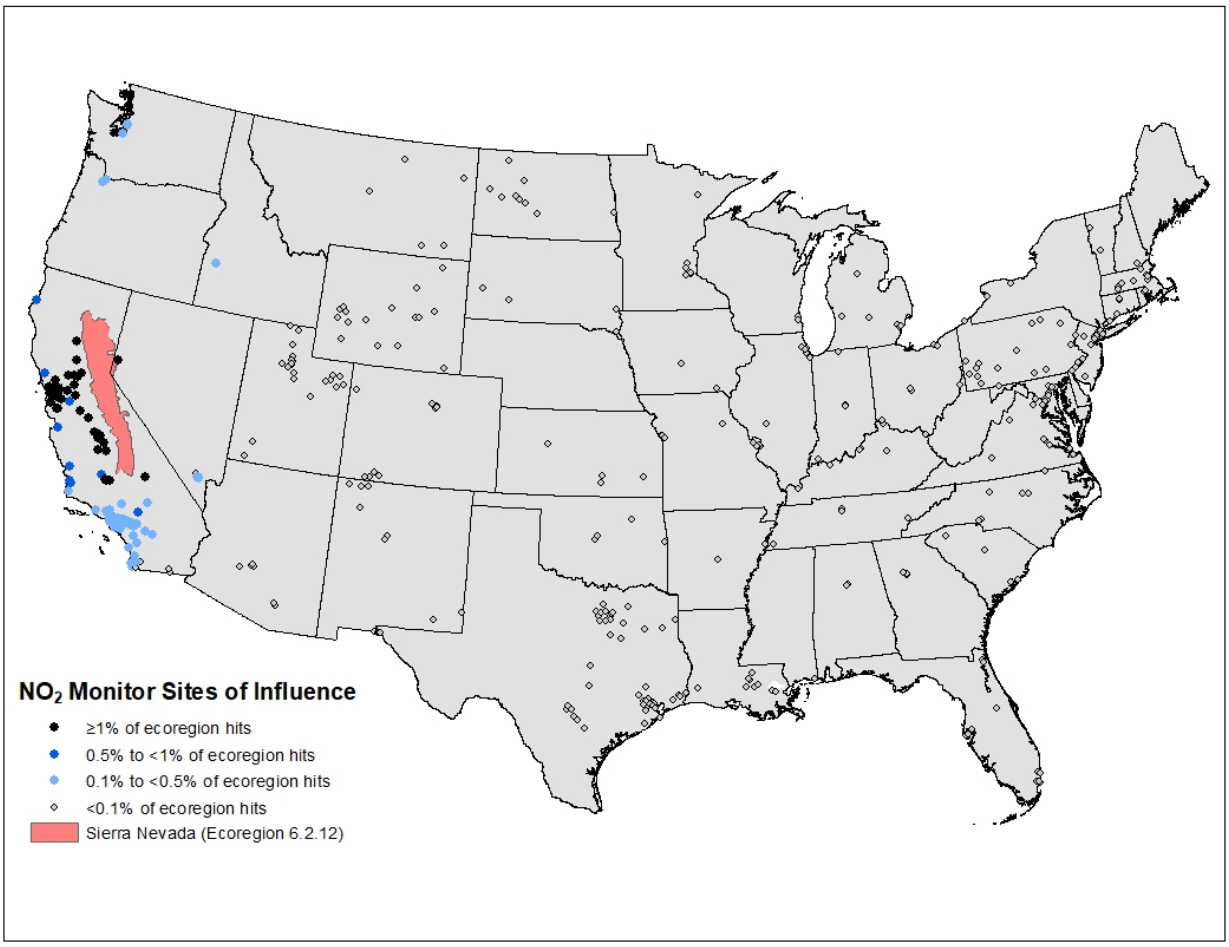

Figure 4. Monitoring sites (annual NO_2 metric) of potential influence for ecoregion 6.2.12 (red shaded region).

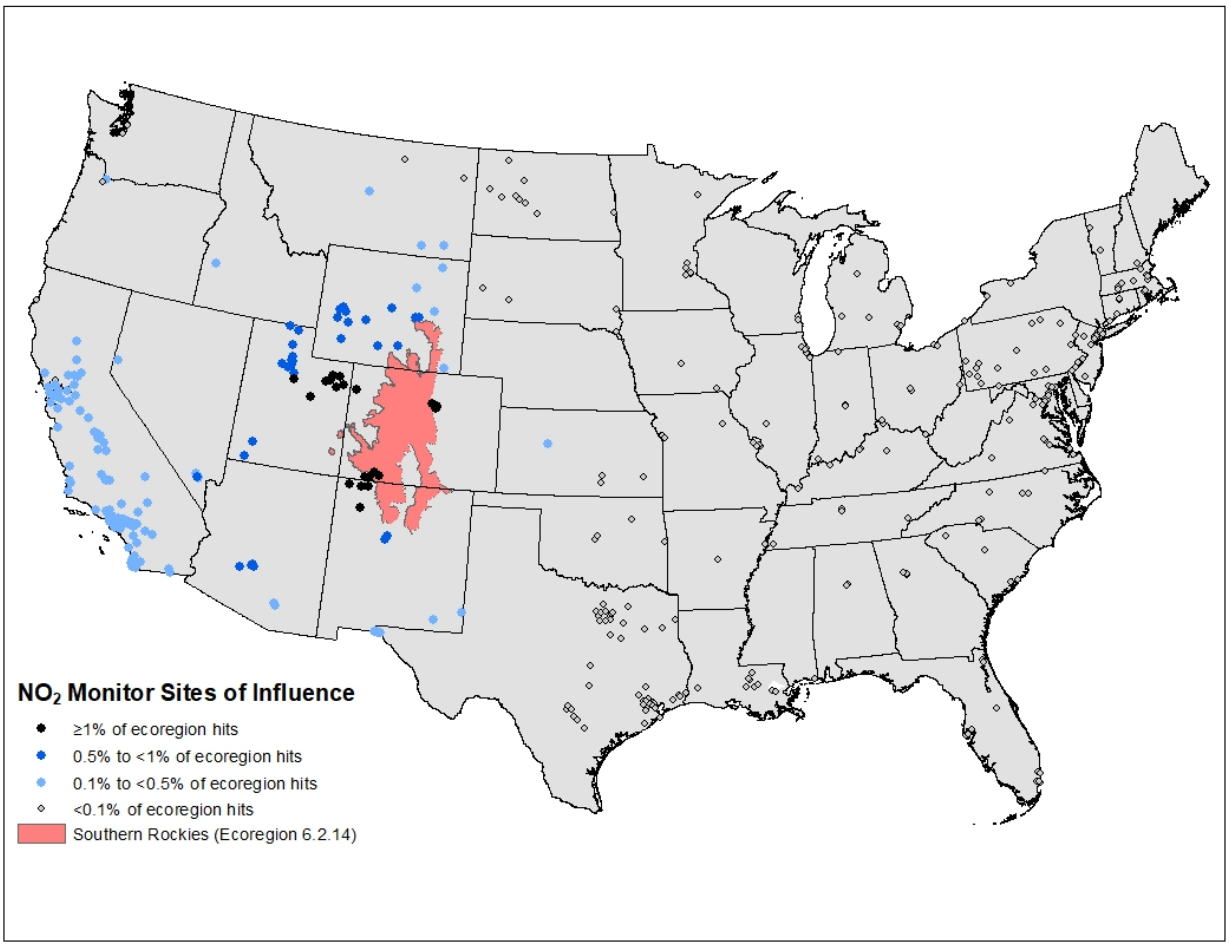

Figure 5. Monitoring sites (annual NO_2 metric) of potential influence for ecoregion 6.2.14 (red shaded region).

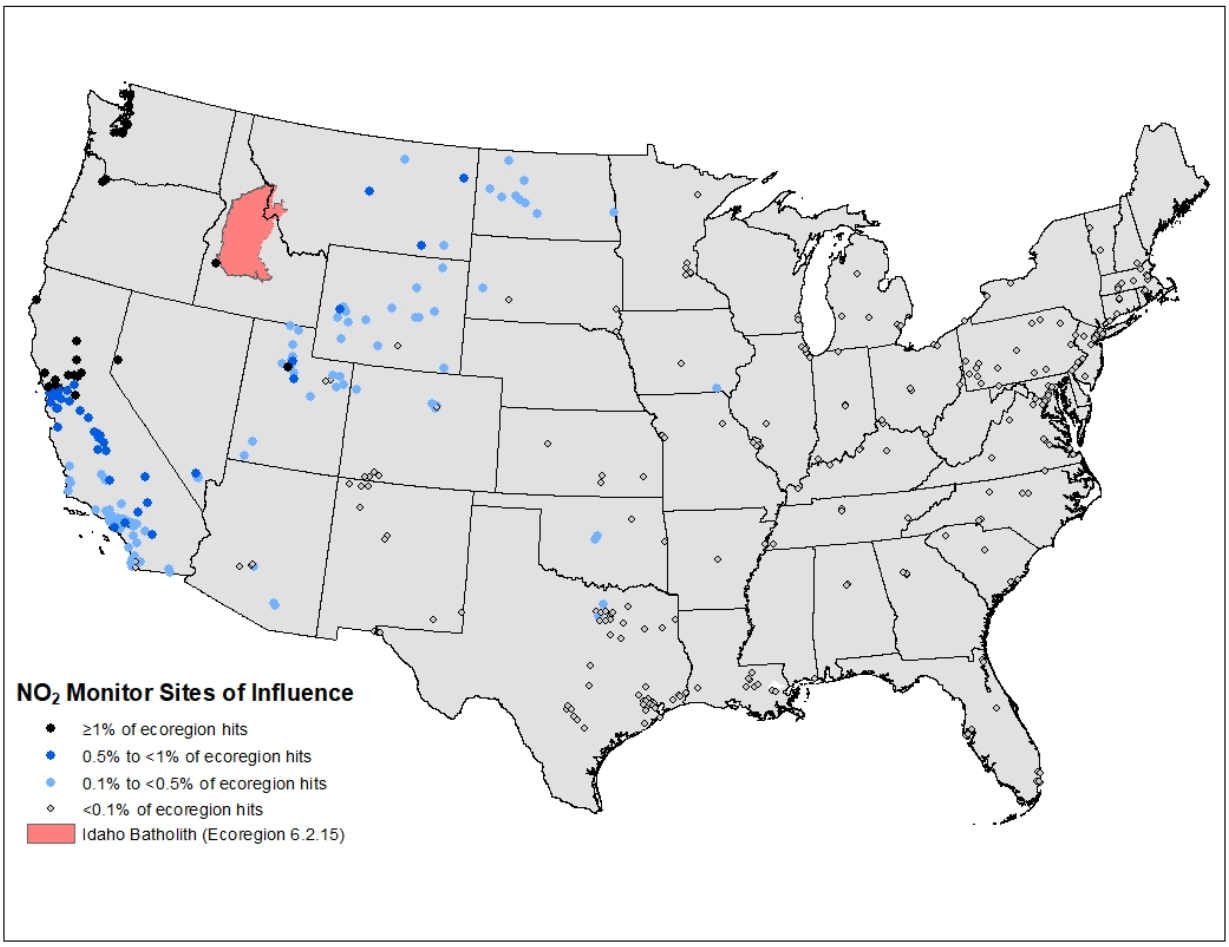

Figure 6. Monitoring sites (annual NO_2 metric) of potential influence for ecoregion 6.2.15 (red shaded region).

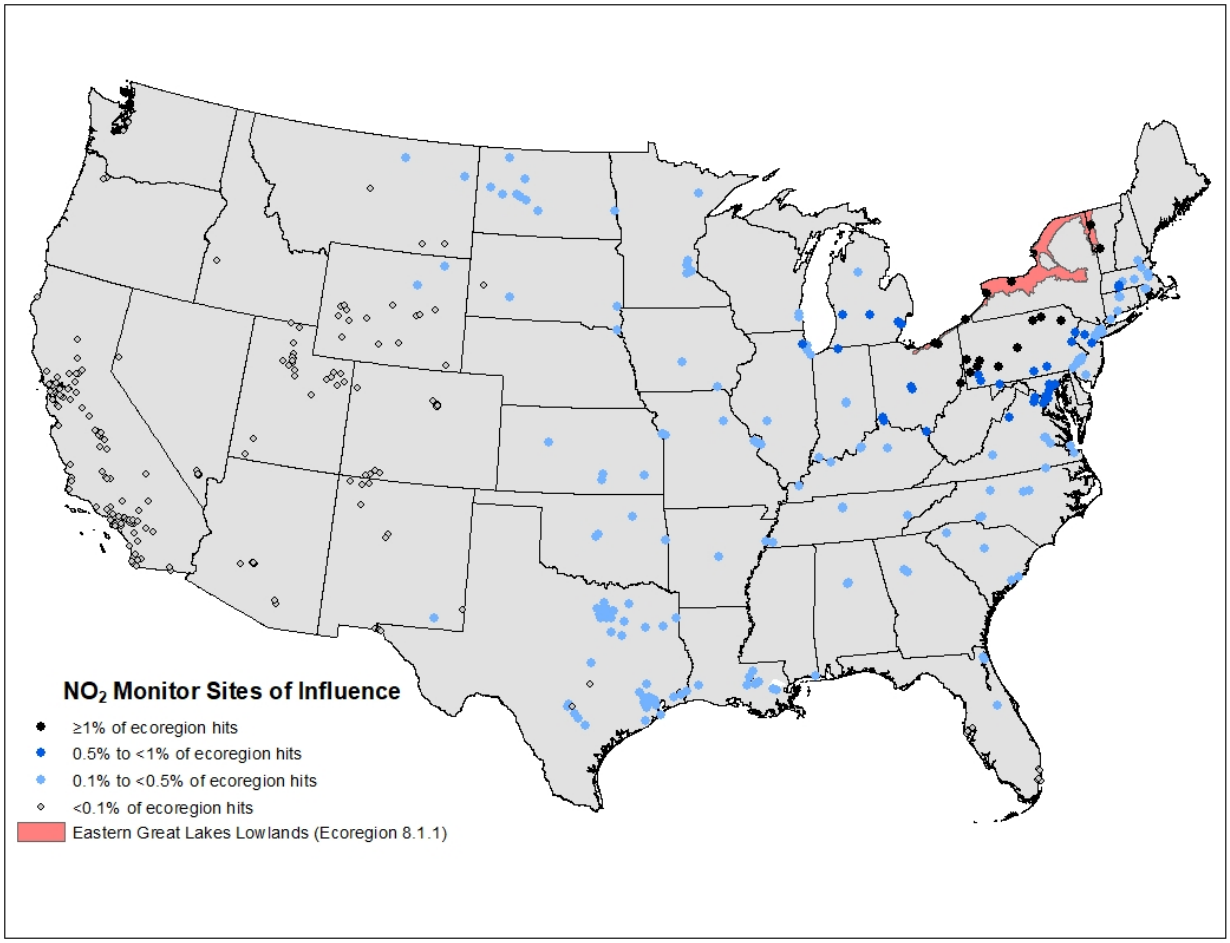

Figure 7. Monitoring sites (annual NO_2 metric) of potential influence for ecoregion 8.1.1 (red shaded region).

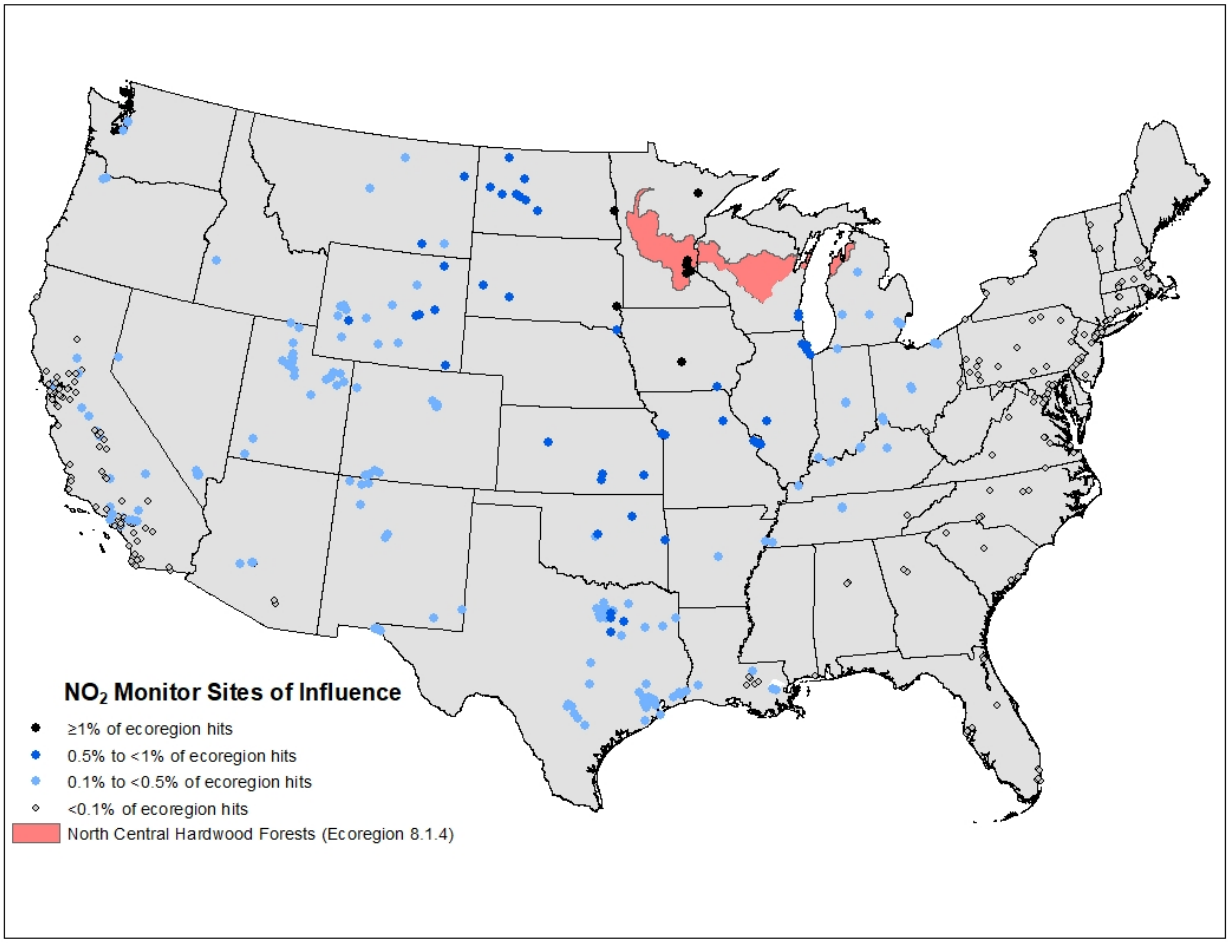

Figure 8. Monitoring sites (annual NO_2 metric) of potential influence for ecoregion 8.1.4 (red shaded region).

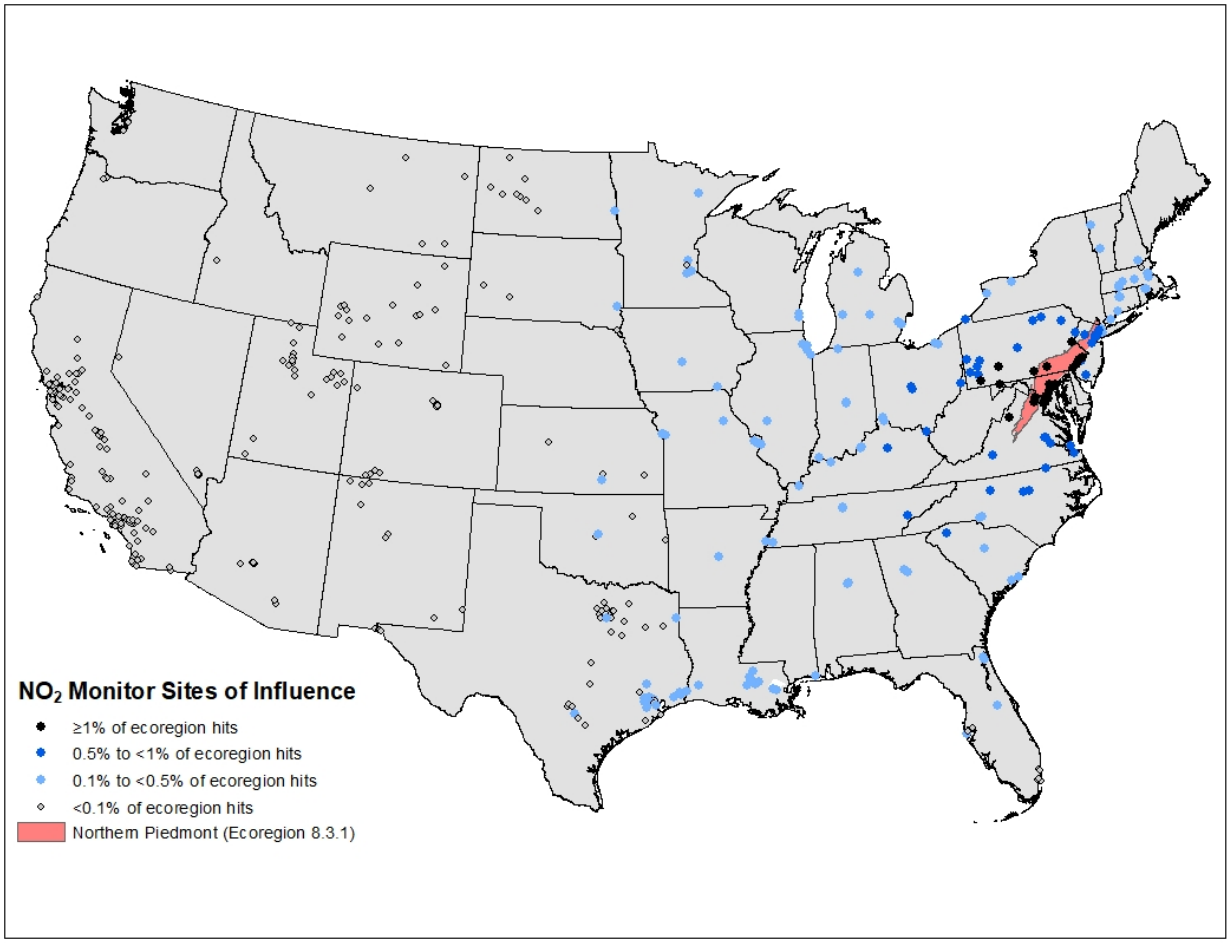

Figure 9. Monitoring sites (annual NO_2 metric) of potential influence for ecoregion 8.3.1 (red shaded region).

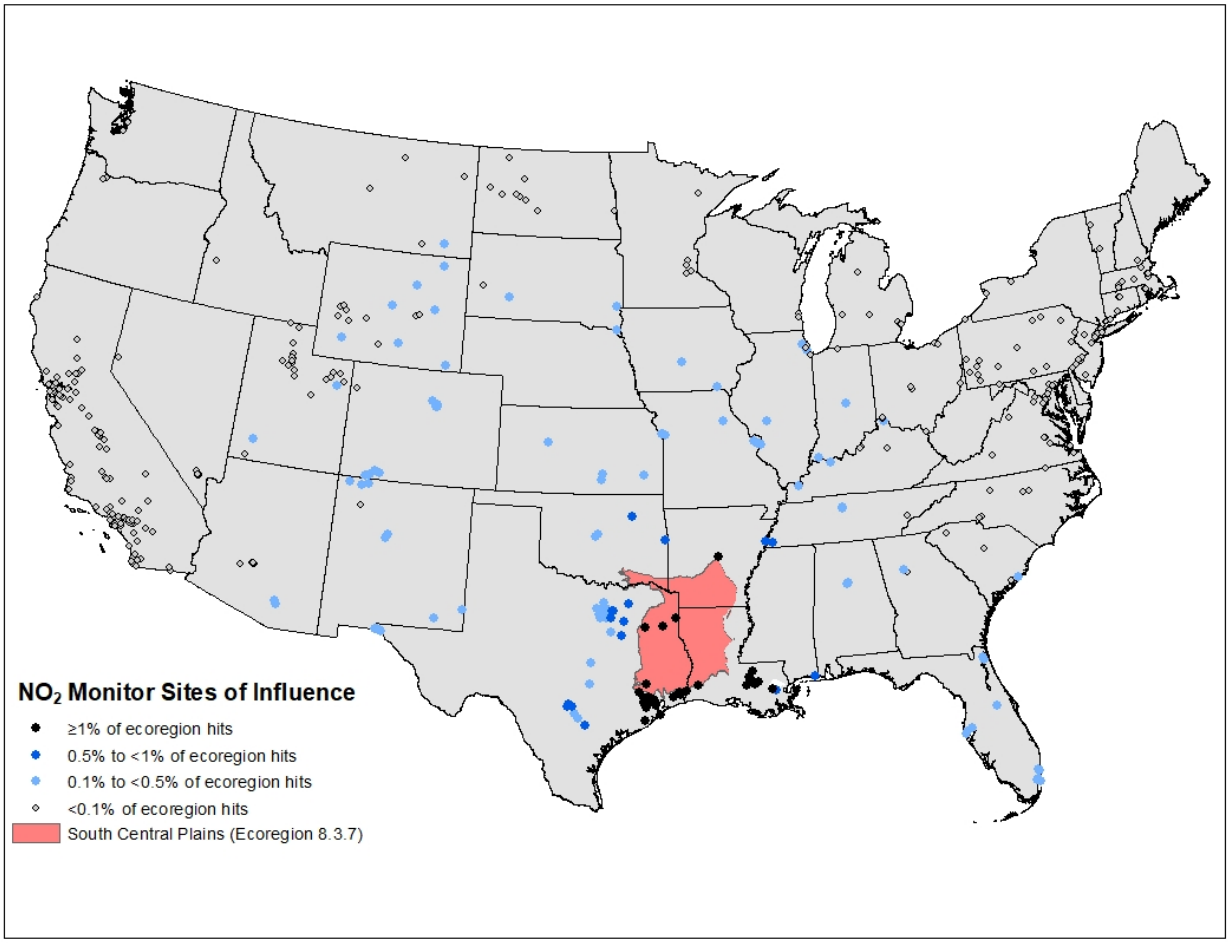

Figure 10. Monitoring sites (annual NO_2 metric) of potential influence for ecoregion 8.3.7 (red shaded region).

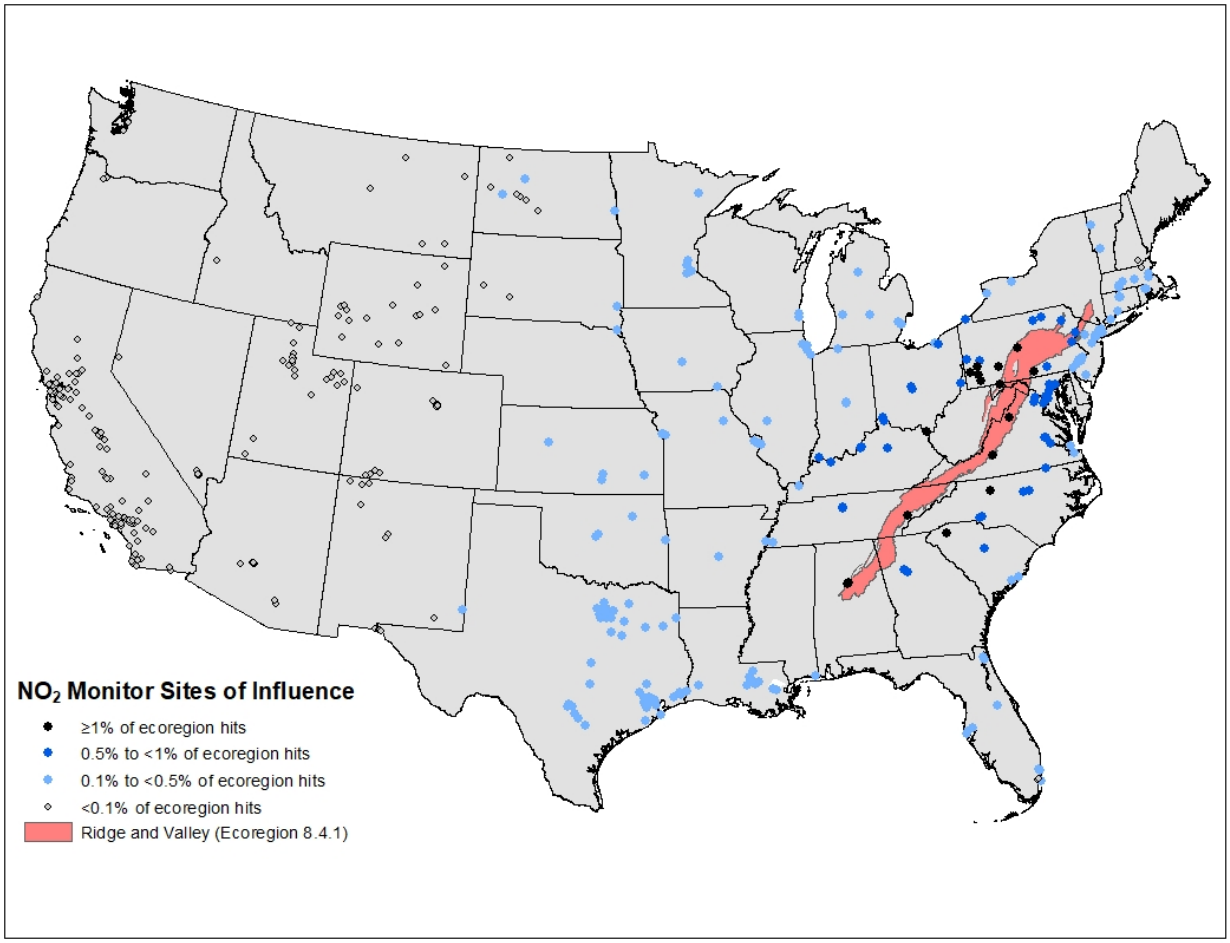

Figure 11. Monitoring sites (annual NO_2 metric) of potential influence for ecoregion 8.4.1 (red shaded region).

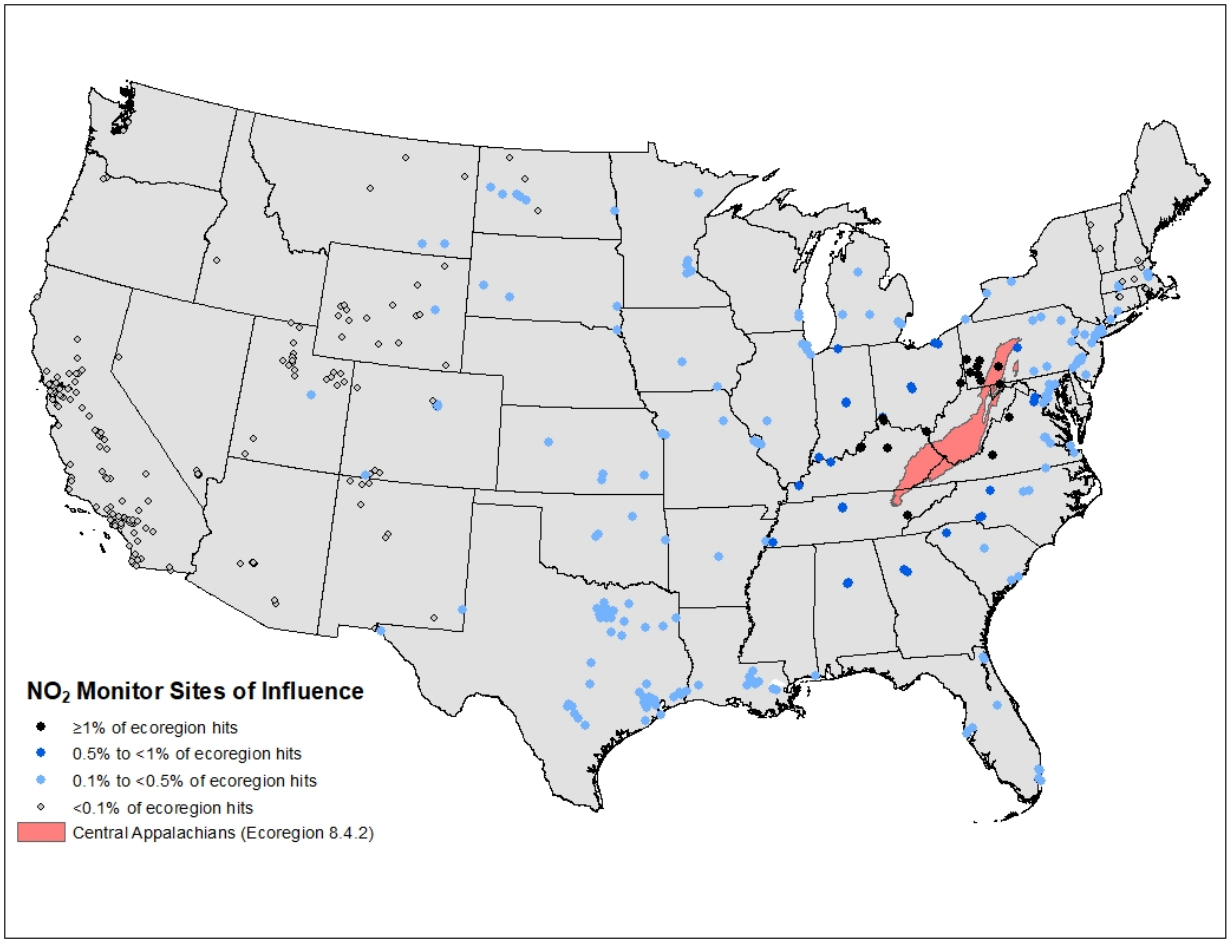

Figure 12. Monitoring sites (annual NO_2 metric) of potential influence for ecoregion 8.4.2 (red shaded region).

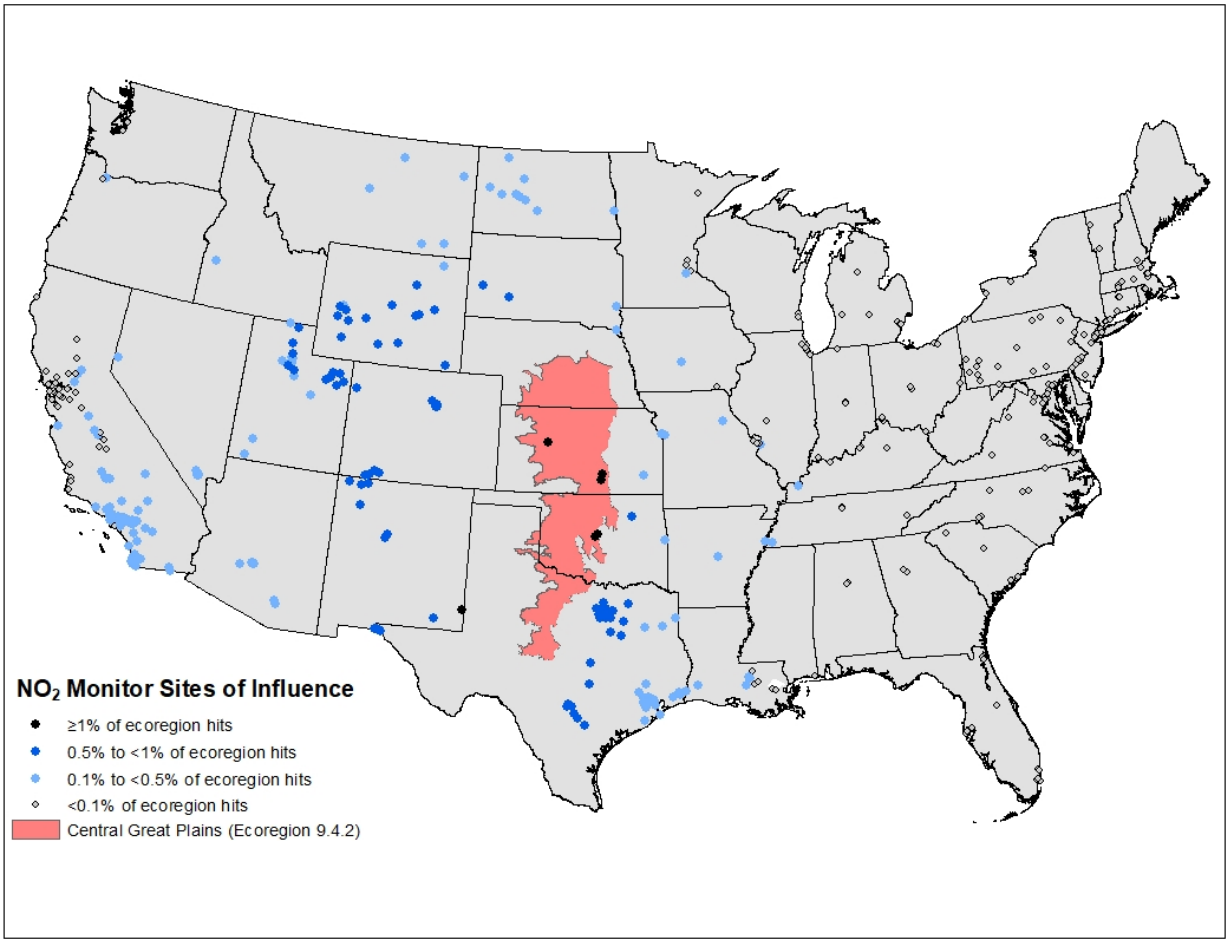

Figure 13. Monitoring sites (annual NO_2 metric) of potential influence for ecoregion 9.4.2 (red shaded region).

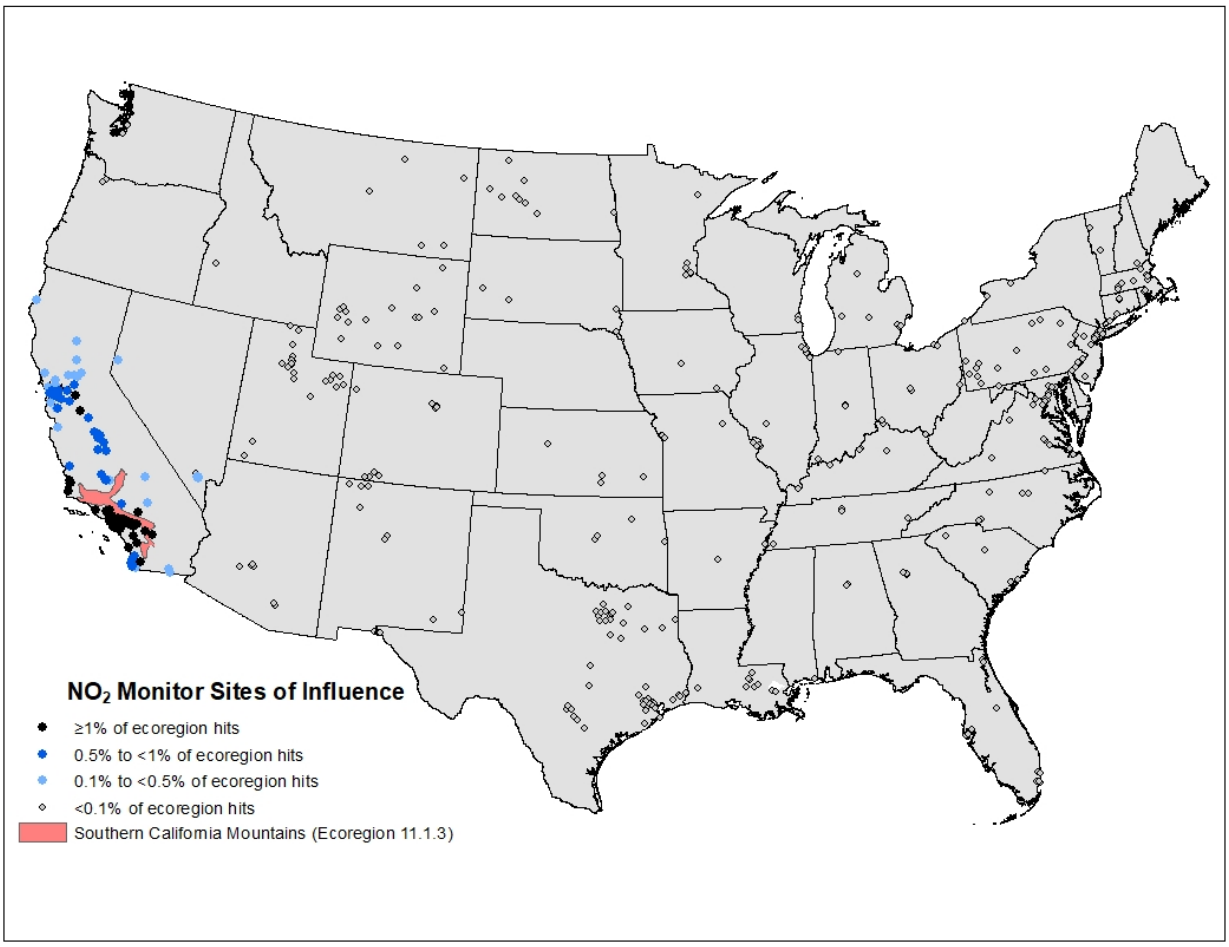

Figure 14. Monitoring sites (annual NO_2 metric) of potential influence for ecoregion 11.1.3 (red shaded region).

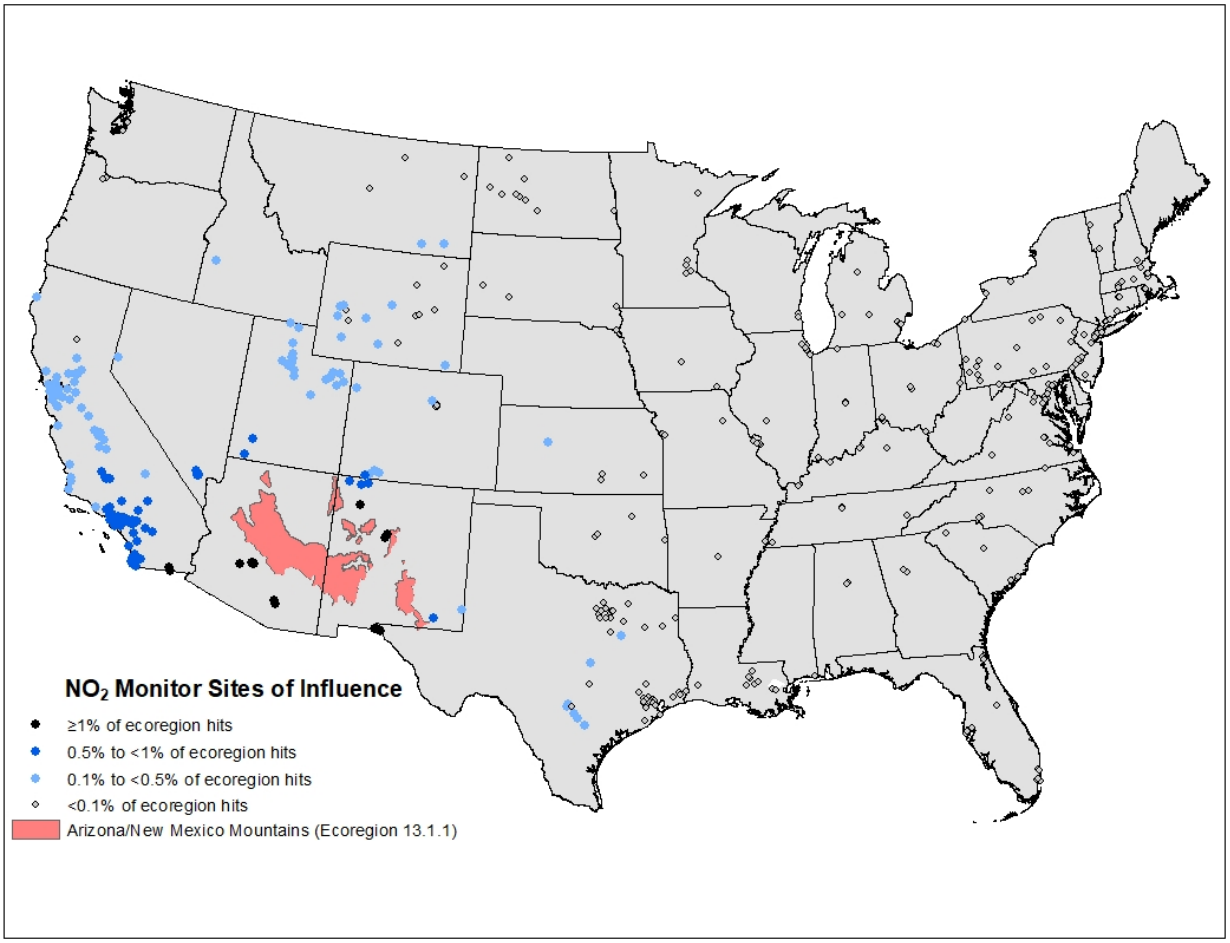

Figure 15. Monitoring sites (annual NO_2 metric) of potential influence for ecoregion 13.1.1 (red shaded region).

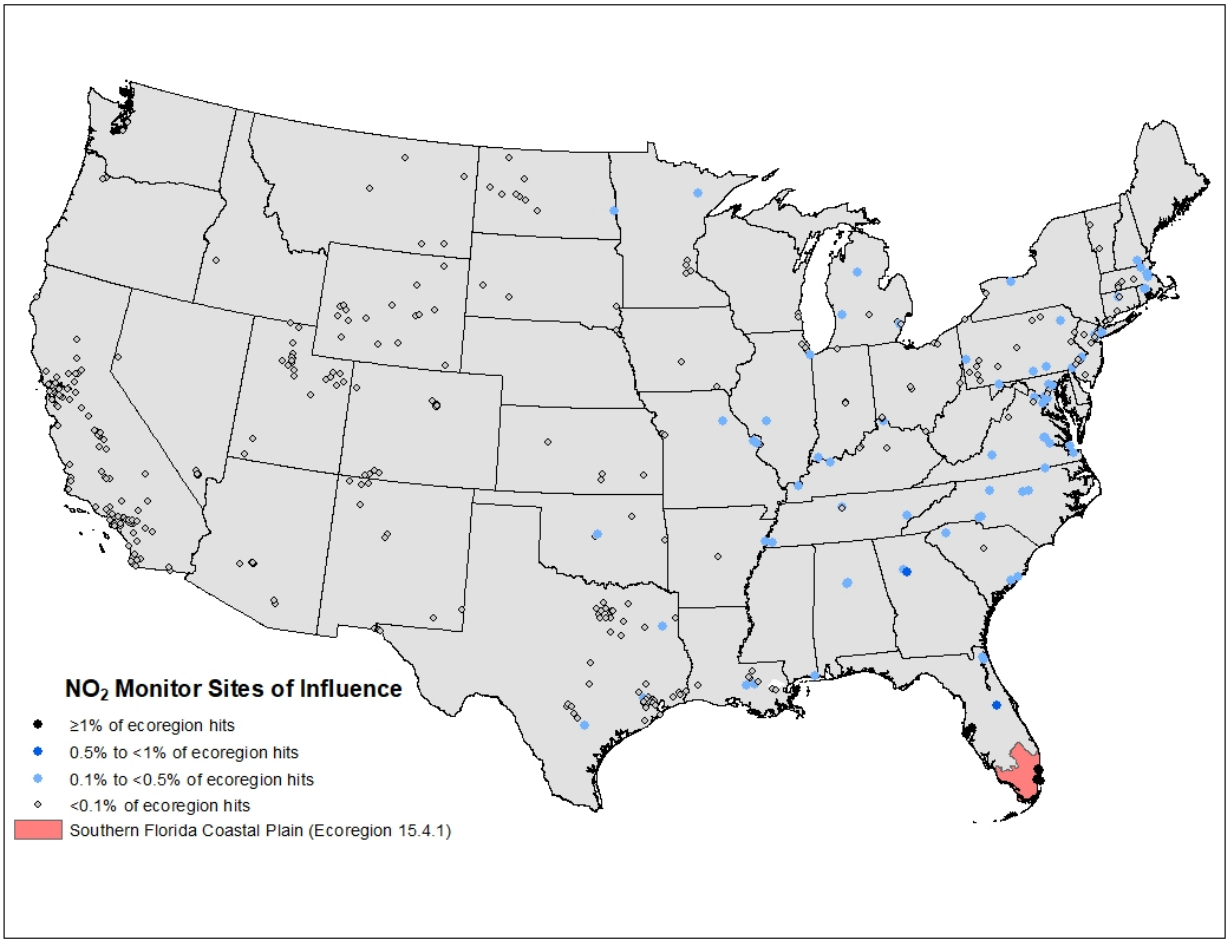

Figure 16. Monitoring sites (annual NO_2 metric) of potential influence for ecoregion 15.4.1 (red shaded region).

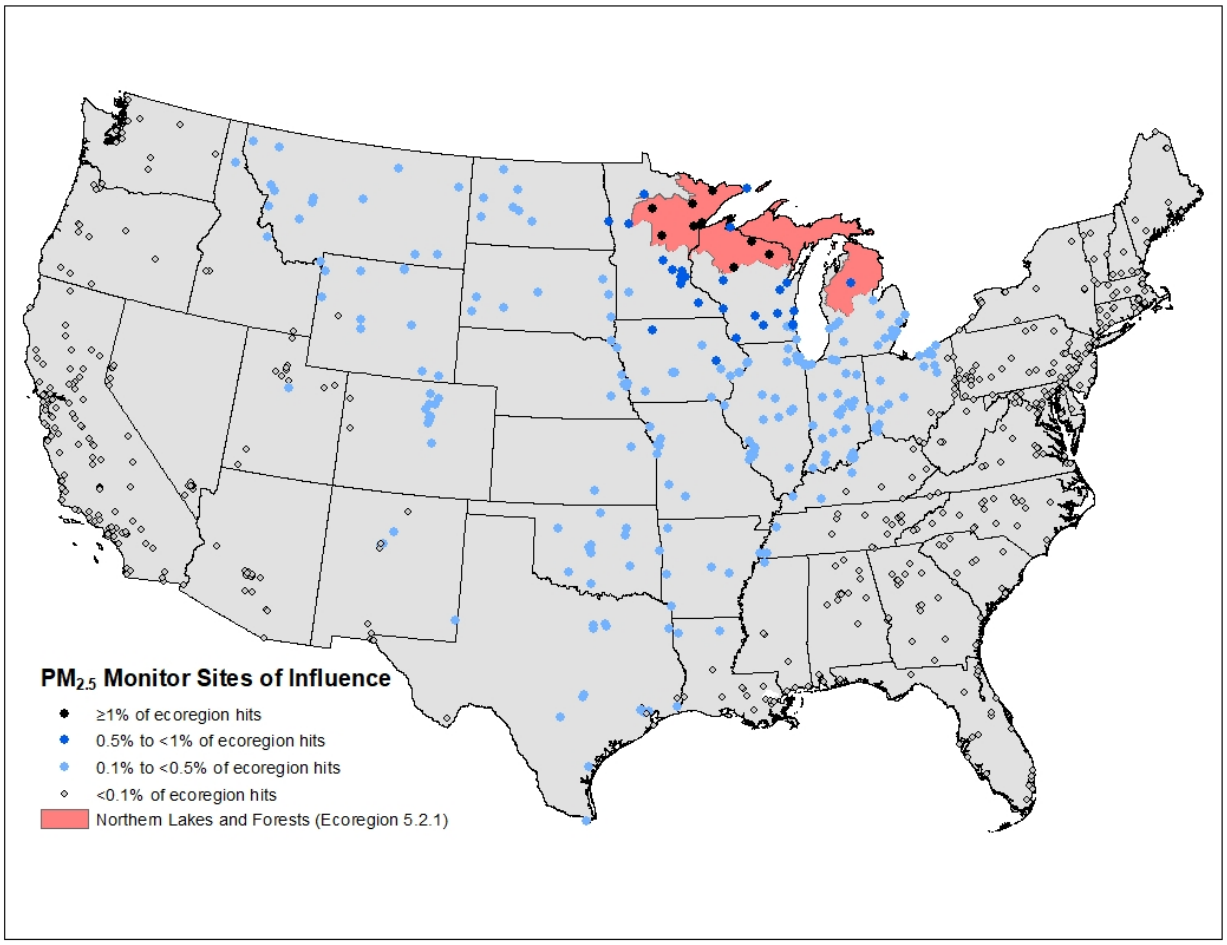

Figure 17. Monitoring sites (annual $PM_{2.5}$ metric) of potential influence for ecoregion 5.2.1 (red shaded region).

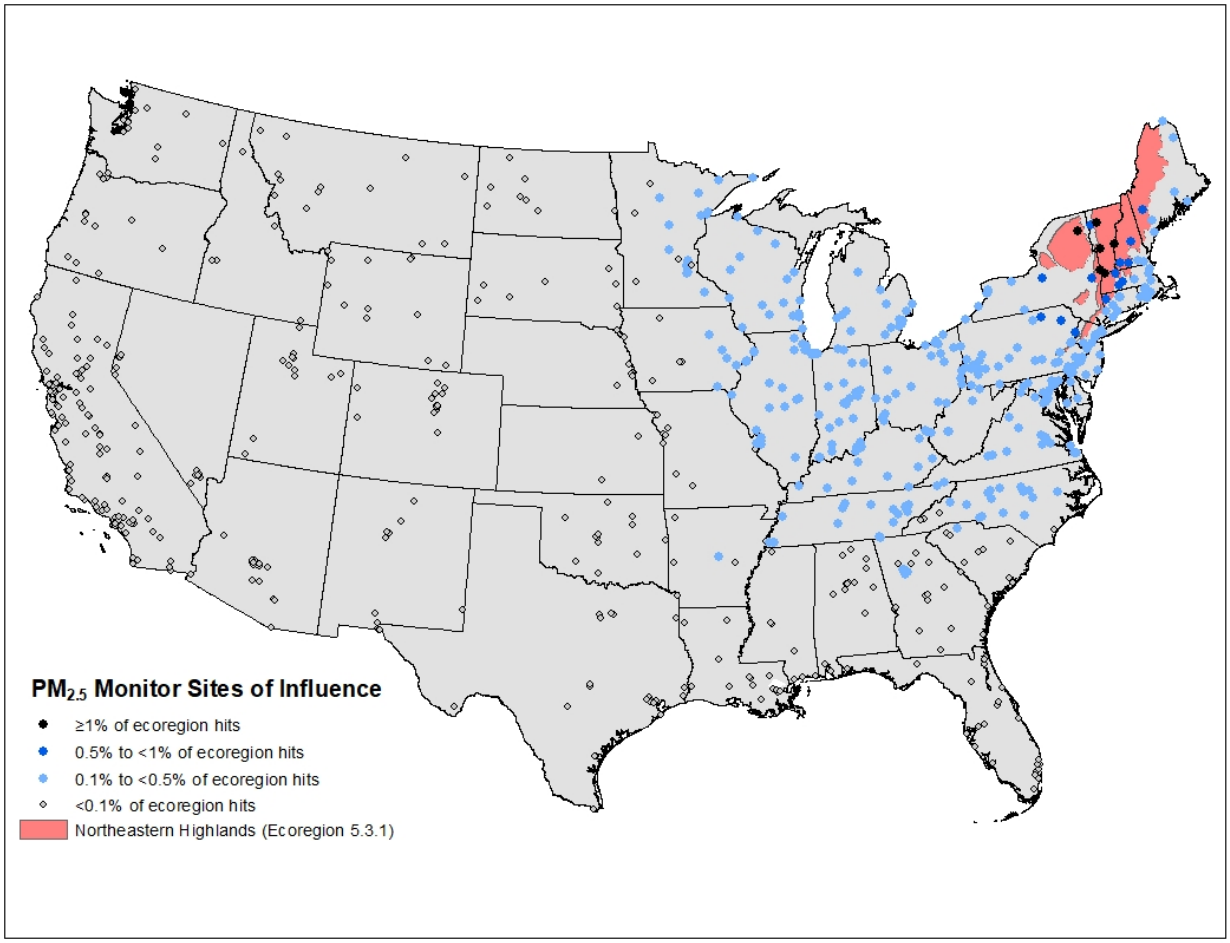

Figure 18. Monitoring sites (annual $PM_{2.5}$ metric) of potential influence for ecoregion 5.3.1 (red shaded region).

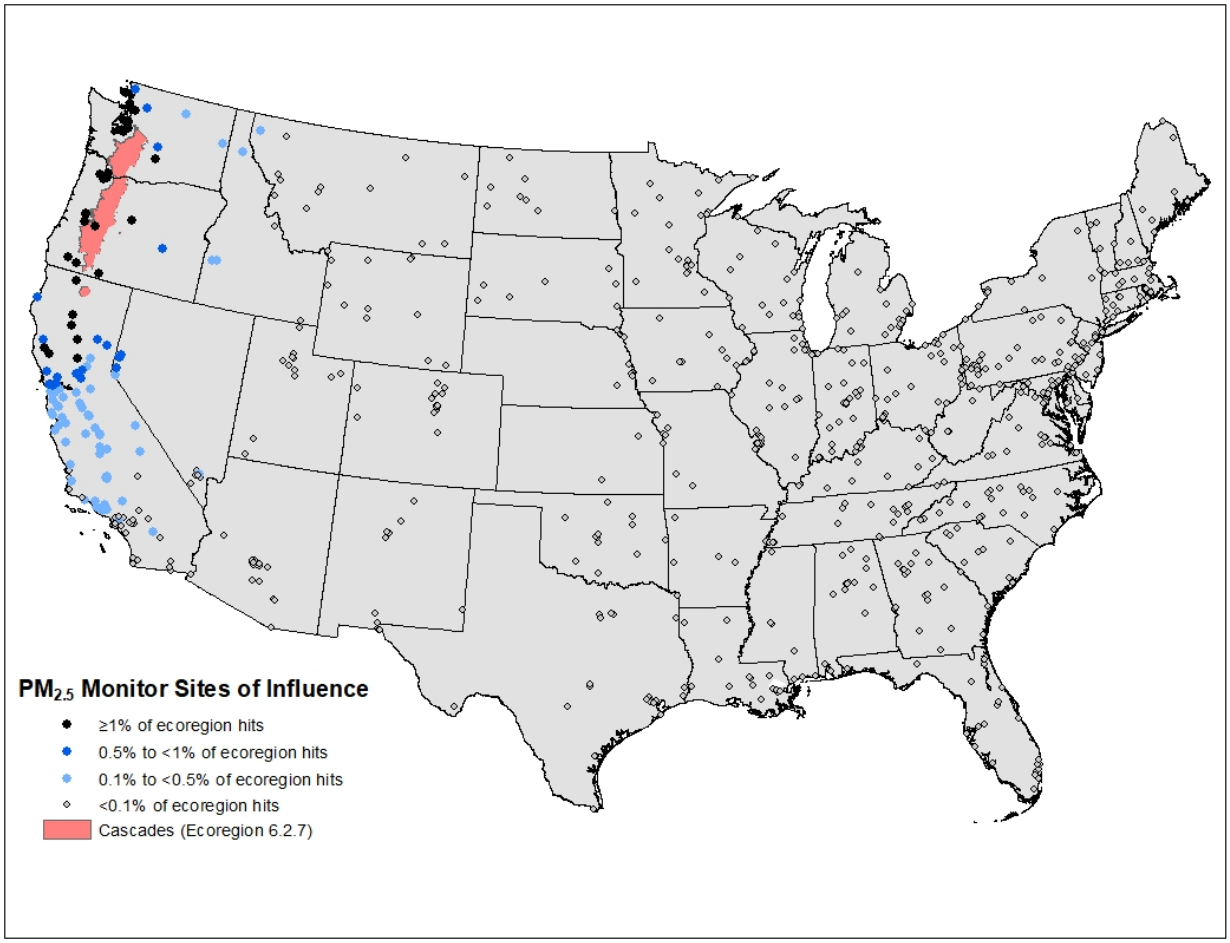

Figure 19. Monitoring sites (annual $PM_{2.5}$ metric) of potential influence for ecoregion 6.2.7 (red shaded region).

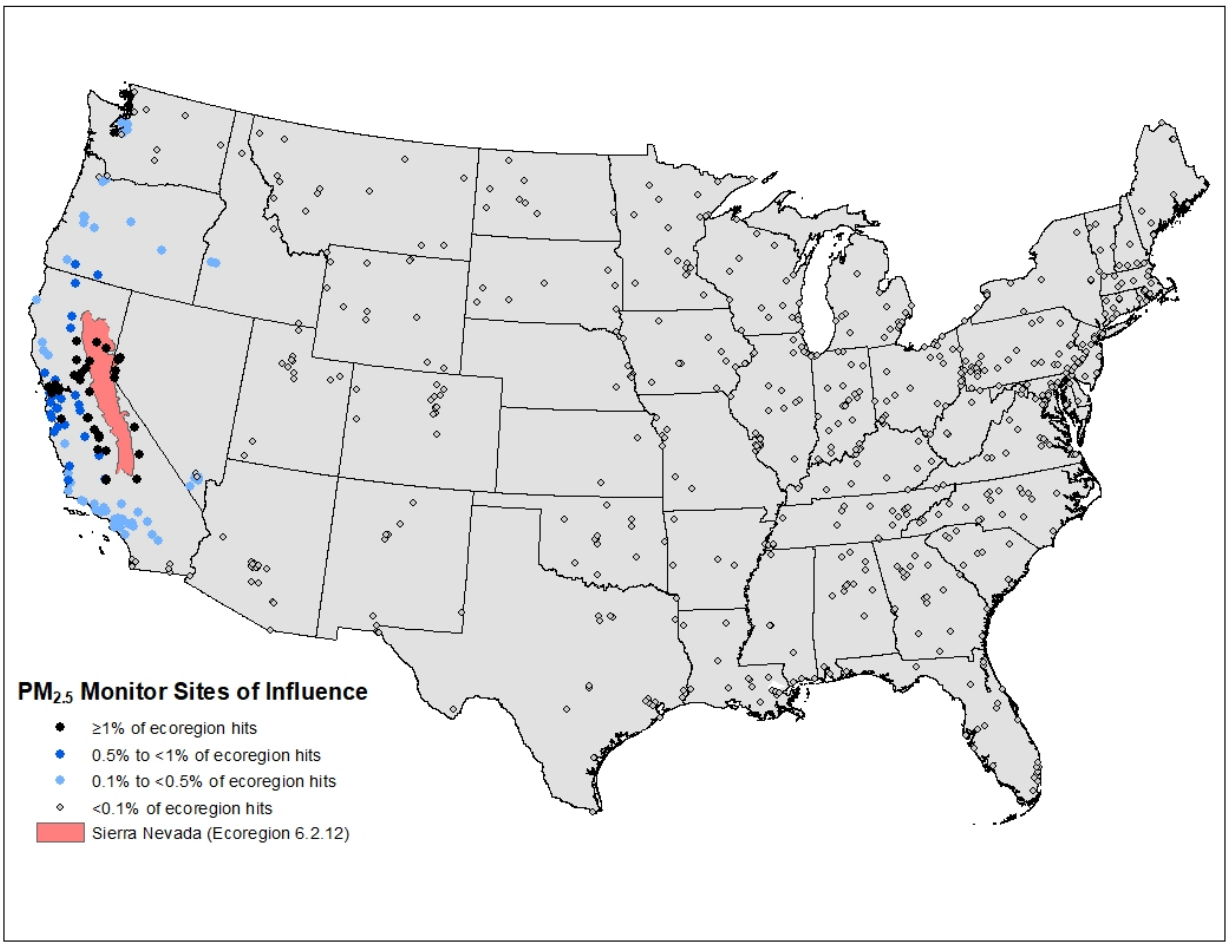

Figure 20. Monitoring sites (annual $PM_{2.5}$ metric) of potential influence for ecoregion 6.2.12 (red shaded region).

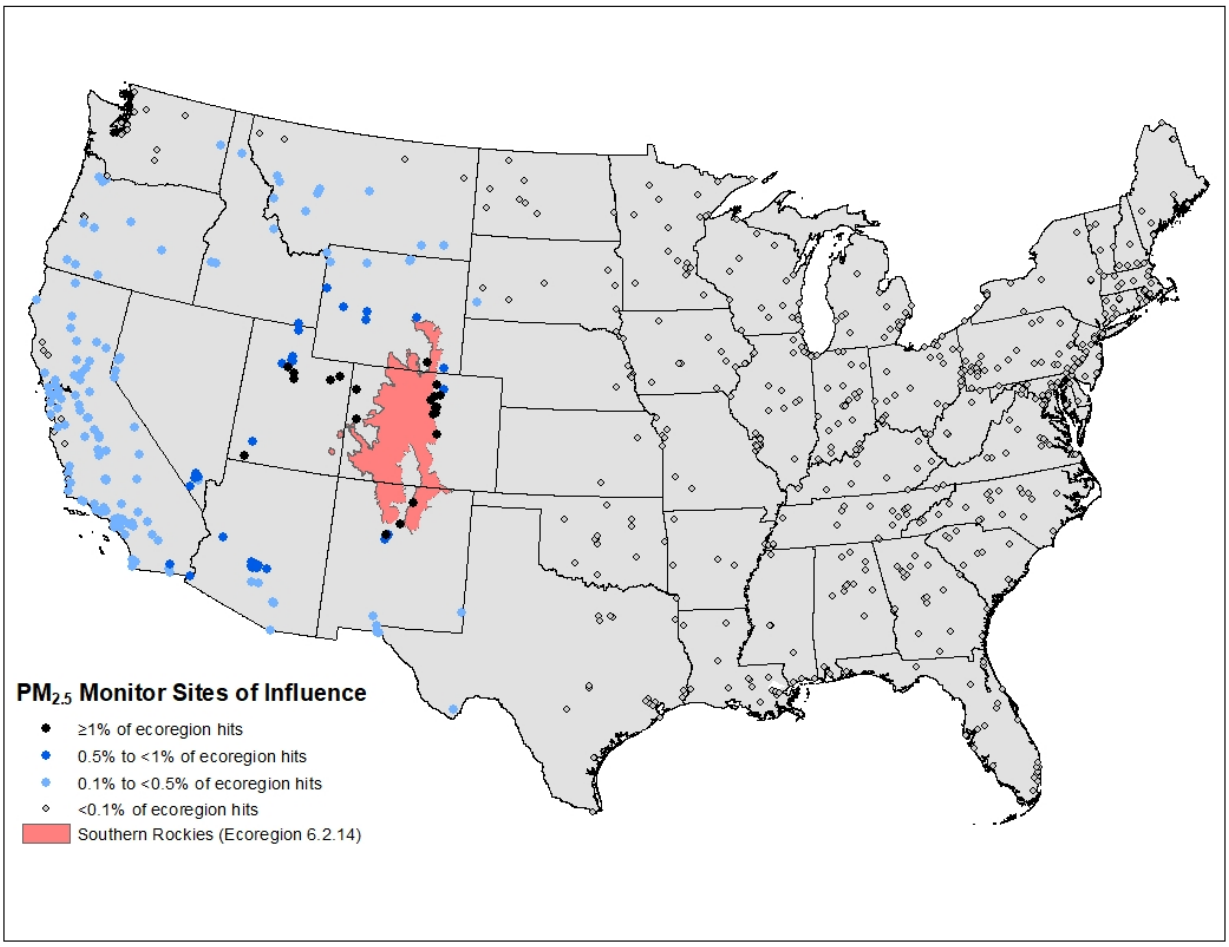

Figure 21. Monitoring sites (annual $PM_{2.5}$ metric) of potential influence for ecoregion 6.2.14 (red shaded region).

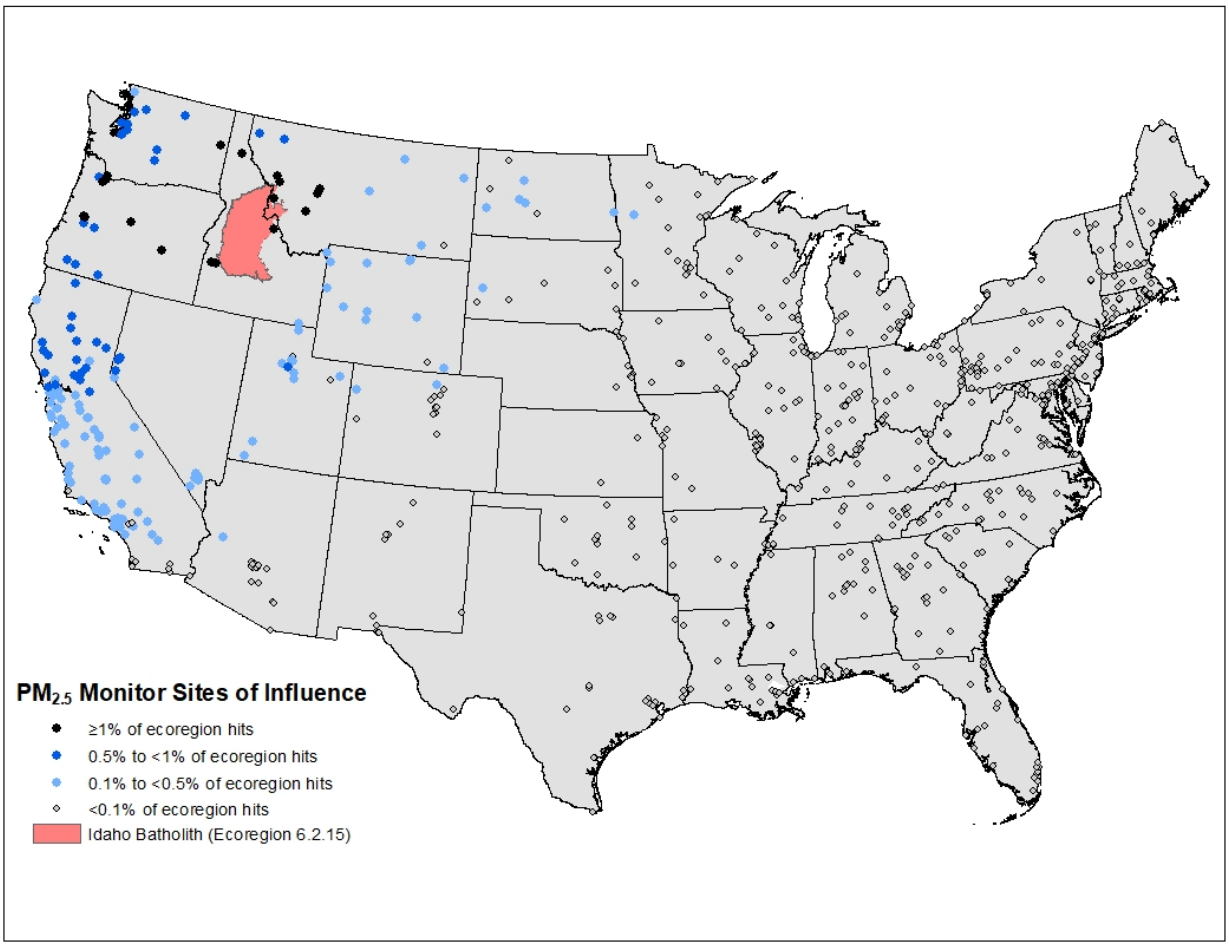

Figure 22. Monitoring sites (annual $PM_{2.5}$ metric) of potential influence for ecoregion 6.2.15 (red shaded region).

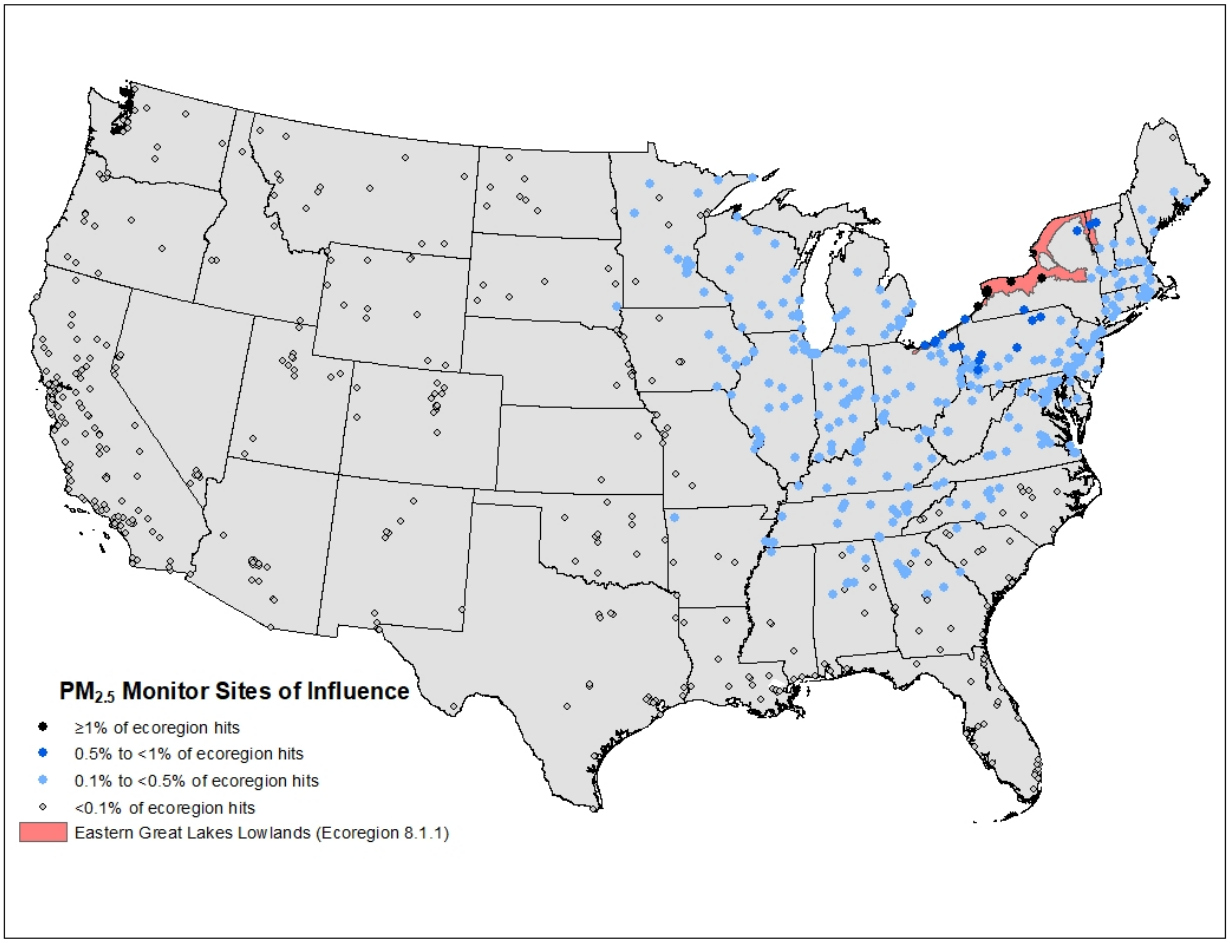

Figure 23. Monitoring sites (annual $PM_{2.5}$ metric) of potential influence for ecoregion 8.1.1 (red shaded region).

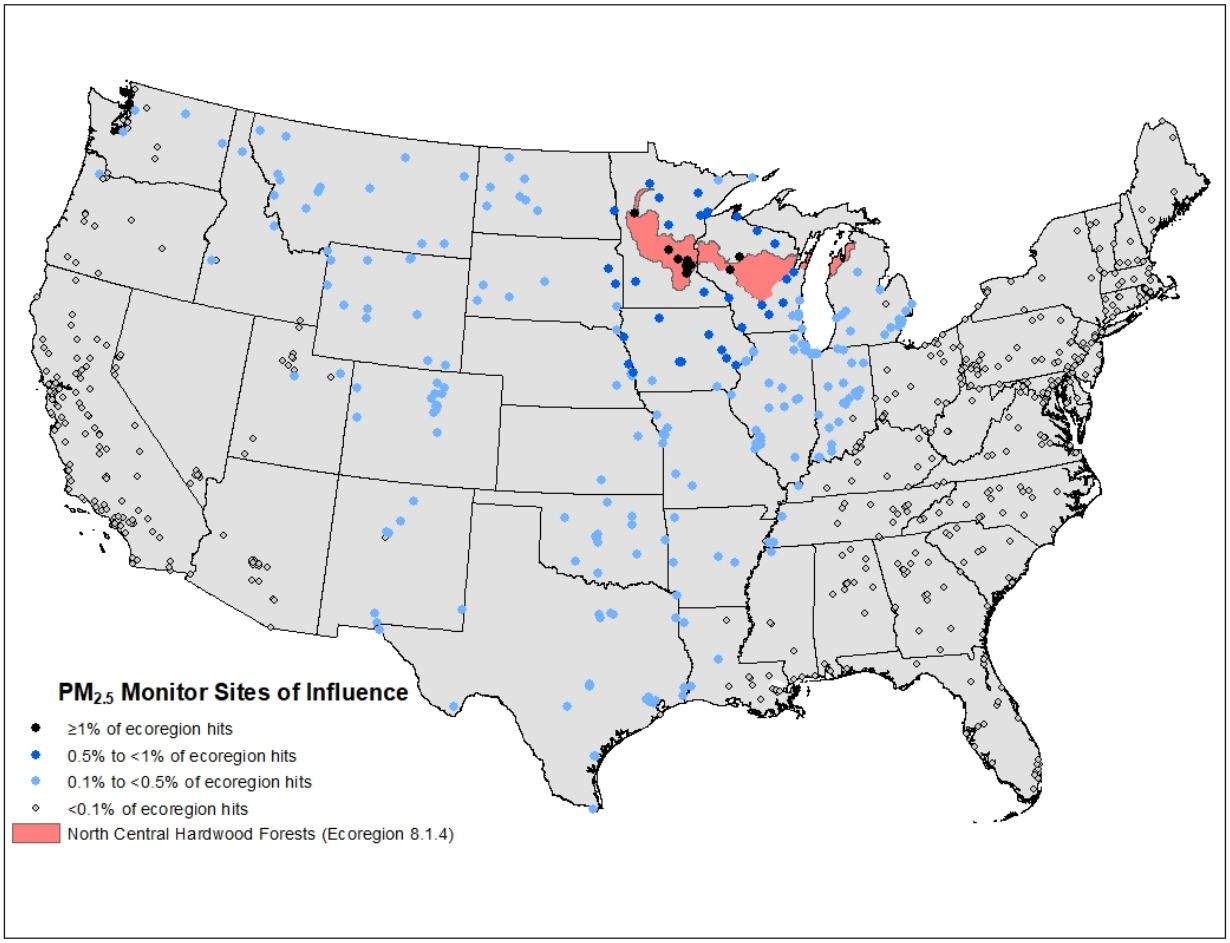

Figure 24. Monitoring sites (annual $PM_{2.5}$ metric) of potential influence for ecoregion 8.1.4 (red shaded region).

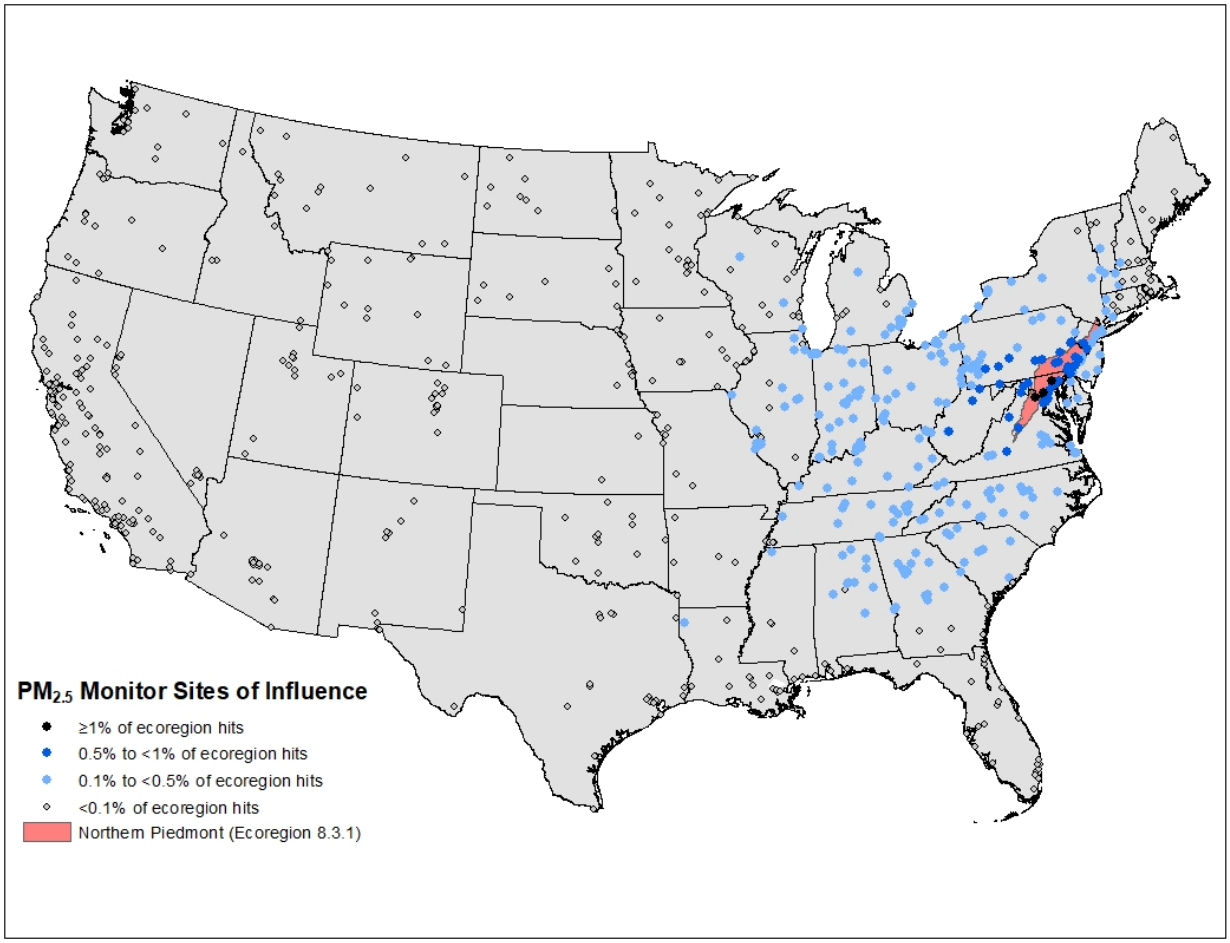

Figure 25. Monitoring sites (annual $PM_{2.5}$ metric) of potential influence for ecoregion 8.3.1 (red shaded region).

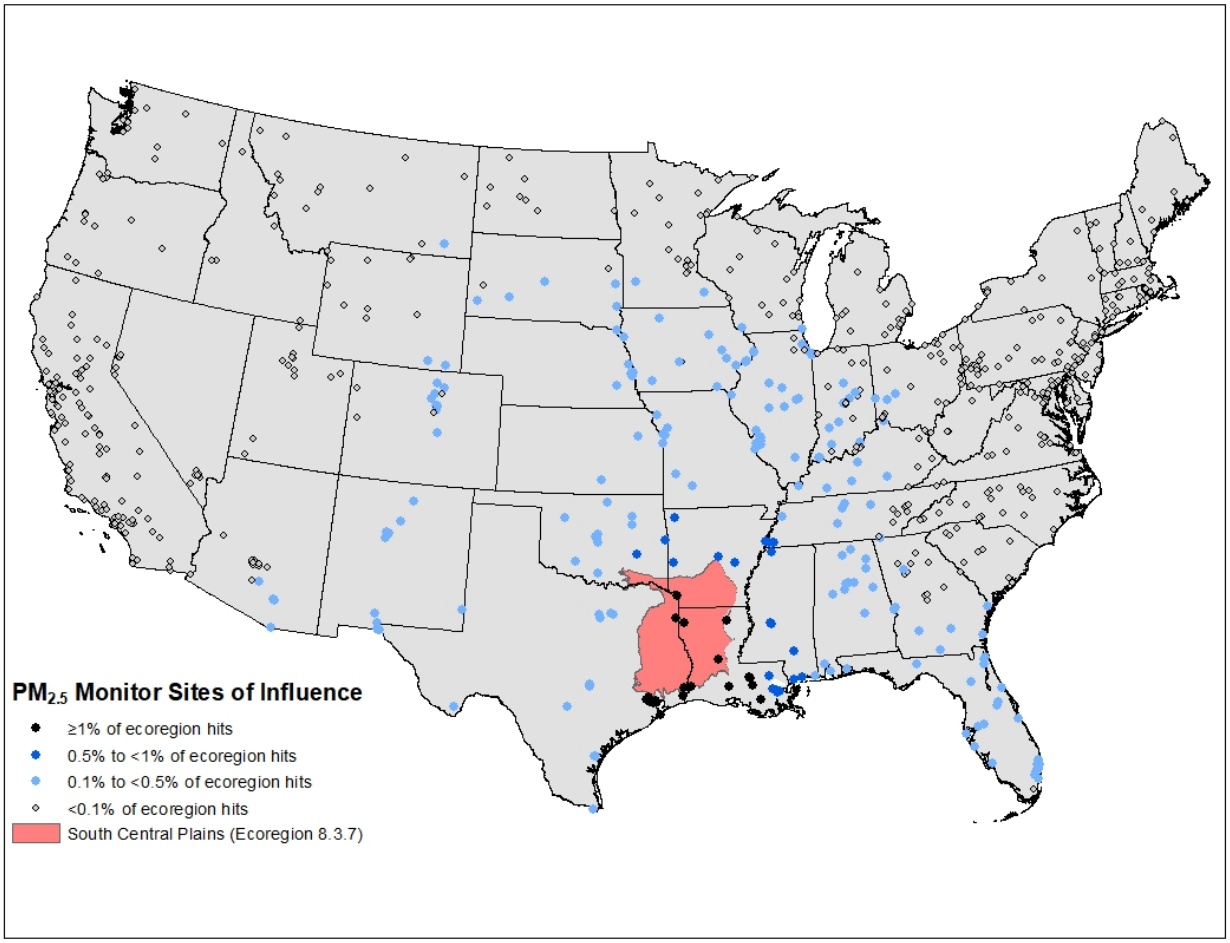

Figure 26. Monitoring sites (annual $PM_{2.5}$ metric) of potential influence for ecoregion 8.3.7 (red shaded region).

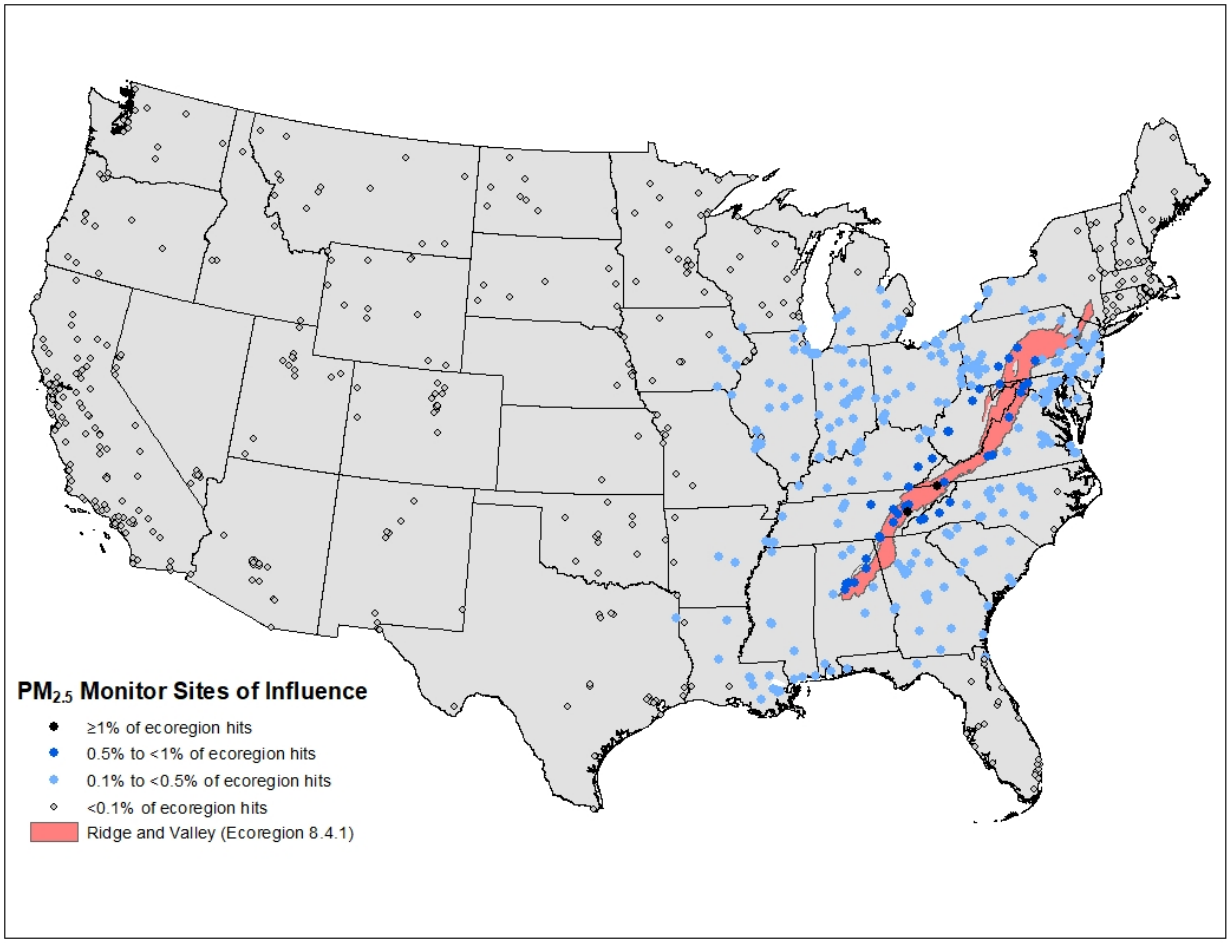

Figure 27. Monitoring sites (annual $PM_{2.5}$ metric) of potential influence for ecoregion 8.4.1 (red shaded region).

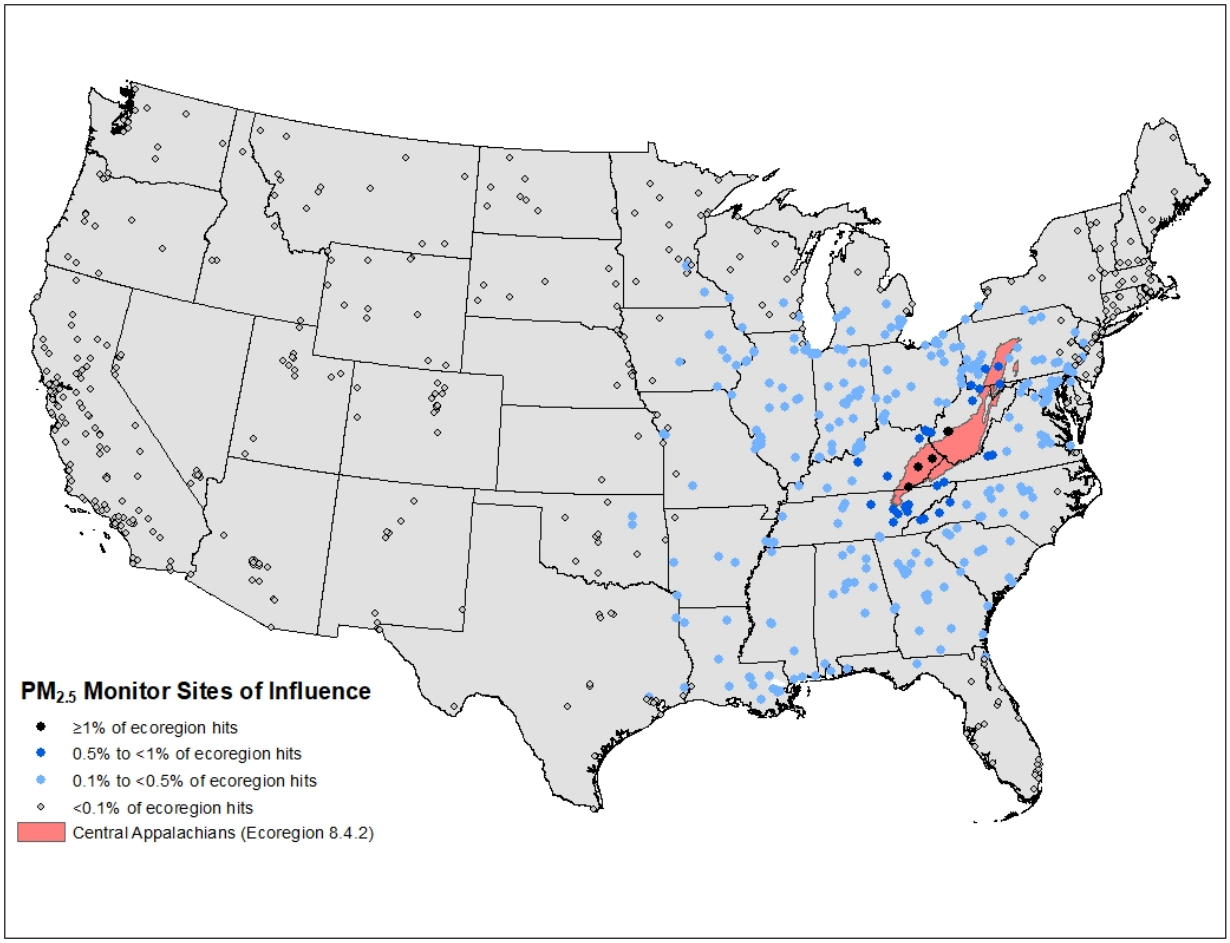

Figure 28. Monitoring sites (annual $PM_{2.5}$ metric) of potential influence for ecoregion 8.4.2 (red shaded region).

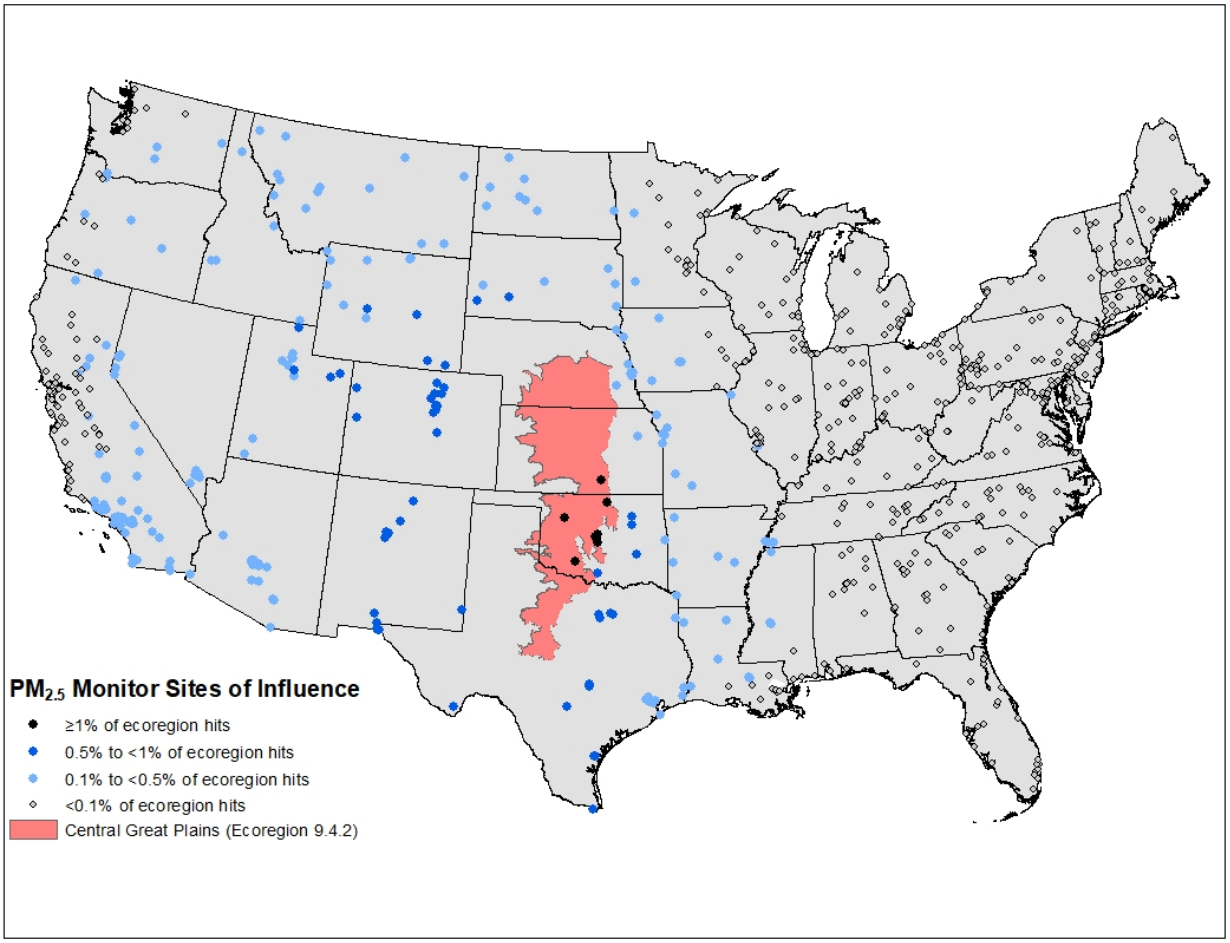

Figure 29. Monitoring sites (annual $PM_{2.5}$ metric) of potential influence for ecoregion 9.4.2 (red shaded region).

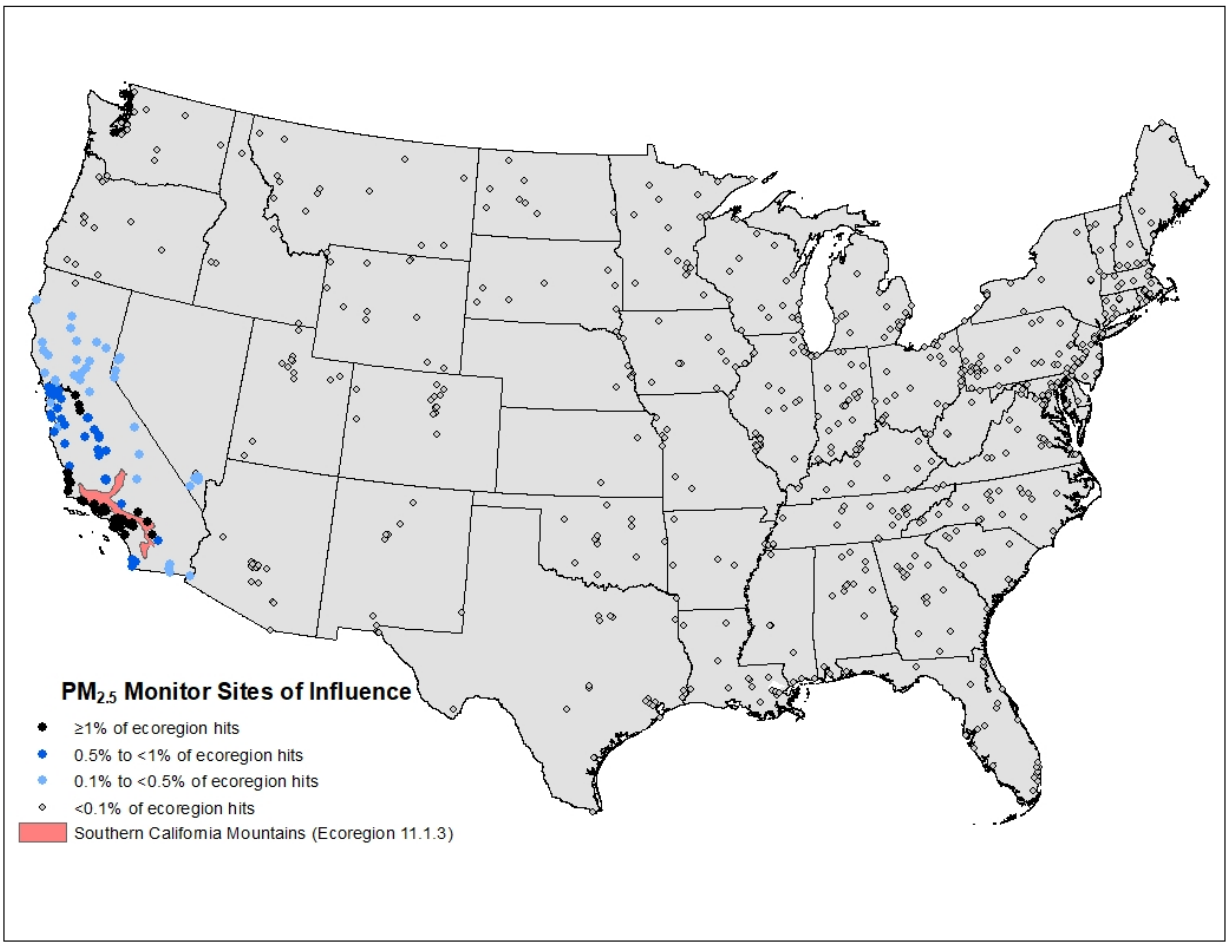

Figure 30. Monitoring sites (annual $PM_{2.5}$ metric) of potential influence for ecoregion 11.1.3 (red shaded region).

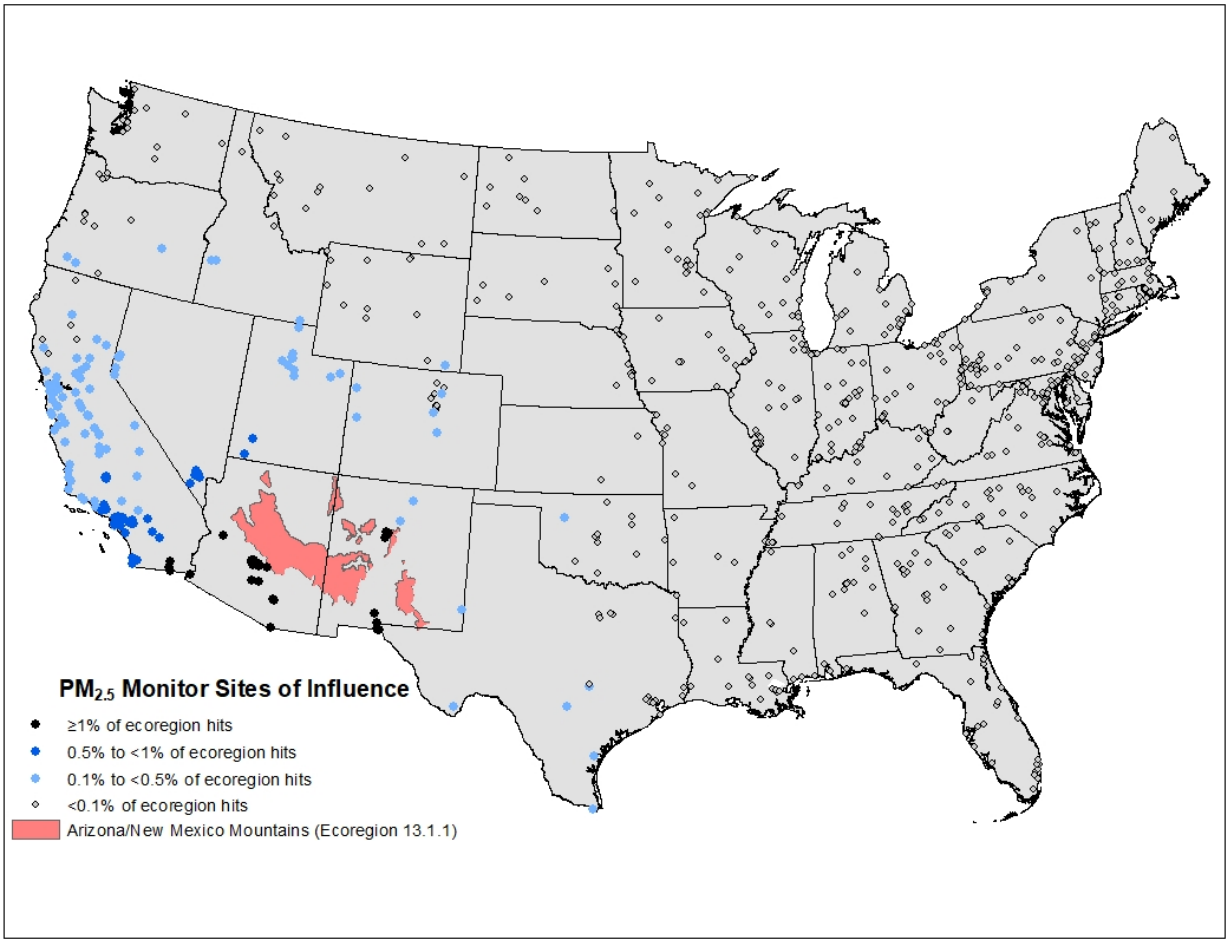

Figure 31. Monitoring sites (annual $PM_{2.5}$ metric) of potential influence for ecoregion 13.1.1 (red shaded region).

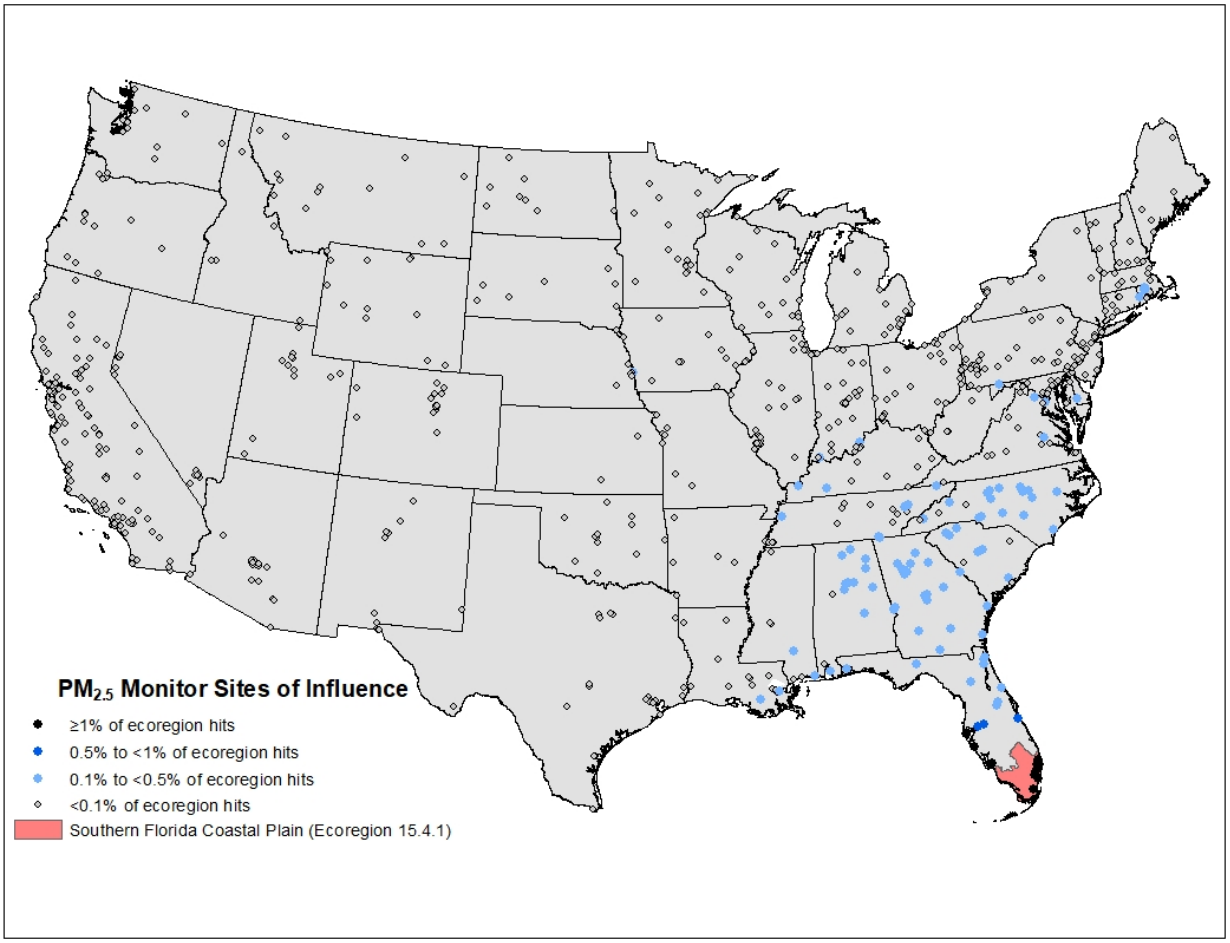

Figure 32. Monitoring sites (annual $PM_{2.5}$ metric) of potential influence for ecoregion 15.4.1 (red shaded region).

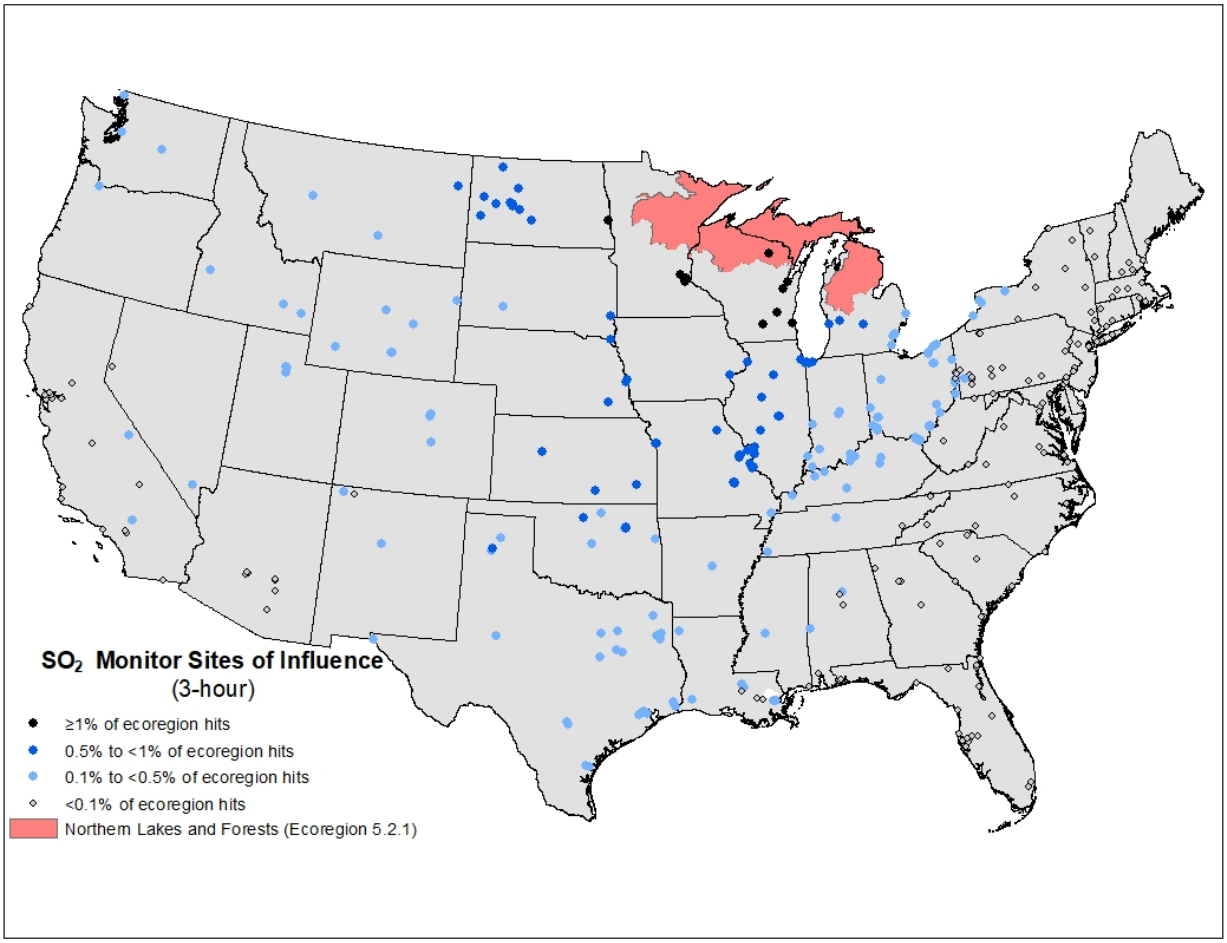

Figure 33. Monitoring sites (3-hour SO_2 metric) of potential influence for ecoregion 5.2.1 (red shaded region).

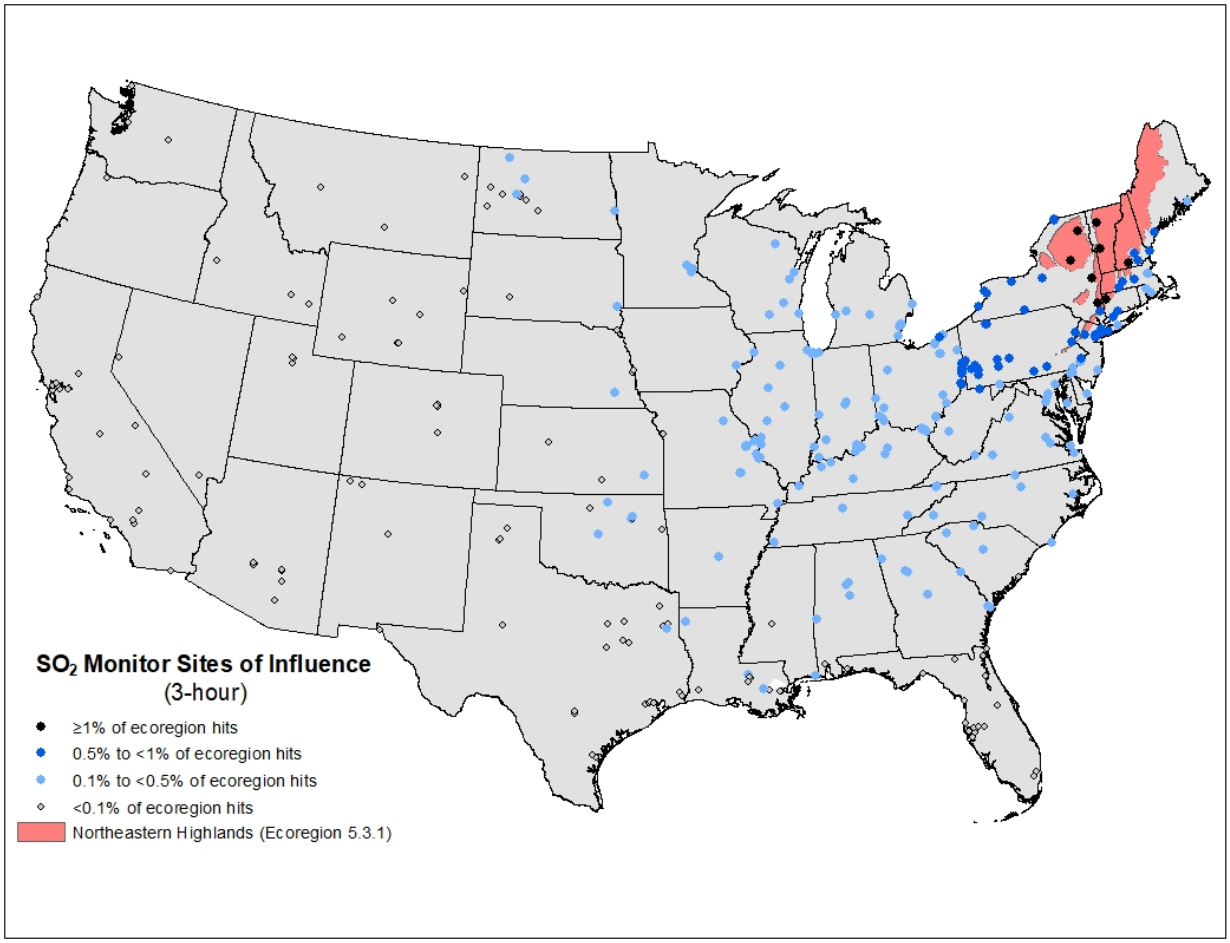

Figure 34. Monitoring sites (3-hour SO_2 metric) of potential influence for ecoregion 5.3.1 (red shaded region).

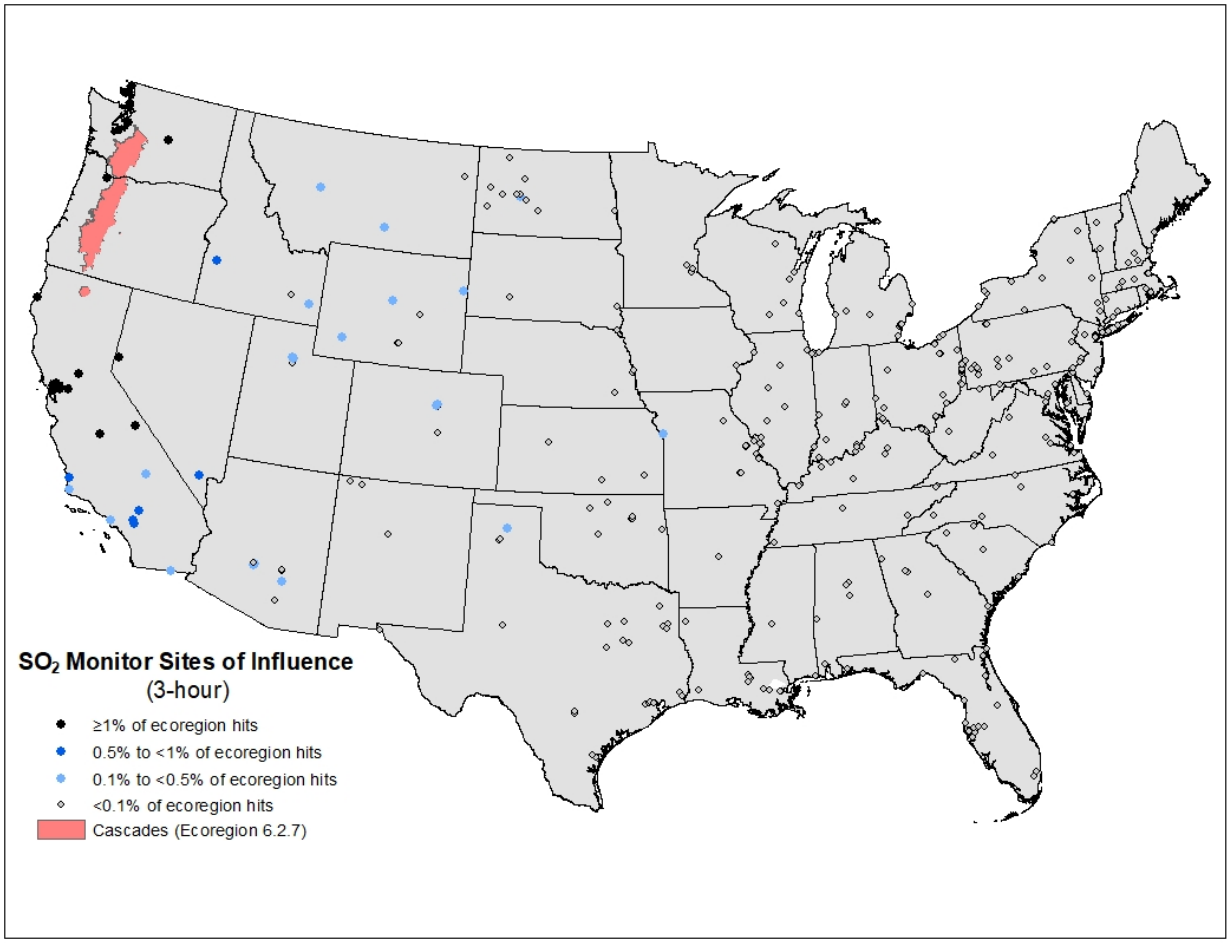

Figure 35. Monitoring sites (3-hour SO_2 metric) of potential influence for ecoregion 6.2.7 (red shaded region).

Figure 36. Monitoring sites (3-hour SO_2 metric) of potential influence for ecoregion 6.2.12 (red shaded region).

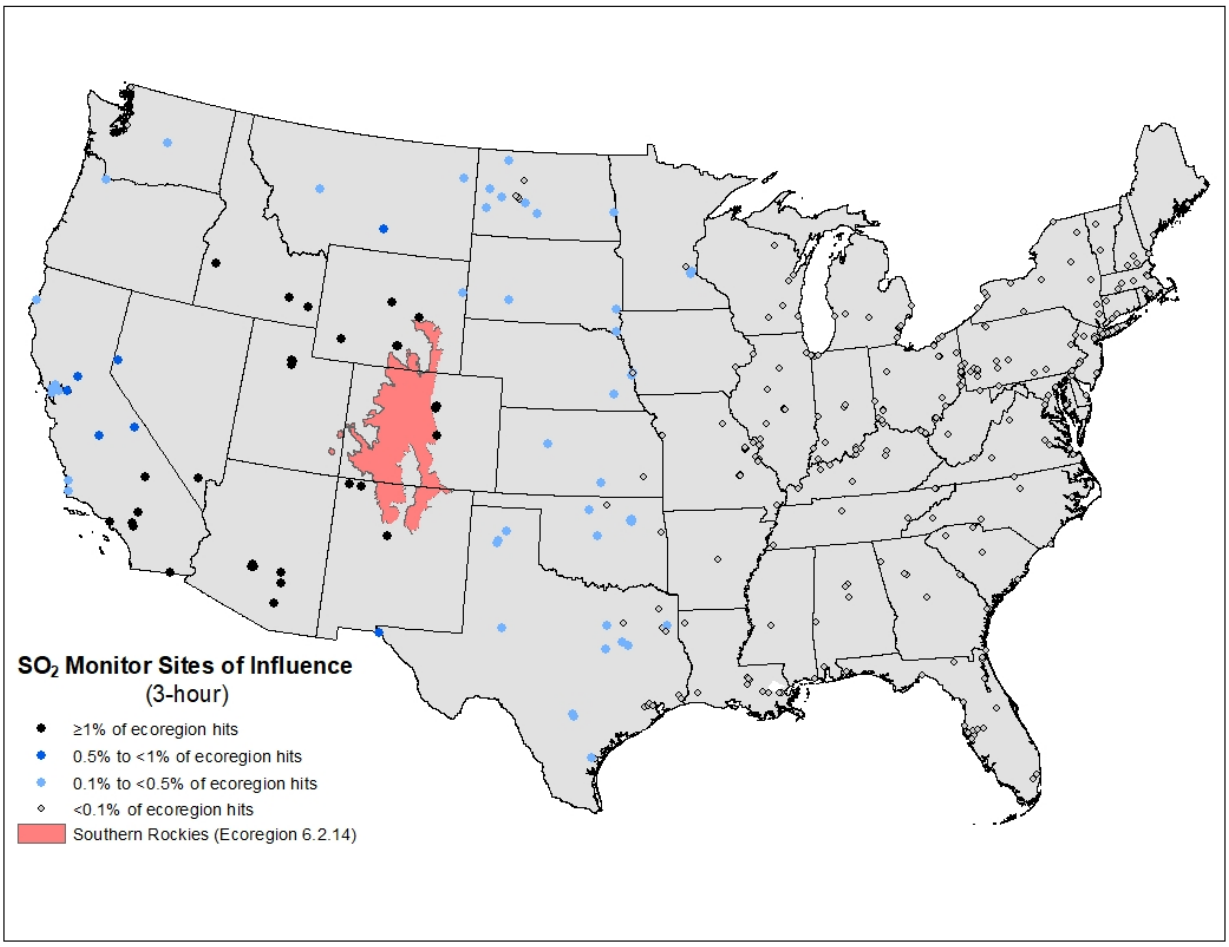

Figure 37. Monitoring sites (3-hour SO_2 metric) of potential influence for ecoregion 6.2.14 (red shaded region).

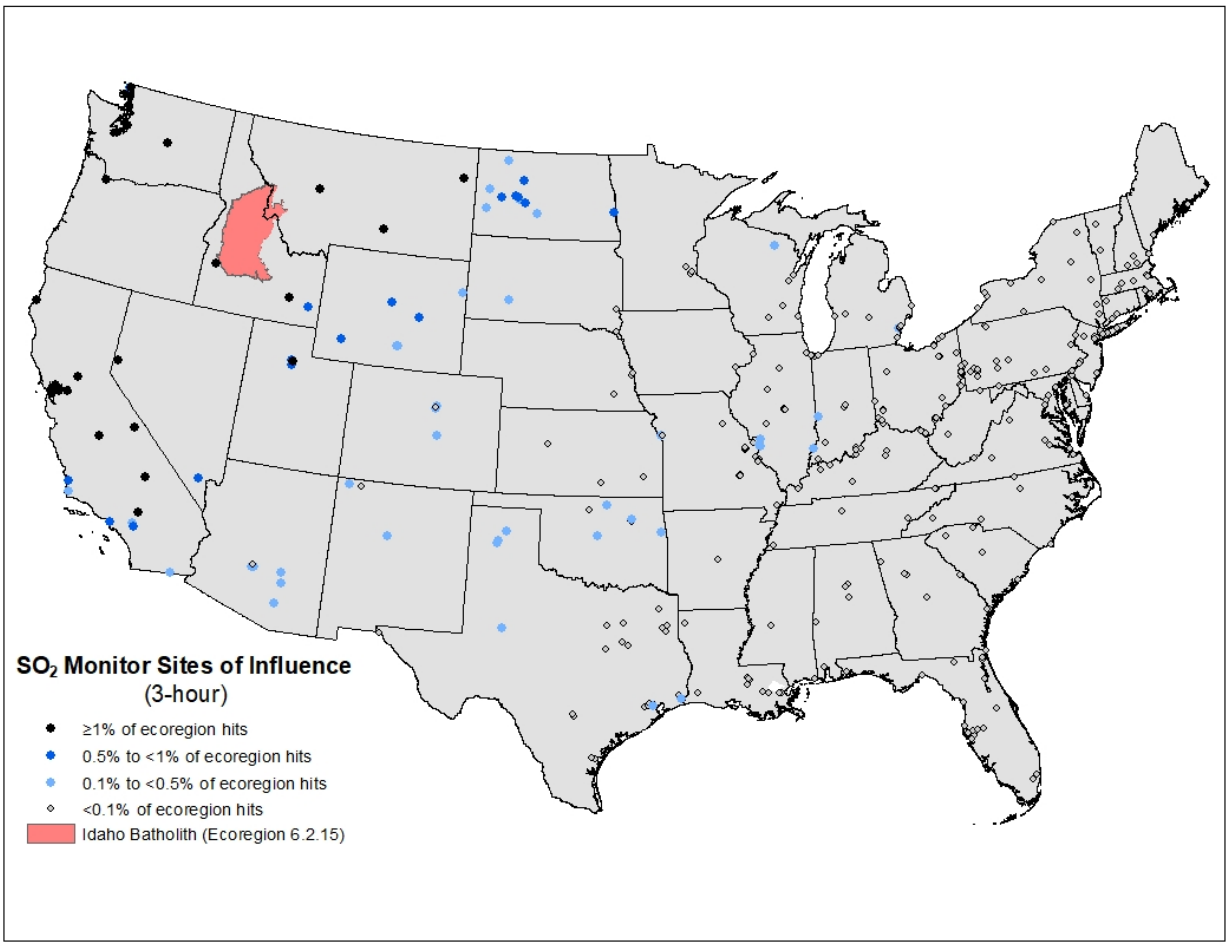

Figure 38. Monitoring sites (3-hour SO_2 metric) of potential influence for ecoregion 6.2.15 (red shaded region).

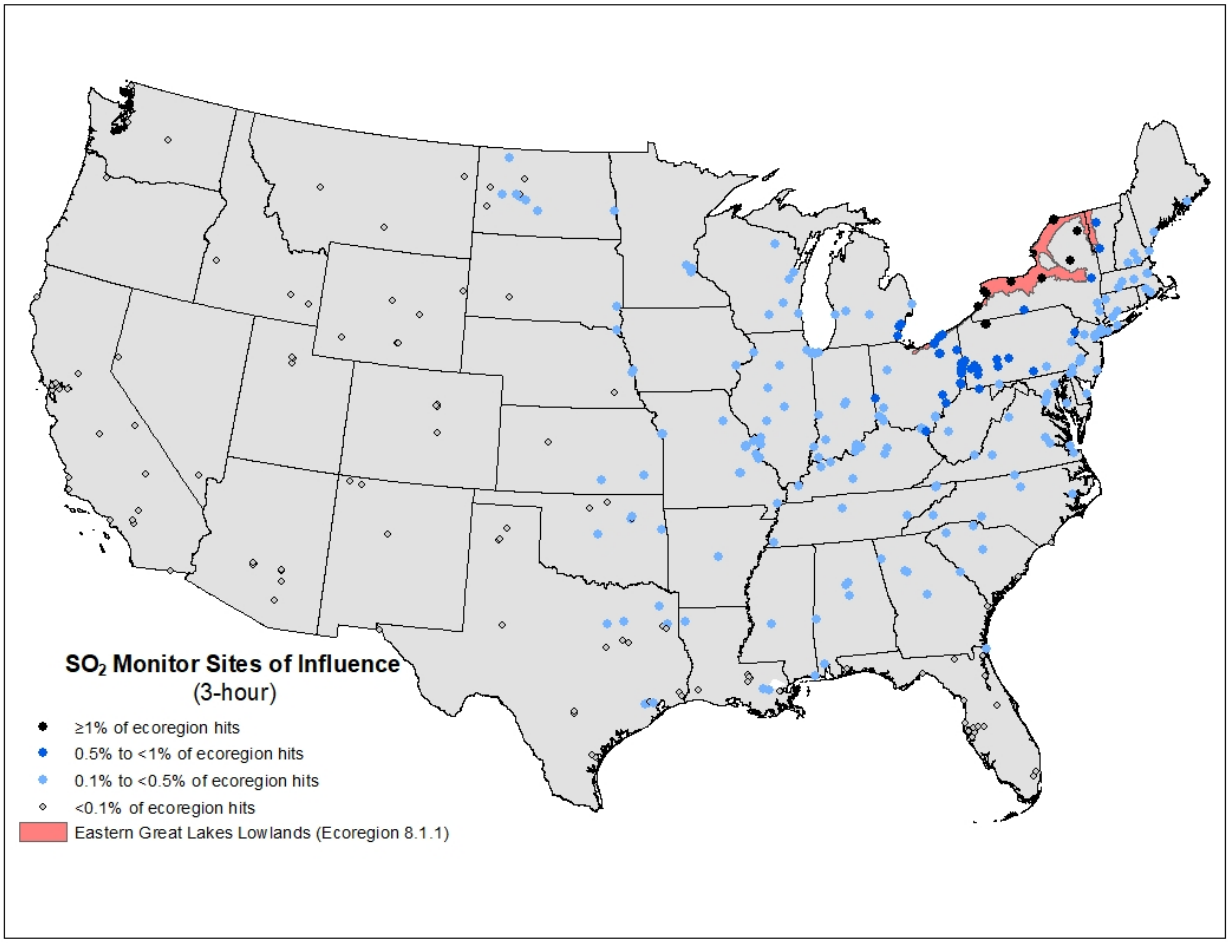

Figure 39. Monitoring sites (3-hour SO_2 metric) of potential influence for ecoregion 8.1.1 (red shaded region).

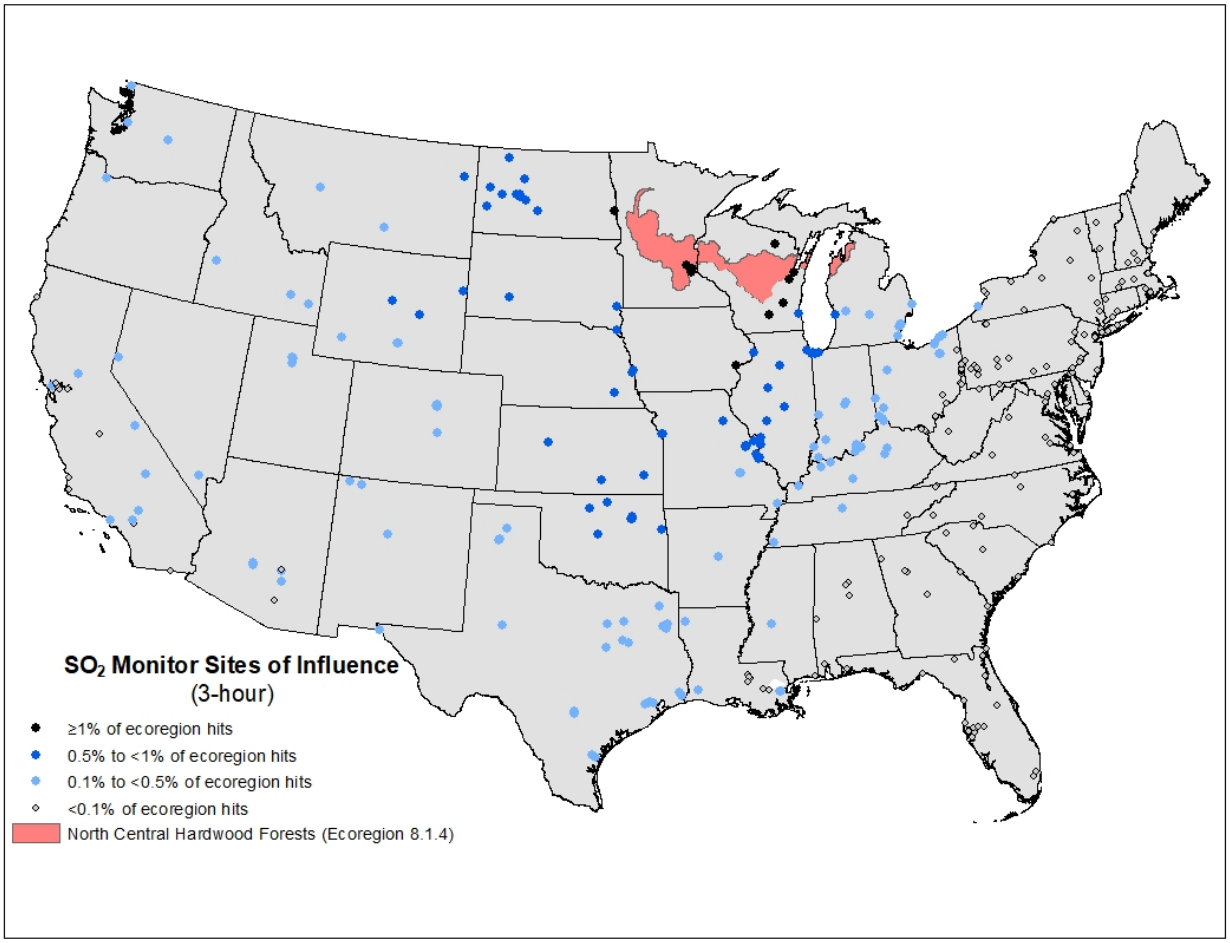

Figure 40. Monitoring sites (3-hour SO_2 metric) of potential influence for ecoregion 8.1.4 (red shaded region).

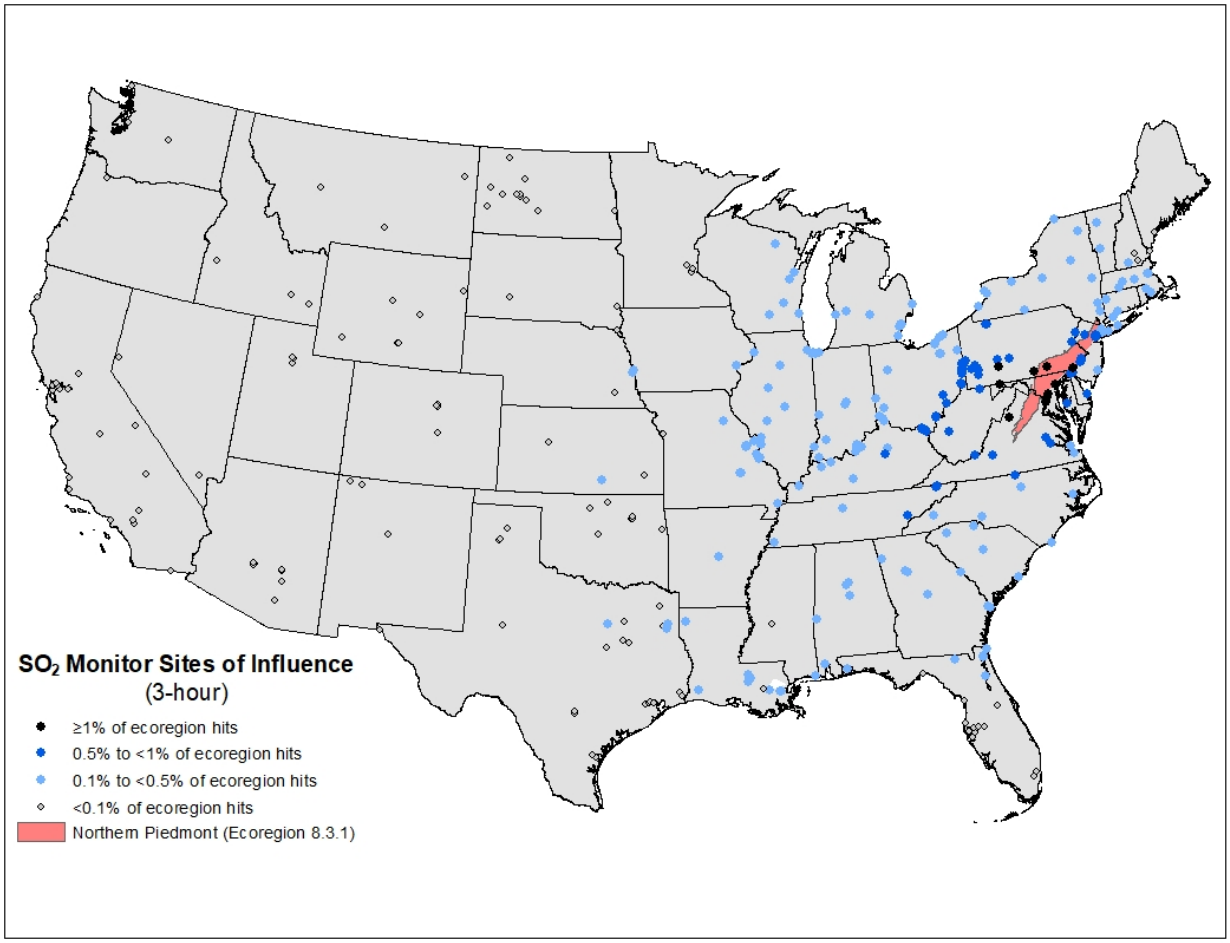

Figure 41. Monitoring sites (3-hour SO_2 metric) of potential influence for ecoregion 8.3.1 (red shaded region).

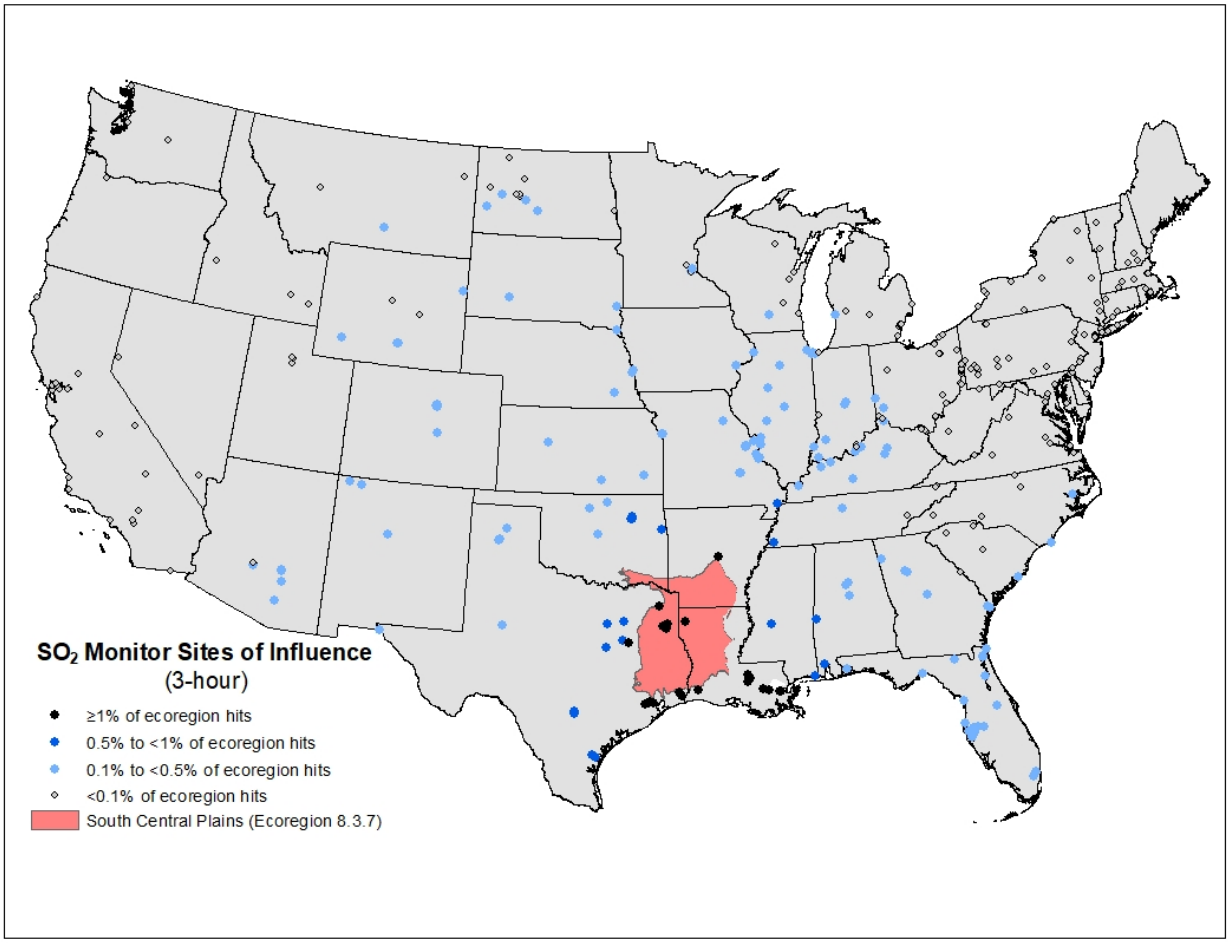

Figure 42. Monitoring sites (3-hour SO_2 metric) of potential influence for ecoregion 8.3.7 (red shaded region).

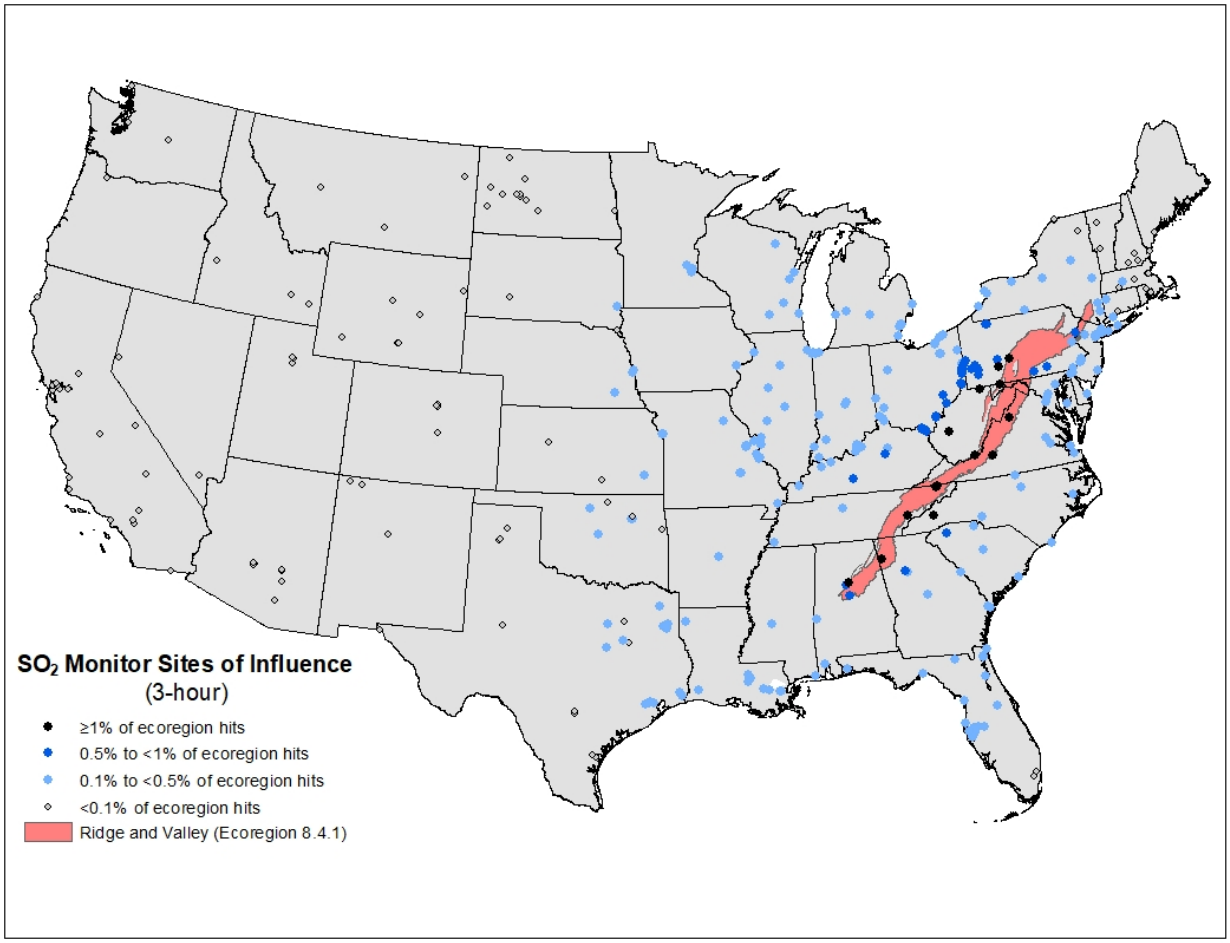

Figure 43. Monitoring sites (3-hour SO_2 metric) of potential influence for ecoregion 8.4.1 (red shaded region).

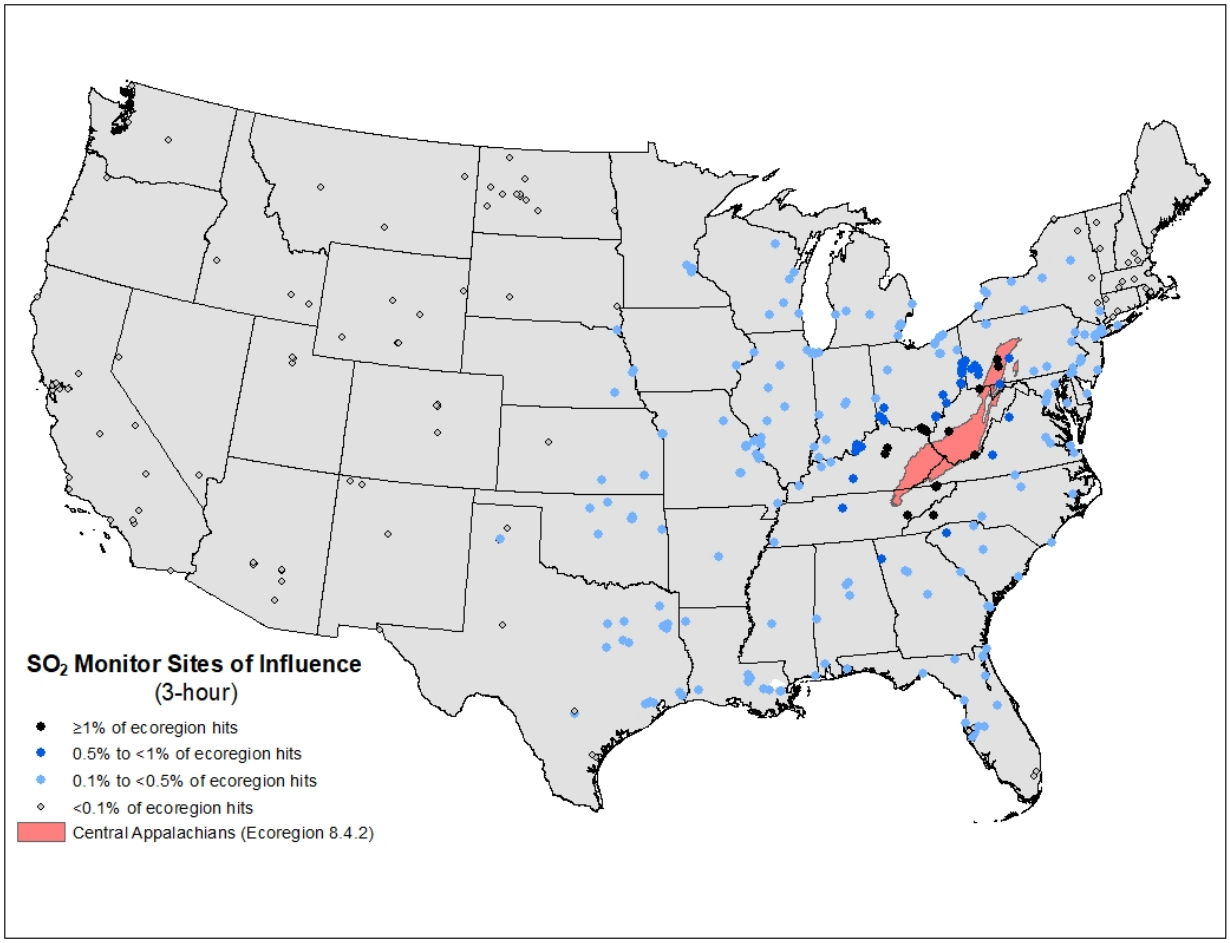

Figure 44. Monitoring sites (3-hour SO_2 metric) of potential influence for ecoregion 8.4.2 (red shaded region).

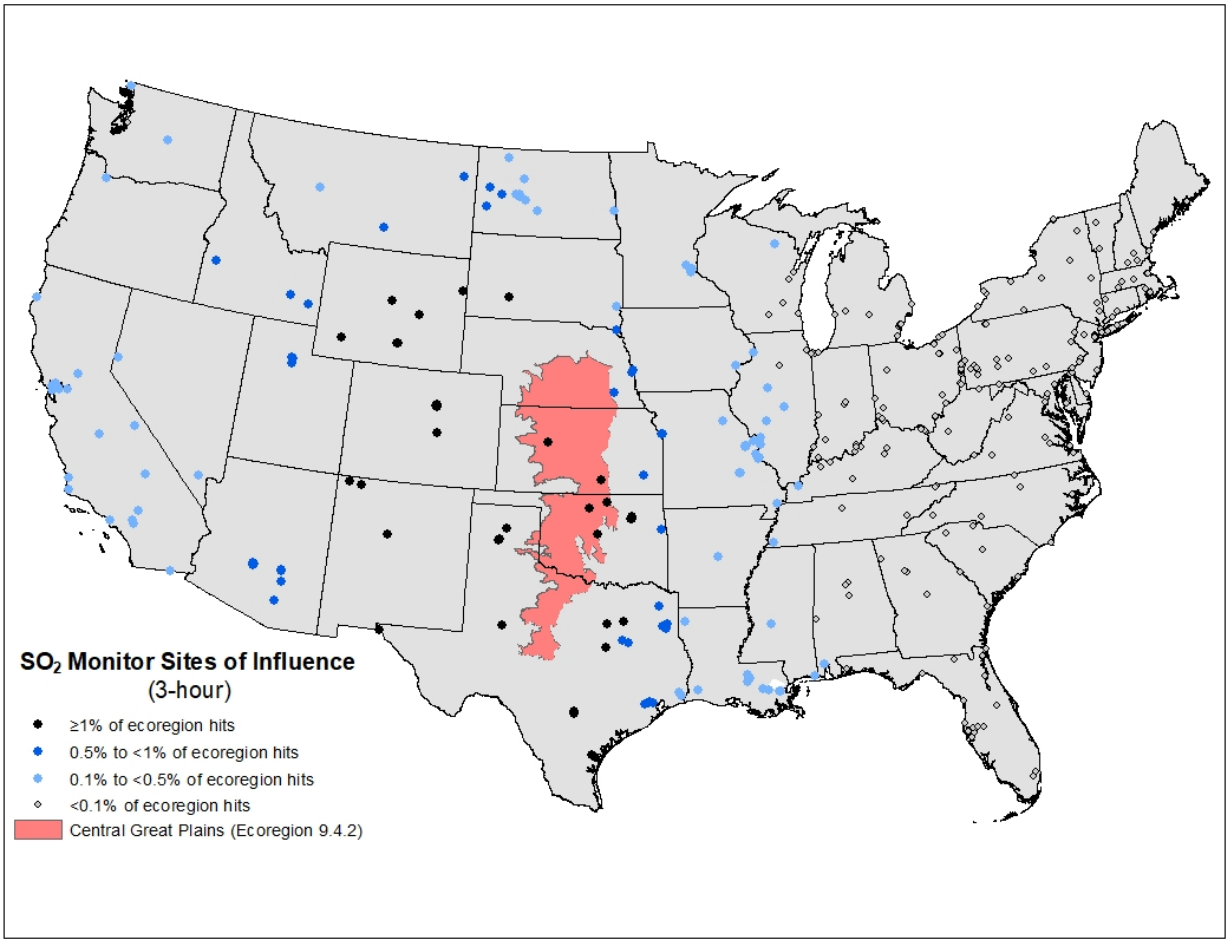

Figure 45. Monitoring sites (3-hour SO_2 metric) of potential influence for ecoregion 9.4.2 (red shaded region).

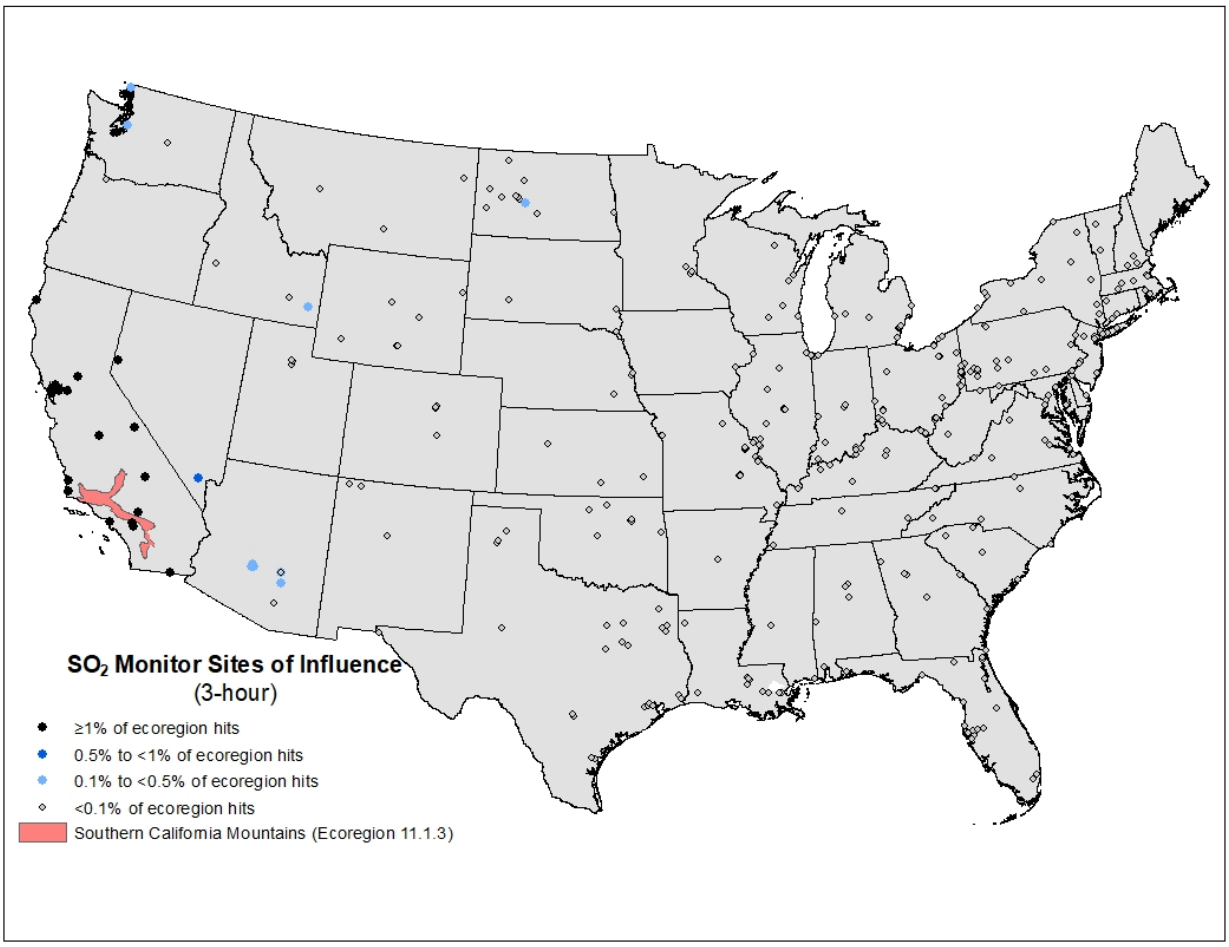

Figure 46. Monitoring sites (3-hour SO_2 metric) of potential influence for ecoregion 11.1.3 (red shaded region).

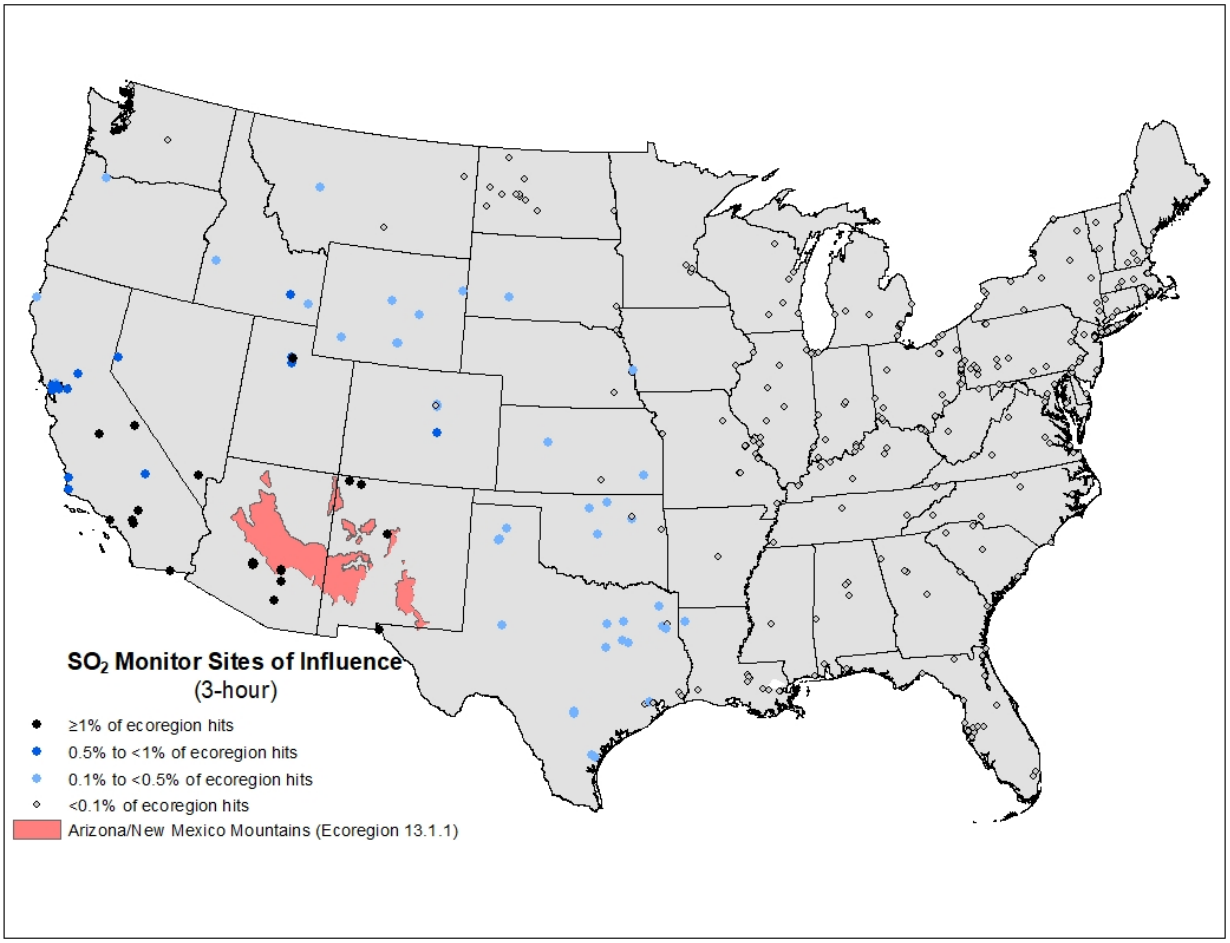

Figure 47. Monitoring sites (3-hour SO_2 metric) of potential influence for ecoregion 13.1.1 (red shaded region).

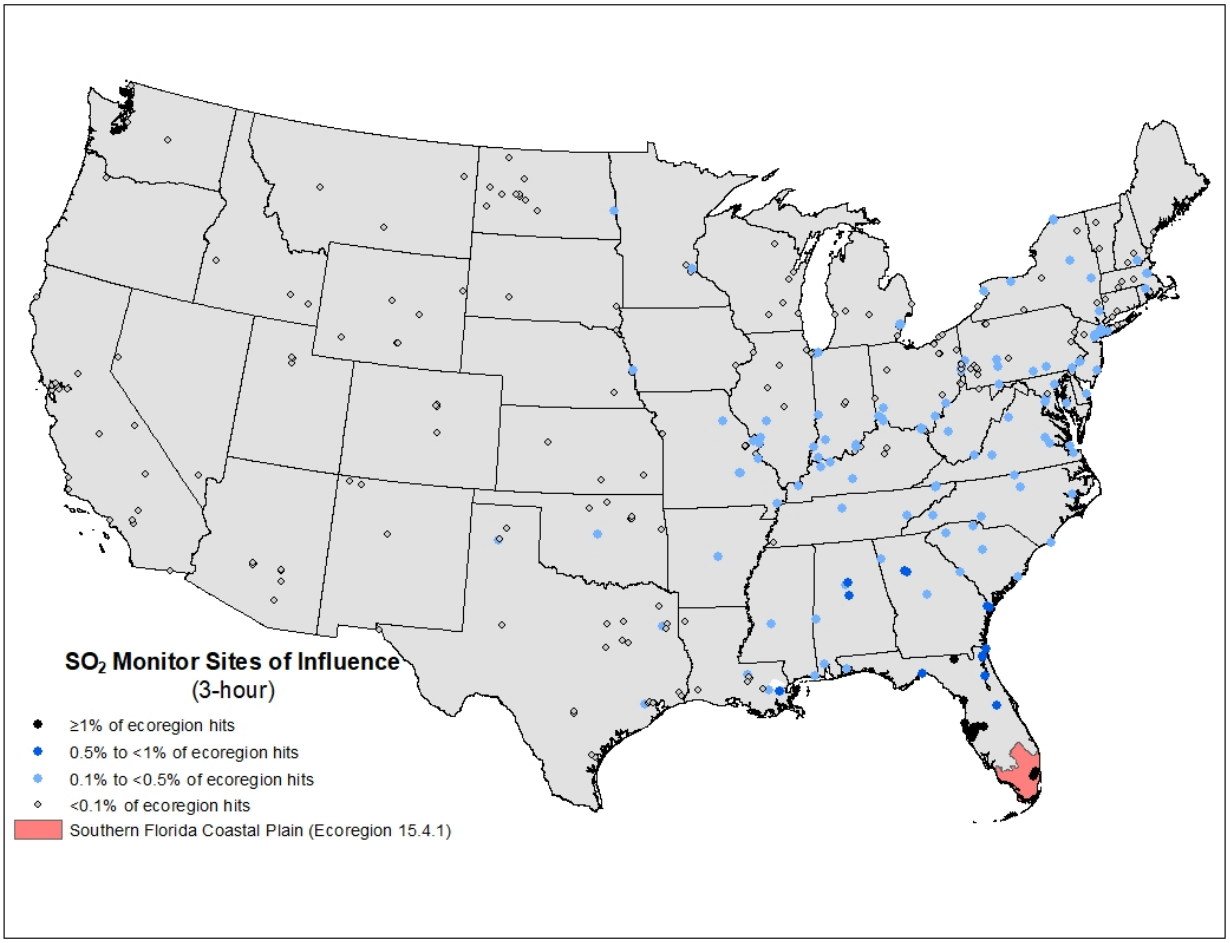

Figure 48. Monitoring sites (3-hour SO_2 metric) of potential influence for ecoregion 15.4.1 (red shaded region).

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