

The EPA Administrator, Michael S. Regan, signed the following notice on 4/3/2024, and EPA is submitting it for publication in the *Federal Register* (FR). While we have taken steps to ensure the accuracy of this Internet version of the rule, it is not the official version of the rule for purposes of compliance. Please refer to the official version in a forthcoming FR publication, which will appear on the Government Printing Office's govinfo website (<https://www.govinfo.gov/app/collection/fr>) and on Regulations.gov (<https://www.regulations.gov>) in Docket No. EPA-HQ-OAR-2014-0128. Once the official version of this document is published in the FR, this version will be removed from the Internet and replaced with a link to the official version.

6560-50-P

ENVIRONMENTAL PROTECTION AGENCY

40 CFR Part 50

[EPA-HQ-OAR-2014-0128; FRL-5788-02-OAR]

RIN 2060-AS35

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

AGENCY: Environmental Protection Agency (EPA).

ACTION: Proposed rule.

SUMMARY: Based on the Environmental Protection Agency's (EPA's) review of the air quality criteria and national ambient air quality standards (NAAQS) for oxides of nitrogen (N oxides), oxides of sulfur (SO_x), and particulate matter (PM), the Environmental Protection Agency (EPA) proposes to revise the existing secondary sulfur dioxide (SO₂) standard to an annual average, averaged over three consecutive years, with a level within the range from 10 to 15 parts per billion (ppb). Additionally, the Agency proposes to retain the existing secondary standards for N oxides and PM, without revision. The EPA also proposes revisions to the data handling requirements for the proposed secondary SO₂ NAAQS.

DATES: Comments must be received on or before **[INSERT DATE 60 DAYS AFTER DATE OF PUBLICATION IN THE FEDERAL REGISTER]**.

Public Hearings: The EPA will hold a virtual public hearing on this proposed rule. This hearing will be announced in a separate *Federal Register* notice that provides details, including specific

dates, times, and contact information for these hearings.

ADDRESSES: You may submit comments, identified by Docket ID No. EPA-HQ-OAR-2014-0128, by any of the following means:

- Federal eRulemaking Portal: <https://www.regulations.gov/> (our preferred method). Follow the online instructions for submitting comments.
- Email: a-and-r-Docket@epa.gov. Include the Docket ID No. EPA-HQ-OAR-2014-0128 in the subject line of the message.
- Mail: U.S. Environmental Protection Agency, EPA Docket Center, Air and Radiation Docket, Mail Code 28221T, 1200 Pennsylvania Avenue NW, Washington, DC 20460.
- Hand Delivery or Courier (by scheduled appointment only): EPA Docket Center, WJC West Building, Room 3334, 1301 Constitution Avenue, NW, Washington, DC 20004. The Docket Center's hours of operations are 8:30 a.m. – 4:30 p.m., Monday – Friday (except Federal Holidays).

Instructions: All submissions received must include the Docket ID No. for this notice.

Comments received may be posted without change to https://www.regulations.gov, including any personal information provided. For detailed instructions on sending comments and additional information on the rulemaking process, see the **SUPPLEMENTARY INFORMATION** section of this document.

FOR FURTHER INFORMATION CONTACT: Ms. Ginger Tennant, Health and Environmental Impacts Division, Office of Air Quality Planning and Standards, U.S. Environmental Protection Agency, Mail Code C539-04, Research Triangle Park, NC 27711; telephone: (919) 541-4072; email: tennant.ginger@epa.gov.

SUPPLEMENTARY INFORMATION:

General Information

Preparing Comments for the EPA

Follow the online instructions for submitting comments. Once submitted to the Federal eRulemaking Portal, comments cannot be edited or withdrawn. The EPA may publish any comment received to its public docket. Do not submit electronically any information you consider to be Confidential Business Information (CBI) or other information whose disclosure is restricted by statute. Multimedia submissions (audio, video, etc.) must be accompanied by a written submission. The written comment is considered the official comment and should include discussion of all points you wish to make. The EPA will generally not consider comments or comment contents located outside of the primary submission (i.e., on the web, the cloud, or other file sharing system). For additional submission methods, the full EPA public comment policy, information about CBI or multimedia submissions, and general guidance on making effective comments, please visit <https://www.epa.gov/dockets/commenting-epa-dockets>.

When submitting comments, remember to:

- Identify the action by docket number and other identifying information (subject heading, *Federal Register* date and page number).
- Explain why you agree or disagree, suggest alternatives, and substitute language for your requested changes.
- Describe any assumptions and provide any technical information and/or data that you used.
- Provide specific examples to illustrate your concerns and suggest alternatives.
- Explain your views as clearly as possible, avoiding the use of profanity or personal threats.

- Make sure to submit your comments by the comment period deadline identified.

Availability of Information Related to this Action

All documents in the dockets pertaining to this action are listed on the www.regulations.gov website. This includes documents in the docket for the proposed decision (Docket ID No. EPA-HQ-OAR-2014-0128) and a separate docket, established for the Integrated Science Assessment (ISA) (Docket ID No. EPA-HQ-ORD-2013-0620) that has been incorporated by reference into the docket for this proposed decision. Although listed in the index, some information is not publicly available, e.g., CBI or other information whose disclosure is restricted by statute. Certain other material, such as copyrighted material, is not placed on the internet and may be viewed with prior arrangement with the EPA Docket Center. Additionally, a number of the documents that are relevant to this proposed decision are available through the EPA's website at <https://www.epa.gov/naaqs/>. These documents include the Integrated Science Assessment for Oxides of Nitrogen, Oxides of Sulfur and Particulate Matter Ecological Criteria (U.S. EPA, 2020a), available at <https://cfpub.epa.gov/ncea/isa/recordisplay.cfm?deid=349473>, and the Policy Assessment for the Review of the Secondary National Ambient Air Quality Standards for Oxides of Nitrogen, Oxides of Sulfur, and Particulate Matter, (U.S. EPA, 2024), available at <https://www.epa.gov/naaqs/nitrogen-dioxide-no2-and-sulfur-dioxide-so2-secondary-air-quality-standards>.

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Executive Summary

This document presents the Administrator's proposed decisions in the current review of the secondary NAAQS for SO_x, N oxides, and PM. The existing secondary standards are: for SO₂, 0.5 ppm as a 3-hour average not to be exceeded more than once in a year; for NO₂, 53 ppb as an annual average; for PM_{2.5}, 15.0 µg/m³ as the 3-year average of annual averages, and 35 µg/m³ as the 3-year average of annual 98th percentile 24-hour averages; and, for PM₁₀, 150 µg/m³ as a 24-hour average, not to be exceeded more than once per year on average over three years. Sections 108 and 109 of the Clean Air Act (CAA, the Act) require the EPA to periodically

review the air quality criteria—the science upon which the standards are based—and the standards themselves. Under section 109(b)(2) of the Act, 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.” In conducting this review of the secondary SO_x, N oxides, and PM NAAQS, the EPA has carefully evaluated the currently available scientific literature on the ecological effects of SO_x, N oxides, and PM,¹ focusing particularly on the new literature available since the conclusion of the previous reviews in 2012 and 2013, respectively, as described in the Integrated Science Assessment (ISA). The ecological effects addressed in this review include direct effects of N oxides and SO_x, and PM loading, on vegetation surfaces, as well as ecological effects related to atmospheric deposition of S and N compounds in sensitive ecosystems.

Sulfur oxides and N oxides, their transformation products (which include particulate compounds), and N- and S-containing components of PM in ambient air can contribute to atmospheric deposition of S and N compounds. Emissions of SO_x, N oxides, PM and PM precursors have declined dramatically over the past two decades, continuing a longer-term trend. In response to the reductions in S- and N-containing compounds, levels of S and N deposition have also been reduced, although the declining trend in N deposition in the last decade has slowed and, in some areas, reversed, due to increasing ammonia emissions.

¹ The ecological effects of PM that are the focus of this action were not considered in EPA’s recently completed reconsideration of the primary and secondary NAAQS for PM. In the review of the PM secondary standards completed in 2020, and reconsidered more recently, the EPA considered effects on visibility and climate and materials damage, but did not consider the ecological effects that are addressed here (89 FR 16202, March 6, 2024).

The Administrator's proposed decision in this review is to revise the existing secondary SO₂ standard and to retain the existing secondary standards for N oxides and PM. In this document, the EPA summarizes the background and rationale for the Administrator's proposed decisions in this review. The EPA solicits comment on the proposed decisions described here and on a number of alternate options, and requests commenters also provide the rationales supporting the views articulated in submitted comments.

The Administrator's proposed decisions are based on his consideration of the characterization of the available scientific evidence in the ISA, quantitative and policy analyses presented in the Policy Assessment (PA), and advice from the Clean Air Scientific Advisory Committee (CASAC).² In conveying its advice in this review, the CASAC provided consensus advice that the existing SO₂ and NO₂ secondary standards were adequate to protect against direct effects of S and N oxides on plants and lichens. With regard to deposition-related effects and SO₂, the majority of CASAC recommended an annual secondary standard of 10-15 ppb, and the minority recommended a secondary standard identical to the existing primary standard. In consideration of deposition-related effects and the NO₂ and PM_{2.5} secondary standards, the CASAC majority recommended revision of the levels of the existing annual NO₂ and PM_{2.5} secondary standards, and the minority recommended adopting secondary standards identical to the existing annual NO₂ and PM_{2.5} primary standards.

Based on his consideration of the ecological effects evidence in the ISA, the evaluations

² Over the course of this review, the EPA developed planning documents, an ISA and a PA, drafts of which were made available for public comment and reviewed by the CASAC Oxides of Nitrogen, Oxides of Sulfide and Particulate Matter Secondary NAAQS Panel (<https://www.epa.gov/naaqs/nitrogen-dioxide-no2-and-sulfur-dioxide-so2-secondary-air-quality-standards>).

and quantitative information in the PA, including the quantitative REA for aquatic acidification, and advice from the CASAC, the Administrator is proposing that the current short-term secondary SO₂ standard is not requisite to protect the public welfare from known or anticipated adverse effects associated with the presence of SO_x in ambient air, including particularly deposition-related effects, and that it should be revised to also provide such protection against effects related to deposition of sulfur (S) compounds to ecosystems. Specifically, the EPA is proposing to revise the existing standard to be an annual average standard, averaged over three years, with a level within the range from 10 to 15 parts per billion (ppb) based on the Administrator's proposed judgment that a standard in this range would provide protection for both direct effects on vegetation surfaces and ecosystem deposition-related effects. The EPA solicits comments on this proposal, including the averaging time, form and range of levels for the revised standard. The EPA also solicits comments on a number of alternative options for a new secondary SO₂ standard. The EPA solicits comment on setting the level for a new annual average standard (averaged over three years) in the range from 5 to 10 ppb, and on revising the existing secondary standard to be identical to the existing primary standard in all respects. Further, the EPA solicits comments on retaining the existing 3-hour standard, in addition to establishing a new annual secondary standard.

The Administrator is also proposing to retain the secondary nitrogen dioxide (NO₂) and PM standards, without revision. With regard to the secondary NO₂ standard, the Administrator finds that the evidence related to the N oxides, NO₂ and nitrogen oxide (NO), does not clearly call into question the adequacy of protection provided by the existing standard for direct effects or for deposition-related effects (in light of the currently diminished role of N oxides in total N deposition, particularly in areas with highest deposition), such that revision is not warranted. The

EPA solicits comment on the proposed decision to retain the existing secondary NO₂ standard, without revision, and also on the alternative approach of revising the form of the existing standard to a 3-year average and the level to a value within the range from 35 to 40 ppb.

With regard to PM, the Administrator proposes to conclude that the current evidence does not call into question the adequacy of the existing PM standards with regard to direct effects and deposition of pollutants other than S and N compounds. Further, he judges that protection of sensitive ecosystems from S deposition is more effectively achieved through a revised SO₂ standard than a PM standard, and that a revised PM standard is not warranted to provide public welfare protection against adverse effects related to S or N deposition. The Administrator additionally proposes to conclude that PM_{2.5} is not an appropriate indicator for a secondary standard intended to provide protection of the public welfare from adverse effects related to S or N deposition. Thus, based on consideration of the PA analyses and conclusions, and consideration of CASAC advice, the Administrator proposes to conclude that no change to the annual PM_{2.5} secondary standard is warranted and he proposes to retain the existing PM_{2.5} secondary standard, without revision. The EPA solicits comment on the proposed decision. Additionally, in recognizing that there may be alternate views with regard to whether and to what extent a standard with a PM_{2.5} indicator might be expected to provide control of N deposition, and in light of the rationale provided by the CASAC minority, the EPA also solicits comment on the alternative approach of revising the secondary PM_{2.5} (with PM_{2.5} referring to particles with a nominal mean aerodynamic diameter less than or equal to 2.5 micrometers) annual standard to a level of 12 micrograms per meter cubed (µg/m³). With regard to other PM secondary standards, based on evaluations and conclusions of the PA, including consideration of recommendations from the CASAC, the Administrator proposes to retain the existing 24-hour secondary PM_{2.5}

standard, without revision. Further, based on the lack of evidence calling into question the adequacy of the secondary PM₁₀ standards for protection of ecological effects, he also proposes to retain the secondary PM₁₀ standards without revision.

This document additionally includes proposed revisions related to implementation of the proposed secondary SO₂ annual standard. Specifically, the EPA is proposing revisions to the data handling requirements in Appendix T of 40 CFR part 50 to include specifications needed for the proposed new annual average standard. This document also describes the SO₂ monitoring network and its adequacy for surveillance for the proposed annual standard. Lastly, the document discusses implementation processes pertinent to implementation of the proposed new standard.

I. Background

A. 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 of the Act (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.”³

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.⁴

³ 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.”

⁴ This section of the Act requires the Administrator to complete these reviews and make any revisions that may be appropriate “at five-year intervals.”

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 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 ($\mu\text{g}/\text{m}^3$). 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.

B. Related Control Programs

States are primarily responsible for ensuring attainment and maintenance of ambient air quality standards once the EPA has established them. Under CAA sections 110 and Part D, Subparts 1, 5, and 6 for nitrogen and sulfur oxides, and Subparts 1, 4, and 6 for PM, and related provisions and regulations, States are to submit, for the EPA's approval, State implementation plans (SIPs) that provide for the attainment and maintenance of such standards through control programs directed to sources of the pollutants involved. The States, in conjunction with the EPA, also administer the prevention of significant deterioration of air quality program that covers these pollutants. *See* 42 U.S.C. 7470-7479. In addition, Federal programs provide for or result in nationwide reductions in emissions of N oxides, SO_x, PM and other air pollutants under Title II of the Act, 42 U.S.C. 7521-7574, which involves controls for motor vehicles, nonroad engines and equipment, and the new source performance standards under section 111 of the Act, 42 U.S.C. 7411.

C. History of the Secondary Standards for N Oxides, SO_x and PM

Secondary NAAQS were first established for N oxides, SO_x and PM 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 2012-2013.⁵

1. N Oxides

⁵ Since the late 1970s, each review of the air quality criteria and standards has generally involved the development of an Air Quality Criteria Document or ISA and a Staff Paper or staff Policy Assessment, which is often accompanied by or includes a quantitative exposure or risk assessment, prior to the regulatory decision-making phase.

The EPA first promulgated NAAQS for N oxides 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).⁶ With regard to welfare effects, the 1971 AQCD described effects of NO₂ 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 parts per million (ppm) NO₂ 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).

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,⁷ drafts of which were reviewed by the CASAC (Wolff, 1995;

⁶ 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 ISA has replaced the AQCD.

⁷ Prior to reviews initiated in 2007, the Staff Paper summarized and integrated key studies and the scientific evidence, and from the 1990s onward, it 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.

U.S. EPA, 1995a). In October 1995, the EPA proposed not to revise the secondary NO₂ NAAQS (60 FR 52874; October 11, 1995). After consideration of the comments received on the proposal, the Administrator finalized the decision not to revise the NO₂ 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 subsection 4 below).

2. SO_x

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 SO₂,⁸ and a 3-hour average standard set at 0.5 ppm SO₂, 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 with SO₂ 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 SO₂ welfare effect for which the

⁸ 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).

evidence base supported a quantitative relationship, stating that although data were not available at that time to establish a quantitative relationship between SO₂ concentrations and other public welfare effects, including effects on materials, visibility, soils, and water, the SO₂ 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, were 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 in guidance provided at the August 20-22, 1980, public meeting of the CASAC 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 -36, 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 SO₂ (53 FR 14926, April 26, 1988). This proposed decision with regard to the secondary SO₂ 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 I.C.3). The EPA decided not to revise the secondary standard, concluding that revisions to the standard to address acidic deposition and related SO₂ welfare effects were not appropriate at that time (58 FR 21351, April 21, 1993). In describing the

decision, the EPA recognized the significant reductions in SO₂ emissions, ambient air SO₂ 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).

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, the program issued a series of reports, including a final report 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 SO₂ 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 Sand N 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 CAA 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 SO₂ and implemented an enhanced monitoring program to track progress (58 FR 21357, April 21, 1993).

4. Most Recent Review of the Secondary Standards for N Oxides and SO_x

In December 2005, the EPA initiated a joint review⁹ of the air quality criteria for oxides of nitrogen and sulfur and the secondary NAAQS for NO₂ and SO₂ (70 FR 73236, December 9, 2005).¹⁰ 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 NO₂ and SO₂ 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).

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 (referred to as 2008 ISA below) was released in December 2008 (73 FR 75716, December 12, 2008; U.S. EPA, 2008a). Based on the scientific information in the ISA, the EPA planned and developed a

⁹ 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).

¹⁰ 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.

quantitative Risk and Exposure Assessment (REA),¹¹ 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 2011 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 2011 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 2011 PA concluded that consideration should be given to a new multipollutant

¹¹ Although the REA for the 2012 review was presented in its own separate document, the REA for a NAAQS review may be presented in its own separate document or as one or more appendices in the PA (e.g., U.S. EPA 2020b, 2020c, and PA for current review [U.S. EPA, 2024]).

standard intended to address deposition-related effects.

On August 1, 2011, the EPA published a proposed decision to retain the existing annual average NO₂ and 3-hour average SO₂ 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 establishing secondary standards that was described in the 2011 PA, the Administrator proposed not to set such a new multipollutant secondary standard in light of a number of uncertainties. Additionally, the Administrator proposed to revise the secondary standards by adding secondary NO₂ and SO₂ standards identical to the 1-hour primary NO₂ and SO₂ standards, both set in 2010, noting that these new primary standards¹² 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 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).

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.

¹² The 1-hour primary standards set in 2010 included the NO₂ standard of 100 ppb, as the 98th percentile of 1-hour daily maximum concentrations, averaged over three years, and the SO₂ standard of 75 ppb, as the 99th percentile of 1-hour daily maximum concentrations, averaged over three years (75 FR 6474, February 9, 2010; 75 FR 35520, June 22, 2010).

The District of Columbia Circuit (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 . . . ’.”¹³ 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.¹⁴

5. PM

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 $\mu\text{g}/\text{m}^3$, 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).¹⁵

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 subsection 2 above, the 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 review (Lippman, 1986; U.S. EPA, 1986). After consideration of public comments on a proposed

¹³ *Center for Biological Diversity, et al. v. EPA*, 749 F.3d 1079, 1087 (2014).

¹⁴ *Id.* at 1088.

¹⁵ Additionally, a guide to be used in assessing implementation plans to achieve the 24-hour standard was set at 60 $\mu\text{g}/\text{m}^3$, as an annual geometric mean (36 FR 8187; April 30, 1971).

decision, the final decision in that review revised the indicator for PM NAAQS from TSP to particulate matter with mass median diameter of 10 microns (PM₁₀) (49 FR 10408, March 20, 1984; 52 FR 24634, July 1, 1987). With an indicator of PM₁₀, two secondary standards were established to be the same as the primary standards. A 24-hour secondary standard was set at 150 µg/m³, with the form of one expected exceedance per year, on average over three years. Additionally, an annual secondary standard was set at 50 µg/m³, 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 for public and 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 final 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. 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 µg/m³, based on the 3-year average of annual arithmetic mean PM_{2.5} concentrations from single or multiple community-oriented monitors;¹⁶ and (2) a 24-hour standard with a level of 65

¹⁶ 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).

$\mu\text{g}/\text{m}^3$, based on the 3-year average of the 98th percentile of 24-hour $\text{PM}_{2.5}$ 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, several parties filed petitions for review, raising a broad range of issues. In May 1999, the U.S. Court of Appeals for the D.C. Circuit upheld the EPA's decision to establish fine particle standards, (*American Trucking Ass'n, 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 (*Id* at 1054-55). Pursuant to the D.C. Circuit's decision, the EPA removed the vacated 1997 PM_{10} standards, leaving the pre-existing 1987 PM_{10} standards 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 (*Id* 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.

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, b). 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 (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 µg/m³, retained the level of the annual PM_{2.5} standards at 15.0 µg/m³, 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 PM₁₀, the EPA retained the 24-hour standards, with levels at 150 µg/m³, and revoked the annual standards.

Several parties filed petitions for review of the EPA's 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 *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 are part of 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 PM₁₀ 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. On judicial review, the revised standards and monitoring requirements were upheld in all respects (*National Association of Manufacturers v. EPA*, 750 F.3d 921, [D.C. Cir. 2014]).

The subsequent review of the PM secondary standards, completed in 2020, and its subsequent reconsideration, focused on consideration of protection provided from visibility effects, materials damage, and climate effects (85 FR 82684, December 18, 2020; 89 FR 16202, March 6, 2024). Those effects – visibility effects, materials damage and climate effects – are not addressed in this review. The evidence for ecological effects of PM is addressed in the review of the air quality criteria and standards described in the PA for this review.

D. 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 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 atmospheric transformation of N and S oxides into particulate compounds, deposition of N and S compounds and associated ecological effects (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 the CASAC reviewed 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).¹⁷ The EPA released the final ISA in October 2020 (85 FR 66327, October 19, 2020; U.S. EPA, 2020a). 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

¹⁷ A change in CASAC membership contributed to an extended time period between the two public meetings.

FR 31755, July 9, 2018; Cox, Kendall, and Fernandez, 2020b; U.S. EPA, 2018; 83 FR 42497, August 22, 2018).

The draft PA, including the REA for aquatic acidification as an appendix, was completed in May 2023 and was 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 final PA was completed in January 2024 (89 FR 2223, January 12, 2024).

Materials upon which this proposed decision is based, including the documents described above, are available to the public in the docket for this review.¹⁸ 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* [No. 4:22-cv-02285-HSG (N.D. Cal.)]).

II. Rationale for Proposed Decisions

This section presents the rationale for the Administrator's proposed decisions in the review of the secondary standards for the ecological effects of SO_x, N oxides and PM. This rationale is based on a thorough review of the full evidence base, including the scientific information available since the last review of the secondary standards for N oxides and SO_x, which is generally published between January 2008 and May 2017 (and considered in the ISA), as well as more recent studies identified during peer review or by public comments (ISA, section

¹⁸ The docket for this review, EPA-HQ-OAR-2014-0128, has incorporated the ISA docket (EPA-HQ-ORD-2013-0620) by reference. Both are publicly accessible at www.regulations.gov.

IS.1.2),¹⁹ integrated with the information and conclusions from previous assessments and presented in the ISA, on ecological effects associated with SO_x, N oxides and PM and pertaining to their presence in ambient air. The Administrator's rationale also takes into account: (1) the PA evaluation of the policy-relevant information in the ISA and presentation of quantitative analyses of air quality, exposure and aquatic acidification risks; (2) CASAC advice and recommendations, as reflected in discussions of drafts of the ISA and PA at public meetings and in the CASAC's letters to the Administrator; and (3) public comments received during the development of these documents.

In presenting the rationale for the Administrator's proposed decisions and their foundations, section II.A provides background on the general approach in this review, including a summary of the basis for the existing standards (section II.A.1), a summary of the prior review of the SO_x and N oxides standards for deposition-related effects (section II.A.2) and the general approach for the current review (section II.A.3).

Section II.B summarizes air quality information and analyses relating S and N deposition to concentrations of SO_x, N oxides and PM. Section II.C summarizes the currently available ecological effects evidence as summarized in the ISA, focusing on consideration of key policy-

¹⁹ In addition to the review's opening "Call for Information" (78 FR 53452, August 29, 2013), multiple search methodologies were applied to identify relevant scientific findings that have emerged since the 2008 ISA. Search techniques for the current ISA identified and evaluated studies and reports that have undergone scientific peer review and were published or accepted for publication between January 2008 (providing some overlap with the cutoff date for the last ISA) and May 2017. Studies published after the literature cutoff date for this ISA were also considered if they were submitted in response to the Call for Information or identified in subsequent phases of ISA development, particularly to the extent that they provide new information that affects key scientific conclusions. References that are cited in the ISA, the references that were considered for inclusion but not cited, and electronic links to bibliographic information and abstracts can be found at: https://hero.epa.gov/hero/index.cfm/project/page/project_id/2965 (ISA, section IS.1.2).

relevant aspects. Section II.D summarizes the exposure and risk information for this review, drawing on the quantitative analyses of aquatic acidification risk, presented in the PA. Section II.E presents the Administrator's proposed conclusions on the current standards and potential alternatives (section II.E.3), drawing on both evidence-based and exposure/risk-based considerations from the PA (section II.E.1) and advice from the CASAC (section II.E.2).

A. Introduction

As is the case for all such reviews, this review is based, most fundamentally, on using the Agency's assessments of the current scientific evidence and associated quantitative analyses to inform the Administrator's judgment regarding secondary standards for SO_x, N oxides and PM that are requisite to protect the public welfare from known or anticipated adverse effects associated with that pollutant's presence in the ambient air. The EPA's assessments are primarily documented in the ISA and PA, both of which have received CASAC review and public comment (82 FR 15702, March 30, 2017; 82 FR 15701, March 30, 2018; 83 FR 29786; June 26, 2018; 83 FR 31755, July 9, 2018; 85 FR 16093; March 20, 2020; 88 FR 34852, May 31, 2023; 88 FR 17572, March 23, 2023; 88 FR 45414, July 17, 2023). In bridging the gap between the scientific assessments of the ISA and the judgments required of the Administrator in his decisions on the current standard, the PA evaluates policy implications of the assessment of the current evidence in the ISA and the quantitative exposure and risk analyses and information documented in the PA. In evaluating the public welfare protection afforded by the current standard, the four basic elements of the NAAQS (indicator, averaging time, level, and form) are

considered collectively.²⁰

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. 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. This approach is 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 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 based on the Administrator's informed

²⁰ 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 SO₂ is the second highest 3-hour average in a year. The level of the standard defines the air quality concentration used for that purpose.

assessment of what constitutes requisite protection against adverse effects to public welfare. A public welfare policy decision draws upon scientific information and analyses about welfare effects, exposure and risks, as well as judgments about the appropriate response to the range of uncertainties that are inherent in the scientific evidence and analyses. The ultimate determination as to what level of damage to ecosystems and the services provided by those ecosystems is adverse to public welfare is not wholly a scientific question, although it is informed by scientific studies linking ecosystem damage to losses in ecosystem services and information on the value of those losses of ecosystem services. In reaching such decisions, the Administrator seeks to establish standards that are neither more nor less stringent than necessary for this purpose.

Thus, 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 these considerations are the nature and degree of effects of the pollutant, including the Administrator's judgments on 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.

1. Basis for Existing Secondary Standards

In the last review of the secondary standards for SO_x and N oxides, completed in 2012, the EPA retained the existing 3-hour SO₂ standard, with its level of 0.5 ppm, and the annual NO₂ standard, with its level of 0.053 ppm (77 FR 20218, April 3, 2012). Both of these secondary standards were established in 1971 (36 FR 8186, April 30, 1971). The basis for both the existing SO₂ and NO₂ secondary standard is to provide protection to the public welfare related to direct effects on vegetation (U.S. DHEW, 1969a; U.S. EPA, 1971).

The welfare effects evidence for SO_x in previous reviews indicates a relationship between short- and long-term SO₂ exposures and foliar damage to cultivated plants, reductions in productivity, species richness, and diversity (U.S. DHEW, 1969a; U.S. EPA, 1982c; U.S. EPA, 2008). At the time the standard was set, concentrations of SO₂ in the ambient air were also associated with other welfare effects, including effects on materials and visibility related to sulfate, a particulate transformation product of SO₂ (U.S. DHEW, 1969a). However, the available data were not sufficient to establish a quantitative relationship between specific SO₂ concentrations and such effects (38 FR 25679, September 14, 1973). Accordingly, direct effects of SO_x in ambient air on vegetation is the basis for the existing secondary standard for SO_x. Effects on materials and visibility (which relate to particles in air, including sulfates) have more recently been considered in the PM secondary NAAQS reviews (e.g., 85 FR 82684, December 18, 2020).

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, 1982c; U.S. EPA, 1993; U.S.

EPA, 2008a). 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 NO₂ secondary standard provided adequate protection related to the “direct” effects of airborne N oxides on vegetation on which the standard is based).

In the last review of the secondary PM standards with regard to protection from ecological effects, completed in 2013, the EPA retained the 24-hour PM_{2.5} standard, with its level of 35 µg/m³, and the 24-hour PM₁₀ standard, with its level of 150 µg/m³ (78 FR 3228, January 15, 2013). With regard to the annual PM_{2.5} standard, the EPA retained the averaging time and level, set at 15 µg/m³, while revising the form to remove the option for spatial averaging consistent with this change to the primary annual PM_{2.5} standard (78 FR 3225, January 15, 2013). The 2013 review considered the PM standards with regard to protection for an array of effects that include effects on visibility, materials damage, and climate effects, as well as ecological effects, and the EPA 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 NO₂ and SO₂ NAAQS (78 FR 3202, January 15, 2013). The ecological effects of PM considered include direct effects on plant foliage as well as effects of the ecosystem loading of PM constituents such as metals or organic compounds (2009 ISA, 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, provided the requisite protection (78 FR 3086, January 15, 2013).

2. Prior Review of Deposition-Related Effects

In the 2012 review of the NO₂ and SO₂ secondary standards, 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 (77 FR 20236, April 3, 2012). Considering the extensive evidence available at that time, the Agency concluded that the most significant risks of adverse effects of N oxides and SO_x to public welfare were 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 multiple quantitative linkages between pollutants in ambient air and ecosystem responses and potential public welfare implications.

While there is extensive evidence of deleterious effects of excessive nitrogen loadings to terrestrial and aquatic ecosystems, 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, 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, limiting the information on the effects of changes in N deposition on forestlands and other terrestrial ecosystems (2011 PA, section 6.3.2). Further, only a fraction of the deposited N was reported to be taken up by the forests, with most of the N retained in the soils, such that forest management practices can significantly affect the nitrogen cycling within a forest ecosystem (2008 ISA section 3.3.2.1 and 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 atmospheric N deposition (2011 PA, section 6.3.2).

The linkages between terrestrial acidification and atmospheric deposition of N and S compounds were also limited by the sparseness of available data for identifying appropriate assessment levels for terrestrial acidification indicators and uncertainties with regard to empirical case studies in the ISA (e.g., the potential for other stressors to confound relationships between deposition and terrestrial acidification effects). However, the evidence in the 2008 ISA and the REA analyses of aquatic acidification provided strong support to the evidence for a relationship between atmospheric deposition of N and S compounds and loss of acid neutralizing capacity (ANC) in sensitive ecosystems, with associated aquatic acidification effects.

In light of the evidence and findings of these analyses and advice from the CASAC, the
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PA concluded it was 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 (2011 PA, section 7.6.2). Conceptually, the AAI is an index that uses the results of ecosystem and air quality modeling to estimate waterbody ANC. The standard level for an AAI-based standard was conceptually envisioned to be a national minimum target ANC for waterbodies in the ecoregions of the U.S. for which data were considered adequate for these purposes (2011 PA, section 7.6.2).

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 ("F factors"), 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 (2011 PA, Chapter 7).²¹ The ecoregion-specific values for these factors would be specified based on then available data and simulations of the Community Multiscale Air Quality (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 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 too great to support establishment of a new standard 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

²¹ 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.

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. General Approach for This Review

As is the case for all NAAQS reviews, this secondary standards review uses the Agency’s assessment of the current scientific evidence and associated quantitative analyses as a foundation 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 for this review of the secondary SO_x, N oxides, and PM standards builds on the last reviews of those

pollutants, 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.

This review of the secondary standards for SO_x, N oxides, and PM assesses the protection provided by the standards from two categories of effects: direct effects of the airborne pollutants and indirect effects of the associated S- and N-containing compounds (in gaseous and particulate form) deposited in ecosystems. In so doing, the review draws on the currently available evidence as assessed in the ISA (and prior assessments) and quantitative exposure, risk, and air quality information in the PA, including the REA for aquatic acidification.

With regard to direct effects, we draw 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. Based on this information, we consider the policy implications, most specifically whether the evidence supports the retention or revision of the current NO₂ and SO₂ secondary standards. With regard to the effects of PM, we 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.

With regard to deposition-related effects, we consider the evidence for the array of effects identified in the ISA (and summarized in section II.B below), including both terrestrial and aquatic effects; and the limitations in the evidence and associated uncertainties; as well as the public welfare implications of such effects. The overall approach takes into account the nature of the welfare effects and the exposure conditions associated with effects in identifying S and N

deposition levels appropriate to consider in the context of public welfare protection. To identify and evaluate metrics relevant to air quality standards (and their elements), we have assessed relationships developed from air quality measurements near pollutant sources and deposition estimates nearby and in downwind ecoregions. In so doing, the available quantitative information both on deposition and effects, and on ambient air concentrations and deposition, has been assessed with regard to the existence of linkages between SO_x, N oxides, and PM in ambient air and deposition-related effects. These assessments then inform judgments on the likelihood of occurrence of deposition-related effects under air quality that meets the existing standards for these pollutants, or potential alternatives.

In considering the information on deposition and effects, we recognize that the impacts from the dramatically higher deposition rates 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 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, Appendix 4, section 4.6.1). Thus, the extent to which S and N compounds (once deposited) are retained in soil matrices (with potential effects on soil chemistry) influences the dynamics of the response of the various environmental pathways to changes in air quality, in addition to the influences of emissions, ambient air concentrations and associated deposition.

The two-pronged approach to this review’s consideration of deposition-related effects
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based on the available information in the ISA (summarized in section II.C and II.D below) includes the consideration of deposition levels that may be associated with ecological effects of potential concern. In this step, we consider and strive to focus on effects for which the evidence is most robust with regard to established quantitative relationships between deposition and ecosystem effects. The information for terrestrial ecosystems is derived primarily from analysis of the evidence presented in the ISA. For aquatic ecosystems, primary focus is given to effects related to aquatic acidification, for which we have conducted quantitative risk and exposure analyses based on available modeling applications that relate acid deposition and acid buffering capability in U.S. waterbodies, as summarized in section II.D below (PA, section 5.1 and Appendix 5A).

In parallel fashion to identification of deposition levels for consideration, air quality and deposition analyses have been employed to inform an understanding of relationships between ambient air concentrations near pollutant sources in terms of metrics relevant to air quality standards (and their elements) and ecosystem deposition estimates. As described in section II.B below, several different types of analyses have been performed in this review for this purpose. Interpretation of findings from these analyses, in combination with the identified deposition levels of interest, and related policy judgments regarding limitations and associated uncertainties of the underlying information, inform the Administrator's proposed conclusions on the extent to which existing standards, or potential alternative standards, might be expected to provide protection from these levels.

In summary, our approach to evaluating the standards with regard to protection from ecological effects related to ecosystem deposition of N and S compounds (presented in the sections that follow) involves multiple components: (1) review of the scientific evidence to

identify the ecological effects associated with the three pollutants, both those related to direct pollutant contact and to ecosystem deposition; (2) assessment of the evidence and characterization of the REA results to identify deposition levels related to categories of ecosystem effects; (3) analysis of relationships between ambient air concentrations of the three pollutants and deposition of N and S compounds to understand key aspects of these relationships that can inform the Administrator's decisions on policy options for ambient air standards to protect against air concentrations associated with direct effects and with deposition-related effects that are judged adverse to the public welfare. As is described in sections II.B and II.E, for two of the pollutants, N oxides and PM, relating ambient air concentrations to deposition (of N compounds) is particularly complex because N deposition also results from an additional air pollutant that is not controlled by NAAQS for N oxides and PM. Thus, separate from the evaluation of standards for SO_x, the evaluation for N oxides and PM also considers current information (e.g., spatial and temporal trends) related to the additional air pollutant, ammonia (NH₃), that contributes to N deposition and to PM components that do not contribute to N deposition. Evaluation of all of this information, together, is considered by the Administrator in reaching his proposed decision, as summarized in section II.E.

B. Air Quality and Deposition

The three criteria pollutants that are the focus of this review (SO_x, N oxides, and PM) include both gases and particles. Both their physical state and chemical properties, as well as other factors, influence their deposition as N- or S-containing compounds. The complex pathway from emissions of these pollutants and their precursors to eventual deposition varies by pollutant and is influenced by a series of atmospheric processes and chemical transformations that occur at multiple spatial and temporal scales (PA, Chapters 2 and 6).

A complication in the consideration of the influence of these criteria pollutants on N deposition (and associated ecological effects) is posed by the contribution of other, non-criteria, pollutants in ambient air, specifically NH₃. As summarized below, although there is a decreasing temporal trend in emissions of N oxides, the coincident increasing trend in NH₃ emissions has reduced the influence of N oxides on N deposition (PA, sections 6.2.1, 6.4.2 and 7.2.3.3). Variability and temporal changes in the composition of PM, including with regard to N- (and S-) containing compounds, is another factor affecting decisions in this review (as discussed in sections II.1.d(3)) and II.3 below).

This section includes a brief summary of the major emission sources of SO_x, N oxides, and PM (section II.B.1). This is followed by a description of how those emissions are transported and transformed within the atmosphere to eventually contribute to S and N deposition (section II.B.1). Available information on current levels of emissions and air concentrations of these three pollutants across the U.S. and their trends is summarized in section II.B.2, accompanied by a description of estimated deposition levels across the U.S. and how they have changed over the past two decades. Finally, while many of the ecological effects examined in this review are associated with deposition of N and S, the NAAQS are set in terms of pollutant concentrations. To that end, section II.B.3 discusses the findings of analyses performed to relate ambient air concentrations of the relevant pollutants and S or N deposition, over a range of conditions (e.g., pollutant, region, time period), and summarizes key observations that may inform the Administrator's judgments in this review.

1. Sources, Emissions and Atmospheric Processes Affecting SO_x, N Oxides and PM

Sulfur dioxide is one of a small group of highly reactive gases collectively known as SO_x. 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, SO₂ is the indicator for the existing NAAQS for SO_x. The main anthropogenic source of SO₂ emissions is fossil fuel combustion (PA, section 2.2.2). Based on the 2020 National Emissions Inventory (NEI), the top three emission sources of SO₂ in the U.S. are: coal-fired electrical generating units (48% of total), industrial processes (27%), and other stationary source fuel combustion (9%).

Once emitted to the atmosphere, the atmospheric lifetime of SO₂ is typically less than 1-2 days; it can either remain in the gas phase or be oxidized to form sulfate particles (SO₄²⁻). Modeling studies suggest that oxidation accounts for more than half of SO₂ removal on a national basis (PA, section 2.1.1). The rate of SO₂ oxidation accelerates with greater availability of oxidants. Oxidants are generally depleted near source stacks, so that more SO₂ is oxidized to SO₄²⁻ in cleaner air downwind of SO_x sources (2008 ISA, section 2.6.3.1). The atmospheric lifetime of SO₄²⁻ particles is longer, ranging from 2 to 10 days. As SO₄²⁻ particles are generally within the fine particle size range, they are a component of PM_{2.5} (PA, section 2.1.1). The spatial distribution of both SO₂ and SO₄²⁻ deposition reflects the distribution of SO_x emissions (i.e., most S deposition is in the eastern U.S.; PA, section 2.5.3) and wind patterns. Precipitation variability also modulates the spatial distribution of S wet deposition. In sum, both SO₂, and the SO₄²⁻ particles converted from SO₂, contribute to S deposition but do so over different time and geographic scales, with dry deposition of SO₂ typically occurring near the source, and wet deposition of sulfate particles being more regional in nature.

The term N oxides refers to all forms of oxidized nitrogen compounds, including nitric oxide (NO), NO₂, nitric acid (HNO₃), and particulate nitrate (NO₃⁻). Most N oxides enter the atmosphere as either NO or NO₂, which are collectively referred to as NO_x (PA, section 2.1.2).

Anthropogenic sources account for the majority of NO_x emissions in the U.S., per the 2020 NEI,

with highway vehicles (26% of total), stationary fuel combustion which includes electric generating units (25%), and non-road mobile sources (19%) identified as the largest contributors to total emissions. Other anthropogenic NO_x sources include agricultural field burning, prescribed fires, and various industrial processes such as cement manufacturing and oil and gas production (PA, section 2.2.1).

Once emitted into the atmosphere, NO_x can deposit to the surface or be chemically converted to other gaseous N oxides, including HNO₃, as well as to particulate NO₃⁻. Unlike particulate SO₄²⁻, which exists almost entirely in the fine particle range, NO₃⁻ particles may occur either in the fine or coarse size range, such that not all particulate NO₃⁻ is a component of PM_{2.5}. Each form of oxidized N is removed from the atmosphere at different rates by both dry and wet deposition. As a general rule, the gas phase species tend to have shorter atmospheric lifetimes, either dry depositing (e.g., as HNO₃) or quickly converting to particulate NO₃⁻. Particulate NO₃⁻ is more efficiently removed by precipitation (wet deposition) and has a similar atmospheric lifetime as particulate SO₄²⁻ (2-10 days).

In addition to N oxides, there is another category of nitrogen pollutants, referred to as reduced nitrogen, which is distinct from N oxides but also contributes to nitrogen deposition. The most common form of reduced N in the air is ammonia gas (NH₃). Sources of NH₃ emissions include livestock waste (49% of total in 2020 NEI), fertilizer application (33%) and aggregate fires (11%). Ammonia tends to dry deposit near sources (PA, section 2.1.3). It can also be converted to particle form, as ammonium (NH₄⁺), which can be transported farther distances and is most efficiently removed by precipitation (PA, section 2.1.3). Ammonia, unlike N oxides or PM_{2.5}, is not a criteria pollutant and is not directly regulated under CAA section 109.

In sum, particulate matter is both emitted to the atmosphere and can be formed in the

atmosphere from precursor chemical gases (such as is the case for NO_x and SO_x). The components of PM_{2.5} mass that contribute to S and N deposition are secondary products formed in the atmosphere after being emitted (e.g., particulate sulfate, particulate NO₃⁻, NH₄⁺). There are other components of PM_{2.5} mass that do not contribute to S and N deposition, e.g., black carbon, organic carbon, dust (PA, section 2.4.3).

2. Recent Trends in Emissions, Concentrations, and Deposition

Emissions of SO_x, oxides of N, and PM have declined dramatically over the past two decades, continuing a longer-term trend (PA, section 2.2). NEI data indicate an 87% decrease in total SO₂ emissions between 2002 and 2022, including reductions of 91% in emissions from electricity generating units and 96% in emissions from mobile sources. Total anthropogenic NO_x emissions have also trended downward across the U.S. between 2002 and 2022 at only slightly smaller percentages than SO₂. Nationwide estimates indicate a 70% decrease in anthropogenic NO_x emissions over this time period, driven in part by large emission reductions in the highway vehicle sector (84%) and from stationary fuel combustion (68%) (PA, section 2.2.1). In contrast with these declining 20-year trends in NO_x and SO_x emissions, the annual rate of NH₃ emissions has increased by over 20 percent since 2002 (PA, section 2.2.3). The two largest contributors are emissions from livestock waste and fertilizer application, which have increased by 11% and 44%, respectively, from 2002 to 2022. These trends in emissions have had ramifications for N deposition patterns across the U.S., as described further below.

As expected, the large reductions in SO_x and NO_x emissions have resulted in substantially lower ambient air concentrations in recent years relative to what was observed in previous periods. The State and Local Air Monitoring Stations (SLAMS) network supports the implementation of the NAAQS. In 2021, all ambient monitoring sites with valid SO₂ design

values (n= 333)²² are less than the level of the existing secondary standard (500 ppb)²³ and more than 75 percent of the sites have design values less than 20 ppb (PA, section 2.4.2). These values reflect a downward trend over the past two decades with median 3-hour secondary SO₂ values down substantially from 2000 levels (from ~50 ppb to ~10 ppb).

Similar trends are evident in the data for the primary SO₂ standard (annual 99th percentile of 1-hour daily maximum concentrations, averaged over 3 years with a level of 75 ppb). In the 2019-2021 period, the maximum design value for the primary SO₂ standard was 376 ppb at a monitoring site near an industrial park in southeast Missouri. It is important to note that peak and mean SO₂ concentrations are higher at source-oriented sites than monitoring locations that are not source-oriented. Additionally, it is not uncommon for there to be high SO₂ values in areas with recurring volcanic eruptions (e.g., Hawaii). In the mid-1990s, the median value of all sites with valid 1-hour SO₂ design values often exceeded 75 ppb (PA, Figure 2-26). Since then, the entire distribution of values (including source-oriented sites) has continued to decline such that the median value across the network of sites is now between 5 and 10 ppb (PA, Figure 2-26). The EPA also evaluated trends in annual average SO₂ data from 2000-2021 and observed improving trends of similar magnitude with the longer-term (annual) averaging time. It is important to note that both peak and mean SO₂ concentrations are higher at source-oriented sites than monitoring locations that are not source-oriented.

²² 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-designvalues>).

²³ The existing secondary standard for SO₂ is 0.5 ppm (500 ppb), as a 3-hour average, not to be exceeded more than once per year.

Regarding NO₂, design values at all 399 sites with valid secondary NO₂ design values (annual average concentrations) in 2021 are less than the 53 ppb level of the existing secondary standard,²⁴ and the majority of sites (98 percent) have design values that are less than 20 ppb. In 2021, the maximum was 30 ppb,²⁵ and the median was 7 ppb. As with SO₂, the more recent NO₂ design values also reflect a downward trend over the past two decades. Median annual NO₂ design values across the U.S. decreased by ~50% between 2000 and 2021 (15 ppb to 7 ppb).

Likewise, the median of the annual average PM_{2.5} concentrations decreased substantially from 2000 to 2021 (from 12.8 µg/m³ to 8 µg/m³). The median of the annual 98th percentile 24-hour PM_{2.5} concentrations at the more than 1000 sites monitored also decreased, from 32 µg/m³ in 2000 to 21 µg/m³ in 2021. Although both the annual average and 98th percentile 24-hour PM_{2.5} concentrations decreased steadily from the early 2000s until 2016, these values have fluctuated in recent years due to large-scale wildfire events (PA, section 2.4.3; U.S. EPA, 2023, Figures 23 and 24).

These emission reductions and subsequent downward trends in air concentrations have also contributed to a nationwide decrease in N and S deposition (PA, sections 2.5.3 and 6.2.1). Total S deposition and N deposition declined by 68% and 15%, respectively, calculated as a nationwide, three-year average between 2000-2002 and 2019-2021 (PA, section 6.2.1). The trend in S deposition is more robust than for N because of the offsetting influence of increasing emissions of reduced forms of nitrogen over the same timeframe. The largest reductions in total

²⁴ Sites in the contiguous U.S. have met the existing NO₂ secondary standard since around 1991 (PA, Figure 2-22).

²⁵ The maximum annual average NO₂ concentrations has been at, slightly above or slightly below 30 ppb since about 2008, with the highest 3-year average value just above 30 ppb (PA, Figures 2-22 and 7-9).

S and N deposition are seen in regions downwind of point sources and transportation corridors related to emission reductions from electricity generating units and mobile sources.

3. Relationships Between Concentrations and Deposition

As the NAAQS are set in terms of pollutant concentrations, analyses in the PA evaluated relationships between criteria pollutant concentrations in ambient air and ecosystem deposition across the U.S. We examined these relationships over a range of conditions (e.g., pollutant, region, time period), and considered deposition both near sources and at distance (allowing for pollutant transport and associated transformation). The findings of these analyses, described in detail in Chapter 6 and Appendix 6A of the PA, have informed consideration of indicators and levels for potential secondary standards based on consideration of deposition-related effects (PA, Chapter 7).

As is evident from the air quality-deposition analyses, relating ecosystem deposition to ambient air concentrations is not straightforward. Deposition rates vary across ecosystems nationally, and there is not a simple one-to-one relationship between ambient air concentrations of any one indicator and S or N deposition. As discussed above, the atmospheric processes that lead from pollutant emissions leading to eventual deposition to the earth's surface are complex. Multiple chemicals, both gaseous and particulate, from multiple types of sources contribute to S and N deposition. Further, both criteria pollutants and non-criteria pollutants contribute to N deposition. There are also multiple deposition pathways (i.e., dry deposition and wet deposition) that can influence the spatial and temporal scales at which deposition occurs, which vary by pollutant and pollutant phase.

In light of these challenges, the PA employed five different approaches for considering relationships between S and N deposition rates and ambient air concentrations. First, as part of a

“real-world experiment,” the PA analyses leveraged the recent downward trends in NO_x and SO_x emissions and corresponding air quality concentrations as well as the trends in deposition estimates (TDep or total deposition) to examine the correlation between observed decreases in emissions and concentration and observed changes in deposition over the past two decades (PA, section 6.2.1). The TDep estimates used in these analyses are based on a hybrid approach that involves 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).²⁶ For the second approach, we assessed how air quality concentrations and associated deposition levels are related within a chemical-transport model (CMAQ²⁷) both nationally and then at certain Class I areas²⁸ (PA, section 6.2.2.1) where additional monitoring data are collected as part of the Clean Air Status and Trends Network (CASTNET) and the Interagency Monitoring of Protected Visual Environments (IMPROVE) networks. As a third approach, we analyzed the relationships across a limited number of monitoring locations (in Class I areas) where both air quality data (CASTNET and IMPROVE) and wet deposition of S

²⁶ Other than the estimates associated with the CMAQ analysis (second approach referenced above), the deposition estimates used in these analyses are those provided by the National Atmospheric Deposition Program, TDep Science Committee. 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).

²⁷ The CMAQ is a state of the science photochemical 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. See <https://www.epa.gov/cmaq> for more detail.

²⁸ 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.

and N was measured to evaluate the associations between concentrations and deposition at a local scale (PA, section 6.2.2.2 and 6.2.2.3). The fourth approach also considered the local associations between the two terms at the local scale but did so using a broader set of ambient air concentration measurements (i.e., all valid SO₂, NO₂, and PM_{2.5} measurements at SLAMS across the U.S.) and a hybrid set of deposition estimates (TDep) (PA, section 6.2.3).

Finally, in recognition of the fact that air quality at upwind locations can also influence downwind deposition, the fifth approach used a trajectory model (HYSPLIT – The Hybrid Single-Particle Lagrangian Integrated Trajectory model) to identify upwind areas where emissions might be expected to influence deposition at downwind ecoregions (PA, section 6.2.4 and Appendix 6A).²⁹ Once those potential zones of influence were established, we evaluated the relationships between air quality metrics for the three pollutants³⁰ at sites within those zones with deposition estimates in the downwind ecoregion, as 3-year averages for five periods: 2001-2003, 2006-2008, 2010-2012, 2014-2016 and 2018-2020. The metrics, Ecoregion Air Quality Metrics (EAQMs), include a weighted-average (EAQM-weighted) and a maximum metric (EAQM-max). The EAQM-max is the maximum metric value among the sites linked to the downwind ecoregion and, for the EAQM-weighted, the value of each site linked to the downwind ecoregion 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 (PA, section 6.2.4.1).

²⁹ Upwind sites of influence were identified for all 84 ecoregions (level III categorization) in the contiguous U.S.

³⁰ For SO₂, there were two sets of metrics: one based on an annual average and one based on the 2nd highest 3-hour maximum concentration in the year. Both the NO₂ and PM_{2.5} metrics are annual averages. For relating to 3-year average deposition, all are averaged across three years.

As with any assessment, there are uncertainties and limitations associated with the analyses summarized above. These are more fully discussed in the PA (PA, sections 6.3 and 6.4). The evaluation of measured air quality concentrations (SO₂, NO₂, and PM_{2.5}) and TDep estimates of deposition at all SLAMS (generally composed of sites that use either a Federal Reference Method [FRM] or a Federal Equivalence Method [FEM]) is a robust analysis (i.e., large number of monitors distributed across the U.S.) and particularly relevant given that compliance with the current standards (both primary and secondary) is judged using design value metrics based on measurements at the current SO₂, NO₂ and PM_{2.5} monitors. However, these site-based comparisons do not account for deposition associated with the transport of pollutants emitted some distance upwind. Each of the other analyses completed to bolster this analysis have their own limitations ranging from model uncertainty to limited geographical scope.

The full set of quantitative results of the characterization of air quality and deposition relationships are discussed more thoroughly in Chapter 6 and Appendix 6A of the PA. In combination, these analyses supported the PA conclusion of a strong association between SO₂ and S deposition. Regarding N oxides and PM, however, the results, and associated information, indicated more variable relationships between NO₂ concentrations and N deposition, and PM_{2.5} concentrations with either S or N deposition.

For SO₂, annual monitored SO₂ concentrations, at existing monitors within the SLAMS network, averaged over 3 years at the national scale were highly correlated to S deposition estimates in the TDep dataset at the local scale (correlation coefficient of 0.70),³¹ especially in

³¹ The correlation coefficients are based on Spearman's rank correlation coefficient. These coefficients are generally used to assess how well the relationship between two variables can be described via a monotonic function. The term "r value" is sometimes used as shorthand for this

the earlier periods of the record and across the eastern U.S. (PA, section 6.2.3). This association was confirmed by the relationships between SO₂ annual values at the identified upwind sites of influence and S deposition estimates from TDep in downwind ecoregions, especially in those locations where the annual average SO₂ concentrations are greater than 5 ppb (PA, section 6.2.4.2). Finally, we note that the observed declines in national levels of S deposition over the past two decades have occurred during a period in which emissions of SO₂ have also declined sharply (PA, sections 6.2.1 and 6.4.1).

Analyses in the PA also investigated relationships between S deposition and air quality metrics other than the current indicator species (SO₂) in a limited number of circumstances. For example, an evaluation of the association between SO₄²⁻ and total S deposition across 27 Class I areas where data for both parameters were available, concluded that the correlations between particle sulfate and total sulfate (i.e., SO₂ + SO₄²⁻) with total S deposition (correlation coefficients of 0.55 and 0.61, respectively) was lower than what was exhibited for SO₂ and S deposition at the SLAMS (PA, section 6.2.2). The analyses also concluded that there was poor correlation (correlation coefficient of 0.33) between PM_{2.5} mass, as measured at IMPROVE sites, with total S deposition estimates for those sites (PA, sections 2.3.3 and 6.2.2.3). While these analyses are based on data at a relatively limited number of sites, as compared to the SLAMS network, the results suggest that there are no clear advantages to considering PM_{2.5} mass, particle sulfate, or total sulfate as an indicator for a secondary NAAQS, over using SO₂.

Both NO₂ and certain components of PM_{2.5} can contribute to N deposition. As is the case for SO₂ and S deposition, there are multiple pathways for N deposition (dry and wet), and

correlation coefficient. Higher values indicate that the two variables are highly associated with one another (can range from 1.0 to -1.0).

multiple scales of N deposition (local and regional). However, there are some additional complications in the consideration of how air quality concentrations (i.e., NO₂ and PM_{2.5} mass) are associated with eventual N deposition. First, not all N deposition is caused by the criteria pollutants (PA, Chapter 2 and section 6.1.1). 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., NO₃⁻ and NH₄⁺). As a result of these two factors, the association between NO₂ concentrations and N deposition, and PM_{2.5} concentrations and N deposition is less robust than what is observed for SO₂. Our multi-faceted approach to evaluating these relationships confirmed this expectation. For example, when comparing NO₂ observations at SLAMS across the U.S. against the N deposition estimates from TDep, there are weaker associations than what is observed in the similar SO₂ comparisons (PA, section 6.4.2). There is little correlation for N deposition with concentrations of NO₂, as evidenced by a Spearman's correlation coefficient of 0.38, compared to 0.70 for SO₂ and S deposition (PA, Table 6-6 and Table 6-4). Further, the trajectory-based analyses of the relationships between NO₂ annual values in the identified upwind zones of influence and N deposition estimates from TDep in downwind ecoregions indicate negative correlations (PA, Table 6-10). These negative correlations are observed for both the EAQM-weighted and EAQM-max values. This relative lack of association was confirmed by considering national trends over the past 20 years, where sharp declines in NO₂ emissions and concentrations are linked in time with sharp declines in oxidized N deposition (PA, Table 6-2), but not associated with recent trends in total or reduced atmospheric N deposition. Since 2010, NO₂ concentrations have continued to drop while N deposition has remained steady (PA, section 6.2.1). As noted for S deposition and S compound metrics above, the PA also investigated relationships between N deposition and air quality metrics other than the

current indicator species (NO₂). Across the 27 Class I areas where collocated data were available, the PA evaluated the relationships between several air quality parameters (e.g., nitric acid, particulate NO₃⁻, and NH₄⁺) and, as for S deposition and S compound metrics, the PA concluded there were no clear advantages over the consideration of NO₂ or PM_{2.5} mass. In sum, the evidence suggests that NO₂ would be a weak indicator of total atmospheric N deposition, especially in areas where ammonia is prevalent or where PM_{2.5} mass is dominated by species other than NO₃⁻ or NH₄⁺ (PA, section 6.4.2).

C. Welfare Effects Evidence

The information summarized here is based on our scientific assessment of the welfare effects evidence available in this review; this assessment is documented in the ISA³² and its policy implications are further discussed in the PA (and summarized in section II.E.1 below). More than 3,000 studies are newly available since the last review and considered in the ISA.³³ While expanding the evidence for some effect categories, studies on acid deposition, a key group of effects from the last review, are largely consistent with the evidence that was previously available. The subsections below briefly summarize the following aspects of the evidence: the nature of welfare effects of S oxides, N oxides and PM (section II.C.1); the potential public welfare implications (section II.C.2); and exposure concentrations and deposition-related metrics (section II.C.3).

1. Nature of Effects

³² The ISA builds on evidence and conclusions from previous assessments, focusing on synthesizing and integrating the newly available evidence (ISA, section IS.1.1). Past assessments are cited when providing further details not repeated in newer assessments.

³³ The study count and citations are available on the project page for the ISA on the Health & Environmental Research Online (HERO) website documents these studies (https://heronet.epa.gov/heronet/index.cfm/project/page/project_id/2965).

This welfare effects evidence base available in the current review includes decades of extensive research on the ecological effects oxides of nitrogen, oxides of sulfur and PM. In the sections below we summarize the nature of the direct effects of gas-phase exposure to oxides of nitrogen and sulfur (section II.C.1.a), acid deposition-related ecological effects (section II.C.1.b), N enrichment and associated effects (section II.C.1.c), and other effects (section II.C.1.d).

a. Direct Effects of SO_x and N Oxides

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 SO₂, NO₂, NO, HNO₃ and peroxyacetyl nitrate (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. 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 SO₂ and injury to vegetation (ISA, Appendix 3, section 3.6.1), and between gas-phase NO, NO₂ 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 HNO₃ and changes to vegetation, noting that experimental exposure can damage leaf cuticle of tree seedlings and HNO₃ concentrations have been reported to have contributed to declines in lichen species in the Los Angeles basin (ISA, Appendix 3, section 3.6.3).

Specifically for SO_x, 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, SO₂ 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). For lichens, damage from SO₂ exposure has been observed to include reduction in metabolic functions that are vital for growth and survival (e.g., decreases in photosynthesis and respiration), damage to cellular integrity (e.g., leakage of electrolytes), 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 SO₂ 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 SO₂-sensitive tree species (e.g., Thomas et al., 2013). Although the authors attributed the growth response to reductions in SO₂-associated acid deposition, and related recovery from soil acidification, the relative roles of different pathways are unclear as a historical deposition record was not available (ISA, Appendix 3, section 3.2). Other researchers have suggested that the observed 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 SO₂, 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 *Air Quality Criteria Document for Oxides of N* included evidence of phytotoxic effects of NO, NO₂, and PAN on plants through decreasing photosynthesis and induction of visible foliar injury (U.S. EPA, 1993 [1993 AQCD]). The 1993 AQCD additionally concluded that concentrations of NO, NO₂, 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 indicates that HNO₃ had a role in observed declines in lichen species in the 1970s in the Los Angeles basin (ISA, Appendix 3, section 3.3). A 2008 resampling of areas shown to be impacted in the past by HNO₃ 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). The recently available evidence on this topic also included a study of six lichen species that reported changes in physiology and functioning including decreased chlorophyll content and chlorophyll fluorescence, decreased photosynthesis and respiration, and increased electrolyte leakage from HNO₃ exposures for 2-11 weeks (daily peak levels near 50 ppb) in controlled chambers. (ISA, Appendix 3, section 3.4).

b. Acid Deposition-Related Ecological Effects

The connection between SO_x and N oxide emissions to ambient air, atmospheric deposition of N and/or S, and the acidification of acid-sensitive soils and surface waters is well documented with many decades of evidence, particularly in the eastern U.S. (ISA, section IS.5; Appendix 8, section 8.1). In the atmosphere, SO_x and N oxides undergo reactions to form various acidic compounds that are removed from the atmosphere through deposition. 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). These effects depend on the magnitude and rate of deposition, as well as multiple biogeochemical processes that occur in soils and waterbodies.

Soil acidification is influenced by the deposition of inorganic acids (HNO₃ and H₂SO₄),
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and by chemical and biological processes. When NO_3^- , or SO_4^{2-} leach from soils to surface waters, an equivalent number of positive cations, or countercharge, are also transported. If the countercharge is provided by a base cation (e.g., calcium, $[\text{Ca}^{2+}]$, magnesium $[\text{Mg}^{2+}]$, sodium $[\text{Na}^+]$, or potassium $[\text{K}^+]$), rather than hydrogen ions (H^+), the leachate is neutralized, but the soil becomes more acidic from the hydrogen ions 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).

(1) Freshwater Ecosystems

As was the case in the last review, the body of evidence 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).

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 their history of 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).

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, ANC, and inorganic aluminum (Al) concentration (ISA, Appendix 8, Table 8-9).

The effects of waterbody acidification on fish species are especially well understood in
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the scientific literature, and many species (e.g., brown and brook trout and Atlantic salmon) have been documented to have experienced adverse effects from acidification (ISA, Appendix 8, section 8.3). 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 Al (ISA, Appendix 8, section 8.3.6.1). In general, biological effects in aquatic ecosystems are primarily attributable to low pH and high inorganic aluminum concentration (ISA, p. ES-14). Waterbody pH largely controls the bioavailability of Al, which is toxic to fish, and aluminum mobilization is largely confined to waters with a pH below about 5.5, which the ISA describes as corresponding to an ANC in the range of about 10 to 30 $\mu\text{eq/L}$ in low to moderate DOC waters of the Northeast (ISA, Appendix 7, section 7.1.2.6 and Appendix 8, section 8.6.4).

The parameter ANC is an indicator of the buffering capacity of natural waters against acidification. Although ANC does not directly affect biota, it is an indicator of acidification that relates to pH and aluminum levels (ISA, p. ES-14), or watershed characteristics like base cation weathering (BCw) rate (ISA, Appendix 8, sections 8.1 and 8.3.6.3). Accordingly, ANC is commonly used to describe the potential sensitivity of a freshwater system to acidification-related effects. It can be measured in water samples and is also often estimated for use in water quality modeling, as is done in the aquatic acidification risk assessment for this review, as summarized in section II.D below. Water quality models are generally better at estimating ANC than at estimating other indicators of acidification-related risk, such as pH. Acid neutralizing capacity is estimated 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). Thus, values below zero indicate a deficit in the ability to buffer acidic inputs, and increasing values above zero

represent increasing buffering capability for acidic inputs. Further, across waterbodies within impacted areas of Shenandoah National Park streams and Adirondack Mountain lakes, a positive relationship has often been observed between ANC and number of fish species, at least for the ANC range from about zero to 50 $\mu\text{eq/L}$ (ISA, Appendix 7, section 7.1.2.6; Cosby et al., 2006; Sullivan et al., 2006, Bulger et al., 1999).

Values of ANC can also be influenced by high concentrations of naturally occurring organic acids, which can reduce bioavailability of Al, buffering effects usually associated with low pH and high total Al concentrations (Waller et al., 2012; ISA, Appendix 8, section 8.3.6.4); in waters where that occurs, ANC may not be a good indicator of risk to biota.

In addition to acidity of surface waters quantified over weeks or months, waterbodies can also experience spikes in acidity in response to episodic precipitation or rapid snowmelt events. In these events (hours-days), a surge or pulse of drainage water, containing acidic compounds, is routed through upper soil horizons rather than the deeper soil horizons that would usually provide buffering for acidic compounds (ISA, Appendix 7, section 7.1). 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). 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 7, section 7.1.1.2; 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 watersheds and, in the east, their underlying geology (ISA, Appendix 8, section 8.5.1).

National survey data dating back to the early 1980s through 2004, 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). For example, 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 microequivalents per liter ($\mu\text{eq}/\text{L}$) (Sullivan et al., 2006). Values of ANC below 20 $\mu\text{eq}/\text{L}$ in Shenandoah stream sites were associated with fewer fish of sensitive species compared to sites with higher ANC (Bulger et al., 1999). A more recent study of two groups of Adirondack lakes for which water quality data were available from 1982 and 1992, respectively, reported significant increases in ANC in the large majority of those lakes, with the magnitude of the increases varying across the lakes (Driscoll et al., 2016; ISA, Appendix 7, section 7.1.3.1). 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). Despite the appreciable reductions in acidifying deposition that have occurred in the U.S. since the 1960s and 1970s, aquatic ecosystems across the U.S. are still experiencing effects from historical contributions of N and S (ISA, Appendix 8, section 8.6).

(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, 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).

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 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 Ca²⁺ 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).

In addition to Ca^{2+} addition experiments, the recently available evidence also includes addition or gradient studies evaluating relationships between soil chemistry indicators of acidification (e.g., soil pH, base cation to aluminum (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, Tables 5-2 and 5-6). Further, the 2020 ISA reports on several large observational studies evaluating 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 (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 (PA, sections 5.3.2.3 and 5.3.4 and Appendix 5B).

Although there has been no systematic national survey of U.S. terrestrial ecosystem soils, the forest ecosystems considered the most sensitive to terrestrial acidification from atmospheric deposition 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 ISA, Appendix 3, section 3.2.4.2; ISA, Appendix 5, section 5.3). 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). For example, “[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).

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. found 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). Recent 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).

c. Nitrogen Enrichment and Associated Ecological 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 for plants 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 plant species able to most effectively use 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 of vegetation, and potentially further and broader ramifications on ecosystem processes, structure, and function.

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.

(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, ES.5.2). Decomposition of the plant biomass from the subsequent algal die-off contributes to reduced waterbody oxygen, which in turn can affect higher-trophic-level species, e.g., contributing to fish mortality (ISA, p. ES-18).

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, and whether they may serve as a source, sink, or transformer of atmospherically deposited N, is extremely variable and depends on the type of wetland and other factors, such as physiography, and local hydrology, as well as climate (ISA, section IS.8.1 and Appendix 11, section 11.1). 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).

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, which receive 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). For freshwater fens, marshes, and swamps, inputs from ground and surface water are often of similar order of magnitude as that from precipitation, while estuarine and coastal wetlands receive water from multiple sources, with precipitation being among the smaller of those sources (ISA, Appendix 11, section 11.1).

Nitrogen loading and other factors contribute to nutrient enrichment, which contributes to eutrophication. Such nitrogen-driven eutrophication alters freshwater biogeochemistry and can impact physiology, survival, and biodiversity of sensitive aquatic biota. 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. With regard to freshwaters, for example, studies published since the 2008 ISA augment the evidence base for high-elevation waterbodies where the main N source is atmospheric deposition. Recent evidence continues to indicate that N limitation is common in oligotrophic waters in the western U.S., with shifts in nutrient limitation, from N limitation, to between N and phosphorus (P) limitation, or to P limitation, reported in some alpine lake studies (ISA, Appendix 9, section 9.1.1.3). Small inputs of N in such water bodies have been reported to increase nutrient availability or alter the balance of N and P, with the potential to stimulate growth of primary producers and contribute to changes in species richness, community composition, and diversity.

Another type of N loading effect in other types of freshwater lakes includes a role in the composition of freshwater algal blooms and their toxicity (ISA, Appendix 9, section 9.2.6.1). Information in this review, including studies in Lake Erie, 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; Davis et al., 2015; Gobler et al., 2016).

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 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 to total N loading (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 have been attributed to declines in atmospheric N deposition (ISA, Appendix 7, section 7.1.5.1).

Nutrient inputs to coastal and estuarine waters are important influences on the health of these waterbodies. 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 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). Algal blooms and associated die-offs can contribute to hypoxic conditions (most common during summer months), which can contribute to fish kills and associated reductions in marine populations (ISA, Appendix 10). Further, the prevalence and health of submerged aquatic vegetation (SAV), which is important habitat for many aquatic species, has been identified as a biological indicator for N enrichment in estuarine waters (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 SAV is 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).

The degree to which N enrichment and associated ecosystem impacts are driven by
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atmospheric N deposition varies greatly and is largely unique to the specific ecosystem. 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 ES 5.2; 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, and wastewater sources, in addition to atmospheric deposition across the watershed (ISA, section IS 2.2.2; ISA, Appendix 7, section 7.2.1).

There are estimates of atmospheric N loading to estuaries available from 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, however, it was the 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).

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).

(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 require more N. In this manner, the relative representation of different vegetation 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).

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 (ISA, Appendix 6, section 6.2.1). Nitrogen 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 increased productivity and changes in biodiversity associated with N addition to terrestrial ecosystems 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 (PA, section 4.1; ISA, Appendix 6). The organisms affected include trees, herbs and shrubs, and lichen, as well as fungal, microbial, and arthropod communities. 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 (PA, section 4.1; 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). New evidence for forest communities in this review indicates 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). The recent evidence also 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 associations between herb and shrub species richness and N deposition, that were highly dependent on soil pH (ISA, Appendix 6, section 6.3.3.2).

Recent evidence includes associations of variation in lichen community composition with N deposition gradients in the U.S. and Europe, (ISA, Appendix 6, section 6.3.7; 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). The newly available evidence 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).

Nitrogen 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).

d. Other Deposition-Related Effects

Additional categories of effects for which the current evidence is sufficient to infer causal relationships with deposition of S or N compounds or PM 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). 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, and factors that influence the process, such as oxygen content, temperature, pH, and carbon supply, which themselves vary temporally, seasonally, and geographically (ISA, Appendix 12, section 12.3). The currently available evidence is also 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). Sulfur deposition can contribute to sulfide and associated phytotoxicity in freshwater wetlands and lakes, with the potential to contribute to effects on plant community composition in freshwater wetlands (ISA, Appendix 12, section 12.2.3).

With regard to PM deposition, 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). 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, when deposited to ecosystems, 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. Studies involving ambient air PM, however, have generally involved conditions that would not be expected to meet the current secondary standards for PM. Further, 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 its chemical components, such as trace metals, which can be toxic in large amounts (ISA, Appendix 15, sections 15.2 and 15.3.1). The evidence

largely comes from studies involving areas experiencing elevated concentrations of PM, such as near industrial areas or historically polluted cities (ISA, Appendix 15, section 15.4).

2. 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 regarding effects on the public welfare that have been made in NAAQS reviews.

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.

The categories of effects identified in the CAA to be included among welfare effects are quite diverse, 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 section II.C.1 above (ISA, sections IS.5 to IS.9). As noted in the last review of the secondary NAAQS for NO_x 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 (O₃), judgments regarding public welfare significance have given particular attention to effects in areas with special Federal protections (such as Class I areas), 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).³⁴ In the 2015 O₃ 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 O₃-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 (Constanza et al., 2017; ISA, section IS.13). 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).

³⁴ For example, the fundamental purpose of parks in the National Park System “is to conserve the scenery, natural and historic objects, and wildlife in the System units and to provide for the enjoyment of the scenery, natural and historic objects, and wildlife 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)).

In the last review of the secondary NAAQS for NO_x 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 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 section II.C.1 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).

In acid-impacted surface waters, acidification primarily affects the diversity and abundance of fish and other aquatic life, and the ecosystem services derived from these organisms. (2011 PA, section 4.4.5). In addition to other types of services, fresh surface waters support several cultural services, such as aesthetic, recreational, 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. 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, however, 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 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). Since the 2012 review, additional approaches and methods have been applied to estimate the potential effects of aquatic acidification on uses and services of affected aquatic ecosystems; with regard to economic impacts, however, “for many regions and specific services, poorly characterized dose-response between deposition, ecological effect, and services are the greatest challenge in developing

specific data on the economic benefits of emission reductions” (ISA, Appendix 14, p. 14-23).

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). Studies reviewed in the ISA have explored both enumeration of the number of ecosystem services that may be affected by N loading, and the pathways by which this may occur, as well as approaches to valuation of such impacts. A finding of one such analysis was that “better quantitative relationships need to be established between N and the effects on ecosystems at smaller scales, including a better understanding of how N shortages can affect certain populations” (ISA, Appendix 14, sections 14.5 and 14.6). The relative contribution of atmospheric deposition to total N loading varies widely among estuaries, however, and has declined in more recent years (ISA, Appendix 10, section 10.10.1).

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 plants, that may be agricultural and forest crops. Such increased growth and yield may be judged and valued differently than changes in growth of other species. As noted in section II.C.1 above, 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. In considering such services in past reviews for secondary standards for other pollutants (e.g., O₃), 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 O₃ including timber, produce grown for consumption and horticultural plants used for landscaping" (80 FR 65403, October 26, 2015).

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 composition³⁵) to broader effects on the public welfare. The ISA finds that while there is an improved understanding from information available in this review of the number of

³⁵ As recognized in section II.C.1.c above, lichen communities have important roles in ecosystem function, such as in hydrologic cycling, nutrient cycling, and as sources of food and habitat for other species (ISA, Appendix 6).

pathways by which N and S deposition may affect ecosystem services, most of these relationships remain to be quantified (ISA, Appendix 14, section 14.6).³⁶ This gap creates uncertainties when considering the public welfare implications of some biological or geochemical responses to ecosystem acidification or N enrichment, and accordingly complicates 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 with regard to assessment of effects associated with existing deposition rates is that the current, much-improved air quality and associated reduced deposition is 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, affecting ecosystem responses.

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.

³⁶ While “there is evidence that N and S emissions/deposition have a range of effects on U.S. ecosystem services and their social value” and “there are some economic studies that demonstrate such effects in broad terms,” “it remains methodologically difficult to derive economic costs and benefits associated with specific regulatory decisions/standards” (ISA, Appendix 14, pp. 14-23 to 14-24).

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.

3. Exposure Conditions and Deposition-Related Metrics

The ecological effects identified in section II.C.1 above vary widely with regard to the extent and level of detail of the available information that describes the exposure circumstances that may elicit them. The information for direct effects of SO_x, N oxides and PM in ambient air is somewhat more straight-forward to consider as it is generally in terms of concentrations in air. For deposition-related effects, the information may be about S and N compounds in soil or water or may be for metrics intended to represent atmospheric deposition of those compounds. For the latter, as recognized in section II.A.3 above, we face the challenge of relating that information to patterns of ambient air concentrations.

With regard to the more complex consideration of deposition-related effects such as ecosystem acidification and N enrichment, there is also 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 (in 1971), 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 faced in identifying deposition targets that might be expected 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.

Critical loads (CLs) 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. 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 result in an ecological effect of interest, has multiple interpretations and applications (ISA, p. IS-14). The dynamic nature of ecosystem pollutant processing and the broad array of factors that influence it adds complications to critical load identification and interpretation. Time is an important dimension, which is sometimes unstated, (e.g., in empirical or observational analyses) and is sometimes explicit (e.g., in steady-state or dynamic modeling analyses) (ISA, section IS.2.2.4). Further, this variety in meanings stems in part from differing judgments and associated identifications regarding the ecological effect (both type and level of severity) on which the critical load focuses, and judgment of its significance or meaning.

Studies, based on which CLs are often identified, vary widely with regard to the specific ecosystem characteristics being evaluated (or judged), 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, and associated uncertainty, of using critical load information from such studies for different applications. The summary that follows 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 aquatic acidification REA in this review, presented in the PA and summarized in section II.D below. In those analyses, the concept of a critical load is employed with steady-state modeling that relates deposition to waterbody acid neutralizing capacity.

While recognizing the inherent connections between watersheds and waterbodies, such as lakes and streams, the organization of this section 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 (PA, Chapter 5). Further, we recognize the generally greater role of atmospheric deposition in waterbodies impacted by aquatic acidification compared to its role in eutrophication-related impacts of surface waters, particularly rivers and estuaries in and downstream of populated watersheds, to which direct discharges have also long contributed, as recognized in section II.C.1.c(1) above (ISA, Appendix 13, section 13.1.3.1; ISA, Appendix 7, section 7.1.1.1; 2008 ISA, section 3.2). Therefore, with regard to deposition-related effects, we focus first on the quantitative information for aquatic ecosystem effects in sections II.C.3.a. Section II.C.3.b 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 II.C.3.c 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.

a. Acidification and Nitrogen Enrichment in Aquatic Ecosystems

Prior to the peak in S deposition levels that occurred in the 1970s and early 1980s, surface water sulfate concentrations were increasing in response to the extremely high S deposition of the preceding years. Subsequently, and especially more recently, surface water sulfate concentrations have generally decreased, particularly in the Northeast (Robinson et al., 2008; ISA, section 7.1.5.1.4). Some studies of long-term projections in some waterbodies (e.g., in the Blue Ridge Mountains region in Virginia), however, continue to indicate little or slow reduction in acidic ions, even as emissions have 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,³⁷ which some modeling has suggested will delay chemical recovery in those water bodies (ISA, Appendix 7, sections 7.1.2.2 and 7.1.5.1).

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

³⁷ Some modeling studies in some areas have indicated the potential for a lagged response even as emissions and deposition decline; this lag reflects a reduction in soil absorption of SO_4^{2-} and leaching of previously accumulated S from watersheds (ISA, Appendix 7, section 7.1.2.2).

review, the significance of non-air N loading to rivers, estuaries and coastal waters (as recognized in section II.C.1 above) continues to complicate the assessment of nutrient enrichment-related risks specifically related to atmospheric N deposition. Accordingly, the REA analyses developed in this review focus on aquatic acidification. The REA and its findings regarding deposition rates associated with different levels of aquatic acidification risk are summarized in section II.D below. Thus, the paragraphs below focus on the available quantitative information regarding atmospheric deposition and N 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, based on which the ISA has made determinations of causality; 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 II.C.1 above.³⁸ Separate quantitative analyses have not been performed in this review for N enrichment-related effects in these waterbodies in 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. Rather, we have reviewed the evidence as characterized in the ISA with regard to deposition level estimates that studies have related to various degrees of different effects with associated differences in potential for or clarity in public welfare significance. The overview provided here draws on the summary in the PA (PA, section

³⁸ 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). While the evidence was sufficient to support causal determinations between S deposition and these effects, quantitative information to support quantitative analysis in this review.

5.2).

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 less informative for other uses, 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 section II.C.1 above.

There is also considerable information available for estuaries and coastal systems. The relationship between N loading and algal blooms, and associated water quality impacts, has led to numerous water quality modeling projects, over the past few decades, that have quantified eutrophication processes in multiple estuaries, near coastal marine ecosystems and large river systems, to relate N loading to various water quality indicators to inform water quality management decision-making in multiple estuaries, including Chesapeake Bay, Narraganset Bay, Tampa Bay, Neuse River Estuary and Waquoit Bay (ISA, Appendix 7, section 7.2). These projects often use indicators of nutrient enrichment, such as chlorophyll a, dissolved oxygen, and reduced 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).

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 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.

The establishment of target N loads to surface waterbodies is in many areas related to implementation of the total maximum daily load (TMDL) requirements of section 303(d) of the Clean Water Act.³⁹ 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 phosphorus 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

³⁹ 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).

control of urban sprawl (2008 ISA, section 3.3.8.3). Studies since 2000 estimate atmospheric deposition as a major N source in the overall N budget for Chesapeake Bay⁴⁰ (ISA, section 7.2.1; Howarth, 2008; Boyer et al., 2002). The TMDL established for Chesapeake Bay in 2010, under requirements of section 303(d) of the Clean Water Act, included a loading allocation for atmospheric deposition of N directly to tidal waters, 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).⁴¹

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 Bay, at 20% of the total (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).⁴² Another modeling study in the Waquoit Bay estuaries in Cape Cod, Massachusetts, using data since 1990, estimated

⁴⁰ For example, a 2011 analysis estimated atmospheric deposition to the Chesapeake Bay watershed to account for approximately 25% of total N inputs to the estuary (ISA, Appendix 7, section 7.2.1).

⁴¹ As recognized on the EPA webpage describing this activity, the TMDL, formally established in December 2010 “is designed to ensure that all pollution control measures needed to fully restore the Bay and its tidal rivers are in place by 2025.” The website also indicates that “EPA expects practices in place by 2017 to meet 60 percent of the necessary reductions,” and for some areas to recover before others, but for it to take years after 2025 for the Bay and its tributaries to fully recover (<https://www.epa.gov/chesapeake-bay-tmdl/frequent-questions-about-chesapeake-bay-tmdl>).

⁴² For example, across the 74 estuaries in the 3-state coastal region studied, N from atmospheric deposition to estuary watersheds was generally estimated to account for less than 25% of total N inputs, while estimates for a few small estuaries in CT were higher than 51% (but below 75%) (Latimer and Charpentier, 2010).

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 N loading from multiple sources on nutrient enrichment⁴³ and associated water quality indicators, including chlorophyll a (ISA, Appendix 10, section 10.2).

Nitrogen loading to estuaries has also been considered specifically 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 include flushing and drainage in estuaries (ISA, Appendix 10, section 10.6).

b. Deposition-Related Effects in Terrestrial Ecosystems

The evidence of atmospheric deposition contributing to acidification and N enrichment-related effects in terrestrial systems is strong, as evidenced by the causal determinations made in the ISA and summarized in section II.C.1.b(2) and II.C.1.c(2) above. Accordingly, the

⁴³ One evaluation of progress in achieving mandated N reductions in the Neuse River Basin in NC found that flow-normalized N loading from NO₃⁻ decreased beginning in the 1992-1996 period (ISA Appendix 10, section 10.2; Lebo et al., 2012).

subsections below describe the available information in this review, including that available in the last review, regarding quantitative relationships between atmospheric deposition rates and specific terrestrial effects of interest.

The terrestrial analyses in the 2012 review included 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) for terrestrial acidification 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 analyses in the 2009 REA, and on more recent 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 with investigation into various assessment approaches.

The subsections below discuss the available studies of deposition and risk to trees (section II.C.3.b(1)) and to herbs, shrubs, and lichens in section II.C.3.b(2). 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, each with their various design features and associated strengths and limitations (as noted immediately below), inform consideration of 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 use 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 risk, 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 recovery 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 value). 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) have been considered, with key findings summarized below.

(1) Deposition and Risks to Trees

The available quantitative information regarding S and N deposition and effects on trees including modeling applications, both from the 2012 review and from studies, is described in the ISA. Steady-state modeling analysis performed in the 2009 REA estimated annual amounts of acid deposition at or below which one of three BC:Al target values would be met in a 24-state area in which the acid-sensitive species, red spruce and sugar maple, occur. A range of acid deposition was estimated for each of the three target values. Recent estimates of total S and N deposition in regions of the U.S. appear to meet all but the most restrictive of these targets, for which the uncertainty is greatest (e.g., 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 target values are based (PA, section 5.3.2). There are also 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). More recent publications have employed a new approach to estimating these parameters, including the weathering parameter (BCw), which reduced the uncertainty associated with this parameter (e.g., Phelan et al., 2014; McDonnell et al., 2012; ISA, Appendix 4, sections 4.6.2.1 and 4.8.4 and Appendix 5, section 5.4).

Experimental addition studies of S, or S plus N have been performed in eastern locations and focused on a small set of species, including sugar maple, aspen, white spruce, yellow poplar, and black cherry; these studies generally have not reported growth effects (PA, Appendix 5B, section 5B.3.1; ISA, Appendix 6, sections 6.3.4, 6.3.5 and Table 6-21). 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, e.g., greater than 6 kg N/ha-yr (PA, 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. 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 (PA, Table 5B-1; Magill et al., 2004; McNulty et al., 2005; Pregitzer et al., 2008; 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.

With regard to S deposition, the two large observational 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., 2018⁴⁴). Interestingly,

⁴⁴ The study by Horn et al. (2018) constrained the S analyses to preclude a positive association with S.

survival for the same 9 species groups was also negatively associated with long-term average ozone (Dietze and Moorcroft, 2011). 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 roughly 10-year durations. 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 $\text{kg S ha}^{-1}\text{yr}^{-1}$. The second study covered the more recent time period (2000-2013) and 71 species distributed across the U.S. To draw on this study with regard to S deposition levels of interest, the distribution of S deposition estimates for each species were considered in the PA (Appendix 5B, section 5B.2 and Attachments 2A and 2B). In this national-scale analysis, the range of median S deposition for sites of those species for which negative associations with growth or survival were reported, was 5 to 12 $\text{kg S ha}^{-1}\text{yr}^{-1}$, with few exceptions (Horn et al., 2018).⁴⁵

Regarding N deposition, the three large observational 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 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

⁴⁵ This range is for median S deposition estimates (based on measurement interval average, occurring within 2000-2013) of nonwestern species with negative associations with growth or survival ranged (Horn et al., 2018).

than 10 years. The full range of average NO₃ 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⁻¹yr⁻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⁻¹yr⁻¹ (Horn et al., 2018).

In considering what can be drawn from these studies with regard to deposition levels of potential interest for tree species effects, such as the ranges identified above, a number of uncertainties are recognized. For example, several factors were not accounted for that have potential to influence tree growth and survival. Although O₃ 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. 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 (PA, Appendix 5B, Table 5B-6). 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. 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 deposition patterns across the full time period 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 (e.g., PA, Appendix 5B, Figures 5B-9 through 5B-12).⁴⁶ The appreciable differences in deposition magnitude across the time periods also contribute uncertainty to interpretations related to specific magnitudes of deposition associated with patterns of tree growth and survival. There are few studies of recovery in historically impacted areas in the ISA that might address such uncertainties (e.g., ISA, section IS.4.1, IS.5.1, IS.11.2).

(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

⁴⁶ Sulfur deposition in the U.S. across the full period of these studies (1994-2013) generally exhibited a consistent pattern of appreciable declines, with study plots, particularly in the East, having experienced decades of much higher S deposition in the past. Similarly, N deposition during the combined time period of the studies (1994-2013) has also changed, with many areas experiencing declines and a few areas experiencing deposition increases for some N species and in total N (PA, section 6.2.1).

the order of 6 kg N/ha-yr) and higher (PA, section 5.3.3.1). 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 6.5 kg/ha-yr and low pH sites. In forested sites, negative associations were found above 11.6 kg N/ha-yr in sites with acidic soil pH at or above 4.5 (PA, section 5.3.3).

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, decades of S and N deposition— and elevated levels of airborne pollutants, including ozone and nitrogen oxides, in the U.S. — have affected the ecosystems studied; and these studies generally have not accounted for the influence of historical deposition on the associations observed with more recent deposition metrics. Further, given that observational studies occur 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 elicit specific responses. A number of factors limit such interpretations of the currently available studies (PA, section 5.3.3.2). These factors include uncertainties related to the methods employed to represent N deposition, the potential role of other unaccounted-for environmental factors (including O₃, SO₂, 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 information on exposure conditions associated with effects on lichen species of oxides of N such as HNO₃ in section II.C.3.c below.

c. Direct Effects of N Oxides, SO_x and 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 SO₂ and NO₂ associated with effects on plants, concentrations of NO₂ and HNO₃ 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 SO₂ and HNO₃ 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 SO₂, 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 HNO₃ (than to NO₂). The laboratory and field studies of

oxides of N vary regarding 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, the elevated concentrations of NO₂ and HNO₃ in the Los Angeles area in the 1970s-90s are well documented as is the decline of lichen species in the Los Angeles Basin during that time (PA, section 5.4.2).⁴⁷ Such an assessment of lichen species and elevated concentrations of N oxides is not available for other locations across the U.S.

Ecological effects associated with 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 SO₂ emissions. These studies, however, do not generally report the SO₂ 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 for direct effects 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). 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). Few studies report yield effects from acute exposures, with the available ones reporting

⁴⁷ For example, concentrations of HNO₃ reported in forested areas of California in the 1980s ranged up to 33 ug/m³, and annual average NO₂ concentrations in the Los Angeles area ranged from 0.078 ppm in 1979 to 0.053 ppm in the early 1990s (PA, section 5.4.2). Ambient air concentrations of HNO₃ in the Los Angeles metropolitan area have declined markedly, as shown in Figure 2-23 of the PA, which compares concentrations at CASTNET monitoring sites between 2019 and 1996 (PA, section 2.4.1).

relatively high concentrations, such as multiple hours with concentrations above 1 ppm (1982 AQCD, section 8.3). Effects have also been reported on photosynthesis and other functions in a few lichen species groups, although recovery of these functions was observed from short, multi-hour exposures to concentrations below about 1 ppm (ISA, Appendix 3, section 3.2).

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 NO₂ concentrations below 0.1 ppm (ISA, Appendix 3, section 3.3). The information is more limited regarding exposures to other oxides of N.

The evidence for HNO₃ includes controlled exposure studies describing foliar effects on several tree species. Nitric acid has also been found to deposit on and bind to the leaf or needle surfaces. Studies of ponderosa pine, white fir, California black oak and canyon live oak involving continuous chamber exposure over a month to 24-hour average HNO₃ concentrations generally ranging from 10 to 18 µg/m³ (moderate treatment) or 18 to 42 µg/ m³ (high treatment), with the average of the highest 10% of concentrations generally ranging from 18 to 42 µg/ m³ (30-60 µg/ m³ peak) or 89 to 155 µg/ m³ (95-160 µg/ m³ 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 HNO₃ concentration of 33 ug/ m³ in August 1986 (Padgett et al., 2009; Bytnerowicz and Fenn, 1996), when annual average NO₂ concentrations in the Basin ranged up to 0.058 ppm (U.S. EPA, 1987).

In addition to the observational studies of lichen communities in the Los Angeles Basin impacted by ambient air concentrations from the 70s, the available evidence for lichens includes a recent laboratory study, involving daily HNO₃ exposures for 18 to 78 days, with daily peaks

near 50 ppb ($\sim 75 \mu\text{g}/\text{m}^3$) 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). 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 (PA, Appendix 5B, section 5B.4.1). Over the last several decades, the magnitude of both dry deposition of HNO_3 and annual average HNO_3 concentration in this area, and nationally, and the spatial extent of high deposition rates and concentrations have dramatically declined (PA, Figure 2-23; ISA, Appendix 2, Figure 2-60). 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 and the extent to which similar atmospheric conditions and ecological relationships exist in other locations in the U.S. is uncertain.

Little information is available on welfare effects of airborne PM at concentrations

commonly occurring in the U.S. today, and the available information does not indicate effects to occur under such conditions. The extent to which quantitative information is available for airborne PM concentrations associated with ecological effects varies for the different 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 (ISA, Appendix 15, sections 15.4.3 and 15.4.6). Studies involving ambient air PM have generally involved conditions that are much higher than those common to the U.S. today (ISA, Appendix 15, sections 15.4.3 and 15.4.4).

D. Quantitative Exposure/Risk Assessment for Aquatic Acidification

Our consideration of the scientific evidence available in the current review is informed by results from quantitative analyses of estimated acidic deposition and associated risk of aquatic acidification (PA, section 5.1 and Appendix 5A). These REA analyses, like those in the last review, make use of well-established modeling tools and assessment approaches for this endpoint. Other categories of effects of S and N deposition have been the subject of quantitative analyses, both in the last review (e.g., terrestrial acidification) and in other contexts (e.g., eutrophication of large rivers and estuaries), each with associated complexities and specificity (section II.C.3 above). The PA while focusing the new analyses on aquatic acidification risks, as summarized here, also draws on findings of available analyses for the other categories of effects (e.g., as summarized in section II.C.3 above).

The REA analyses, summarized here and presented in detail in Appendix 5A of the PA, have focused on ANC as an indicator of aquatic acidification risk (PA, section 5.1 and Appendix

5A). This focus is consistent with such analyses performed in the 2012 review and with the longstanding evidence that continues to demonstrate a causal relationship between S and N deposition and alteration of freshwater biogeochemistry and between acidifying S and N deposition and changes in biota, including physiological impairment and alteration of species richness, community composition, and biodiversity in freshwater ecosystems (ISA, Table ES-1), as summarized in section II.C.1 above.

Section II.D.1 summarizes key aspects of the assessment design, including the conceptual approach and tools, indicator reference or benchmark concentrations, the assessment scales, study areas and waterbodies analyzed, and exposure and risk metrics derived. Key limitations and uncertainties associated with the assessment are identified in section II.D.2 and the exposure and risk estimates are summarized in section II.D.3. An overarching focus of these analyses is characterization of aquatic acidification risk in sensitive ecoregions associated with different deposition conditions.

1. Key Design Aspects

The REA for this review entailed a multi-scale analysis of waterbodies in the contiguous U.S. that assessed waterbody-specific aquatic acidification at three spatial scales: national, ecoregion, and case study area (PA, Appendix 5A). The assessment involved evaluation of deposition and water quality response at the waterbody site level, which has been summarized at the national, ecoregion, and case study level. The national-scale analysis involved waterbody

sites across the U.S. for which relevant data were available.⁴⁸ The ecoregion-scale analysis focused on waterbodies in a set of 25 ecoregions generally characterized as acid-sensitive; and the more localized case study-scale analysis focused on waterbodies in five case study areas across the U.S., within each of which were Class I areas.

The impact of acidifying S or N deposition estimated for five different time periods (2001-03, 2006-08, 2010-12, 2014-16 and 2018-20) was evaluated using a CL approach that relied on waterbody location-specific CL estimates derived for other applications and available in the National Critical Loads Database (NCLD)⁴⁹ (PA, Appendix 5A). The CL estimates used in the assessment were largely based on steady-state modeling, and the modeling applications focused on ANC, producing CL estimates (acidifying deposition in terms of kg/ha-yr or meq/m²-yr [milliequivalents per square meter per year] for S and N compounds) for different ANC concentrations. Of the 84 ecoregions in the contiguous U.S., 64 have at least one waterbody site with a CL estimate (PA, Appendix 5A). Given its common use in categorizing waterbody sensitivity, ANC was used as the indicator of acidification risk in this assessment (PA, section 5.1.2.2). Deposition estimates, as 3-year averages of annual TDep estimates, were compared to

⁴⁸ The national-scale analysis focused on the contiguous U.S. as there are insufficient data available for Hawaii, Alaska, and the territories. Of the four hierarchical levels of ecoregion categorization, the REA utilized level III which divides the contiguous U.S. into 84 ecoregions (Omernik and Griffith, 2014). The 69 of these 84 ecoregions in which there was at least one site with sufficient data comprised the national scale.

⁴⁹ 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 Model of Acidification of Groundwater In Catchments (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). Data and CL estimates in the NCLD are generally focused on waterbodies impacted by deposition-driven acidification and are described in documentation for the database version (PA, section 5.1.2.3; Lynch et al., 2022).

the CL estimates for three different ANC benchmark concentrations (targets or thresholds), in recognition of the watershed variability and associated uncertainties, as an approach for characterizing aquatic acidification risk (PA, section 5.1).

The available evidence and scientific judgments were considered in identifying the three ANC benchmark concentrations: 20 $\mu\text{eq/L}$, 30 $\mu\text{eq/L}$, 50 $\mu\text{eq/L}$ (PA, section 5.1.2.2). Selection of these benchmark ANC concentrations reflects several considerations. For example, most aquatic CL studies conducted in the U.S. since 2010 use an ANC of 20 and/or 50 $\mu\text{eq/L}$, because 20 $\mu\text{eq/L}$ has been suggested to provide protection for a “natural” or “historical”⁵⁰ range of ANC, and 50 $\mu\text{eq/L}$ to provide greater protection, particularly from episodic acidification events⁵¹ (Dupont et al., 2005; Fakhraei et al., 2014; Lawrence et al., 2015; Lynch et al., 2022; McDonnell et al., 2012, 2014; Sullivan et al., 2012a, 2012b). For example, levels below 20 $\mu\text{eq/L}$ have been associated with fish species reductions in some sensitive waterbodies of the Shenandoah and Adirondack Mountains. Levels of ANC ranging from 30 to 40 $\mu\text{eq/L}$ have been reported to provide sufficient buffering to withstand acidic inputs associated with episodic springtime rain or snowmelt events. An ANC value of 50 $\mu\text{eq/L}$ has often been cited in the literature as a target for many areas, and in the 2012 review, ANC values at or above 50 $\mu\text{eq/L}$ were described as providing an additional level of protection although with increasingly greater

⁵⁰ For example, dynamic modeling simulations in acid-sensitive streams of the southern Blue Ridge Mountains have predicted all streams to have pre-industrial time ANC levels above 20 $\mu\text{eq/L}$, while also predicting more than a third of the streams to have pre-industrial ANC levels below 50 $\mu\text{eq/L}$ (Sullivan et al., 2011).

⁵¹ As noted in section II.C.1 above, events such as spring snowmelt and heavy rain events can contribute to episodic acidification events. For example, in some impacted northeastern waterbodies, particularly headwater streams, 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).

uncertainty for values at/above 75 $\mu\text{eq/L}$ ⁵² (2011 PA, pp. 7-47 to 7-48). 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 $\mu\text{eq/L}$ (Williams and Labou, 2017; Shaw et al., 2014). For these reasons, this assessment, also develops results for an ANC threshold of 50 $\mu\text{eq/L}$ for sites in the East and 20 $\mu\text{eq/L}$ for sites in the West (denoted as “50/20” $\mu\text{eq/L}$).⁵³ Thus, the set of benchmark concentrations used in this REA include ANC concentrations that are naturally occurring in many areas and also include concentrations that, depending on watershed characteristics, may provide additional buffering in times of episodic acidification events.

Since acidification of waterbodies is controlled by local factors such as geology, hydrology, and other landscape factors, aquatic CLs for acidification were determined at the waterbody level (based on site-specific data) and then summarized at the national, ecoregion, and case study level. National-scale analyses were performed using two approaches: one considering acid deposition of N and S compounds combined and one for S deposition only. Findings from these analyses indicated that across the five different time periods analyzed, the percent of waterbodies exceeding their CLs was similar for the two approaches (PA, Appendix 5A, sections 5A.1.6.2 and 5A.2.1). Thus, to facilitate interpretation of the results, further analysis of the results focused on the findings for S only deposition.

⁵² In considering higher ANC levels (e.g., up to 80 $\mu\text{eq/L}$ and higher), it was also recognized that many waterbodies, particularly in acid-sensitive regions of contiguous never had an ANC that high and would never reach an ANC that high naturally (Williams and Labou 2017; Shaw et al., 2014; PA, section 5.1.2.2). Additionally, in conveying its advice in the 2012 review, the CASAC expressed its view that “[l]evels of 50 $\mu\text{eq/L}$ 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 $\mu\text{eq/L}$ ” (Russell and Samet, 2010a; pp. 15-16).

⁵³ This approach is also used in multiple studies and the NCLD (PA, section 5.1.2.2).

Critical load estimates for specific waterbody sites across the contiguous U.S. were drawn from the NCLD (version 3.2.1)⁵⁴ for comparison to total deposition estimates in the same locations for the five time periods. Comparisons were only performed for sites at which CL estimates were greater than zero, indicating that achievement of the associated target ANC would be feasible.⁵⁵ The results of these analyses are summarized with regard to the spatial extent and severity of deposition-related acidification effects and the protection from these effects associated with a range of annual S deposition.

The ecoregion-scale analyses focused on 25 ecoregions,⁵⁶ 18 in the East and 7 in the West. Ecoregions are areas of similarity regarding patterns in vegetation, aquatic, and terrestrial ecosystem components. The 25 ecoregions in this analysis each had more than 50 waterbody sites (or locations) for which a prior modeling application had developed a CL estimate, which was available in the NCLD (PA, section 5A.2.2.2). Although a total of 32 ecoregions had more than 50 CL sites,⁵⁷ four in the West were excluded as having very low deposition that resulted in no CL exceedances across the complete 20-year analysis period. An additional three ecoregions

⁵⁴ 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, while the CL values were averaged when the publications are from the same timeframe (PA, Appendix 5A, section 5A.1.5).

⁵⁵ Critical load estimates are estimates of the S deposition rate at which a particular waterbody site is estimated to be able to achieve a specified ANC level. A CL estimate at or below zero would indicate that no S deposition estimate would provide for such a result.

⁵⁶ The ecoregion classification scheme used to group waterbody sites into ecoregions is based on that described in Omernik (1987), which 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 (e.g., geology, physiography, vegetation, climate, soils, land use, wildlife, and hydrology).

⁵⁷ In light of the size of the level III ecoregions, 50 was identified as an appropriate minimum number of CL sites within an ecoregion to include it in the analysis.

(i.e., Southeastern Plains , Southern Coastal Plain , and Atlantic Coastal Pine Barrens) were excluded as they are known to have naturally acidic surface waters, and the low CL estimates for these ecoregions (and resulting CL exceedances) are likely driven by natural acidity linked to high levels of dissolved organic carbon, hydrology, and natural biogeochemical processes rather than atmospheric deposition (2008 ISA, section 3.2.4.2; Baker et al., 1991; Herlihy et al., 1991).

The case study scale represents the smallest scale at which CLs and their comparison to deposition estimates were summarized and is intended to give some insight into potential local impacts 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 (details presented in PA, section 5.1.3.3 and Appendix 5A, section 5A.2.1). These areas include a number of national parks and forests that vary in their sensitivity to acidification, but represent high value or protected ecosystems, such as Class 1 areas, wilderness, and national forests (PA, Appendix 5A, section 5A.2.1). The most well studied of these, the Shenandoah Valley Area case study, includes the Class I area, Shenandoah National Park, and waterbodies in each of three ecoregions. The number of waterbody sites with CLs available in the NCLD for the Shenandoah study area (4,977 sites) is nearly an order of magnitude greater than the total for the four other areas combined (524 sites).

The analyses at different scales differed in how results were summarized and evaluated. For example, at the national scale, percentages of water bodies with deposition estimates exceeding their CLs were reported for each of the five time periods for which deposition was assessed. From the case study scale analyses, we focused primarily on the distribution of CL estimates in each study area. In so doing, the CLs for each case study area were characterized in

terms of the average, and two lower percentiles (e.g., the 30th percentile CL, which is the value

below 70% of the CL estimates for that study area, and the 10th percentile).

In the ecoregion-scale analyses, percentages of waterbody sites per ecoregion that exceeded their estimated CLs and percentages of waterbody sites that fell at or below them – for each of the three ANC thresholds - were summarized by ecoregion for each of the five time periods: 2001-2003, 2006-2008, 2010-2012, 2014-2016 and 2018-2020 (PA, section 5.1.3.2 and Appendix 5A, section 5A.2.2). Percentages of waterbody sites that did not exceed their estimated CLs were described as achieving the associated ANC threshold (or target). These results were then considered in two ways. The first is based on a binning of this dataset of ecoregion-time period combinations and percentages by ecoregion median deposition levels (at/below 5 kg/ha-yr, at/below 6 kg/ha-yr, etc). The second approach involved summarizing ecoregion-specific trends in percentage of waterbodies per ecoregion estimated to achieve the three threshold or target ANC values (or estimated to exceed the associated CLs).

2. Key Limitations and Uncertainties

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 (PA, Appendix 5A, section 5A.3). The mainly qualitative approach used to characterize uncertainty here and in quantitative analyses in other NAAQS reviews is described by World Health Organization (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.

Two types of quantitative analyses informed our understanding of the variability and uncertainty associated with the CL estimates developed in this assessment and support the overall uncertainty characterization. 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 note 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, 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, such as soil type. 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 Steady-State Water Chemistry (SSWC) 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 (PA, 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 BC_w 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 (PA, 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 for S CL estimates ranged from 0.1 kg S/ha-yr at the 5th percentile to 5.3 kg S/ha-yr at the 95th percentile. Smaller 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, which are areas for which there are relatively larger site-specific datasets (e.g., for water quality parameters). 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 the vast majority of CLs (those based on the widely used steady-state water chemistry model) was 7.68 meq S/m²-yr or 1.3 kg S/ha-yr, giving a confidence interval of ± 3.84 meq/m²-yr or ± 0.65 kg S/ha-yr. While a comprehensive analysis of uncertainty had not been completed for these estimates prior to this assessment, judgment by EPA experts suggested the uncertainty for combined N and S CLs to be on average about ± 0.5 kg/ha-yr (3.125 meq/m²-yr), which is generally consistent with the range of uncertainty determined from this quantitative uncertainty analysis (PA, Appendix 5A, section 5A.3).

At the ecoregion scale, 51 ecoregions had sufficient data to calculate the 5th to 95th percentile (PA, 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, Blue Ridge, Northern Lakes and Forests, and North Central Appalachians) and Rockies (e.g., Sierra Nevada, Southern Rockies, and Idaho Batholith). Ecoregions with more uncertain CLs included the Northeastern Coastal Zone, Cascades, Coast Range, Interior Plateau, and Klamath Mountains/California High North Coast Range.

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., 2012b;

McDonnell et al., 2014), and (3) Dynamic Models (Model of Acidification of Groundwater In Catchments [MAGIC]) or Photosynthesis EvapoTranspiration Biogeochemical model [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 PA Appendix 5A, section 5A.3.1, 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 (PA, Appendix 5A, section 5A.3).

3. Summary of Results

The findings from the aquatic acidification REA are summarized in terms of S deposition due to the finding of a negligible additional influence of N deposition compared to S deposition on acidification in this assessment⁵⁸ (Appendix 5A, section 5A.2.1). As summarized more fully below, the analyses of five case study areas, including the acidification-impacted Shenandoah Valley area, indicate that with annual average S deposition below 12 and 10 kg/ha yr, the average waterbody in each area (average as to acid-sensitivity) would be estimated to achieve the ANC benchmarks of 20 and 50 $\mu\text{eq/L}$, respectively. Seventy percent of waterbodies in each area would be estimated to achieve these benchmarks with deposition below 10 and 7 kg/ha-yr, respectively. At the ecoregion-scale, the results from the analysis of 25 ecoregions, dominated by

⁵⁸ More specifically, the percentage of waterbodies across the contiguous U.S. estimated to exceed a CL for combined total S and N are very similar or just slightly higher (e.g., by 1-2%) than S only percentages of the waterbodies estimated to not meet the ANC benchmarks. This indicates that most of the N deposition entering the watershed is retained within the watershed and/or converted to gaseous N (PA, Appendix 5A, section 5A.2.1).

acid-sensitive waterbodies, indicate acid buffering capabilities to have improved substantially over the past 20 years, and particularly between the first and second decades of the period. By the 2010-2012 period, the percentages of waterbodies achieving the three ANC benchmarks in all 25 ecoregions exceeded 80%, 80% and 70% (for 20, 30 and 50 $\mu\text{eq/L}$, respectively). By the subsequent analysis period (2014-2016), these percentages were 90%, 80% and 80%. The ecoregion median annual average deposition in all 25 ecoregions was below 8 kg/ha-yr for 2010-2012 and below 5 kg/ha-yr for 2014-2016. An alternate approach to analyzing these estimates (for the 25 ecoregions across all five time periods) suggested that the three ANC benchmarks could be met in more than 80%, 80% and 70% of waterbodies per ecoregion in all ecoregions and time periods for which annual average ecoregion median deposition is estimated to be at or below 7 kg/ha-yr.

Between the three-year period of 2000-2002, which was the analysis year for the 2009 REA, and 2018-2020, the latest period considered in the REA for this review, national average sulfur deposition has declined appreciably 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 2009 REA, in the national-scale analysis of the current REA, few waterbody sites are estimated to be receiving deposition in excess of their CLs 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 the current 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 $\mu\text{eq/L}$ is less than 5% (Table 1; PA, Table 5-1).

Table 1. Percentage of Waterbodies Nationally for which Annual Average S Deposition During the Five Time Periods Assessed Exceed the Waterbody CL (for CLs Greater than 0) for each of the Specified ANC Targets

ANC (µeq/L)	2001-2003	2006-2008	2010-2012	2014-2016	2018-2020
20	22%	16%	5%	3%	1%
30	25%	19%	7%	4%	2%
50	28%	24%	11%	6%	4%
50/20*	28%	23%	10%	6%	4%
* This combination refers to the use of a target of 50 µeq/L in eastern ecoregions and 20 µeq/L in western ecoregions.					

The case study analyses provide estimates of S deposition (with associated uncertainties) that might be expected to allow these geographically diverse locations to meet the three ANC targets (20, 30 and 50 µeq/L). 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,⁵⁹ 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 S/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.

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 findings for several target ANC levels (50, 30 and 20 µeq/L) and the temporal changes across the 20-year period, as described in the PA, section 5.1.3.2. For example, during the two most recent

⁵⁹ The term “average” here refers to the average CL estimated for the specified ANC across all sites with CL estimates in each case study area (PA, Table 5-6).

3-year periods, the ecoregion median S deposition estimates in 2014-16 were below 5 kg/ha-yr in all 25 ecoregions in the analysis and the estimates for 2018-20 were all below 4 kg/ha-yr (Table 2). 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.

Table 2. Summary of Ecoregion Medians Derived as Median of S Deposition Estimates at CL sites Within an Ecoregion

	Ecoregion Median* Total Sulfur Deposition (kg S/ha-yr)				
	2001-03	2006-08	2010-12	2014-16	2018-20
	<i>All 25 Ecoregions</i>				
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
	<i>18 Eastern Ecoregions</i>				
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
	<i>7 Western Ecoregions</i>				
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 the deposition estimates across each ecoregion's waterbody sites with CL estimates.					

The ecoregion-scale results (e.g., percentage of waterbodies per ecoregion estimated to achieve the various ANC targets, or alternatively to exceed the associated CLs) for the 18 eastern and 7 western ecoregions are summarized in two ways. One approach, summarized further below, is framed by the temporal trends in deposition, and the second approach is in terms of ecoregion-time period combinations, using ecoregion deposition estimates (medians of deposition estimates at waterbodies with CLs in each ecoregion) as the organizing parameter. For example, Table 3 presents the percentages of waterbody sites per ecoregion estimated to achieve

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 the 18 eastern ecoregions and five time periods, there are 90 ecoregion-time period combinations, and for each of these, there are waterbody percentages for each of the three ANC targets. In Table 3, the combinations are binned by the ecoregion median S deposition associated with that ecoregion and time period. As can be seen from this table, fewer than half of the eastern ecoregion-time period combinations had an S deposition estimate at or below 4 kg/ha-yr (Table 2).⁶⁰ Table 3 indicates that lower levels of S deposition are associated with improved ANC values and greater percentages of waterbodies expected to reach ANC targets. Across the ecoregion-time period dataset of CL exceedances for the three ANC targets for all 90 eastern ecoregion-time period combinations (for which ecoregion median S deposition was at or below 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 (Table 3). 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 (the lowest ecoregion median deposition bin that includes at least half of the full dataset), 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 µeq/L (Table 3; PA, Table 5-5).

⁶⁰ The ecoregion median S deposition in all seven of the western ecoregions in all five time periods were at or below 2 kg/ha-yr (PA, Table 5-4).

Table 3. 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 $\mu\text{eq/L}$ 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%	
		<i>ANC target of 20 $\mu\text{eq/L}$</i>					<i>ANC target of 30 $\mu\text{eq/L}$</i>					<i>ANC target of 50 $\mu\text{eq/L}$</i>					
2	10	100%	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%	100%	97%	100%	100%	100%	100%
4	41	100%	100%	100%	100%	100%	95%	100%	100%	100%	100%	100%	93%	98%	100%	100%	100%
5	51	96%	98%	100%	100%	100%	92%	98%	100%	100%	100%	100%	82%	94%	96%	98%	100%
6	59	93%	98%	100%	100%	100%	88%	98%	100%	100%	100%	100%	78%	93%	97%	98%	100%
7	63	92%	98%	100%	100%	100%	87%	97%	100%	100%	100%	100%	78%	92%	95%	98%	100%
8	67	87%	94%	100%	100%	100%	82%	91%	99%	100%	100%	100%	73%	87%	93%	96%	100%
9	69	87%	94%	100%	100%	100%	81%	91%	99%	100%	100%	100%	72%	87%	93%	96%	100%
10	73	85%	92%	99%	99%	99%	78%	89%	97%	99%	99%	99%	70%	85%	92%	95%	99%
11	76	83%	91%	97%	99%	99%	76%	88%	96%	99%	99%	99%	68%	83%	91%	95%	99%
12	79	81%	89%	95%	96%	97%	73%	86%	94%	96%	96%	96%	66%	81%	89%	92%	96%
13	81	80%	88%	95%	96%	98%	73%	85%	94%	96%	96%	96%	65%	80%	88%	93%	96%
14	84	77%	86%	93%	95%	96%	70%	83%	92%	94%	95%	95%	63%	79%	86%	90%	94%
15	86	76%	84%	91%	93%	95%	69%	81%	90%	92%	93%	93%	62%	77%	84%	88%	92%
16	88	75%	83%	90%	92%	94%	68%	81%	89%	91%	92%	92%	61%	76%	83%	88%	91%
17	88	75%	83%	90%	92%	94%	68%	81%	89%	91%	92%	92%	61%	76%	83%	88%	91%
18	90	73%	81%	88%	90%	92%	67%	79%	87%	89%	90%	90%	60%	74%	81%	86%	89%

Given the decreasing temporal trend in S deposition across all ecoregions, we also analyzed 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) from a temporal perspective. 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 (e.g., PA, Figure 5-13). 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 $\mu\text{eq/L}$ and at least 85% are able to achieve an ANC at or above 20 $\mu\text{eq/L}$ (Figure 1; PA, Table 7-2). 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/h-yr to 7.3 kg S/h-yr and 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 $\mu\text{eq/L}$ in the 2010-12 period and achieving an ANC of 50 $\mu\text{eq/L}$ in the 2014-16 period. When the 7 western ecoregions are included in a summary based on ANC targets of 20 $\mu\text{eq/L}$ for the West and 50 $\mu\text{eq/L}$ for the East,⁶¹ 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 (Figure 1; PA, Table 7-2).

⁶¹ 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 the PA, section 5.1.2.2).

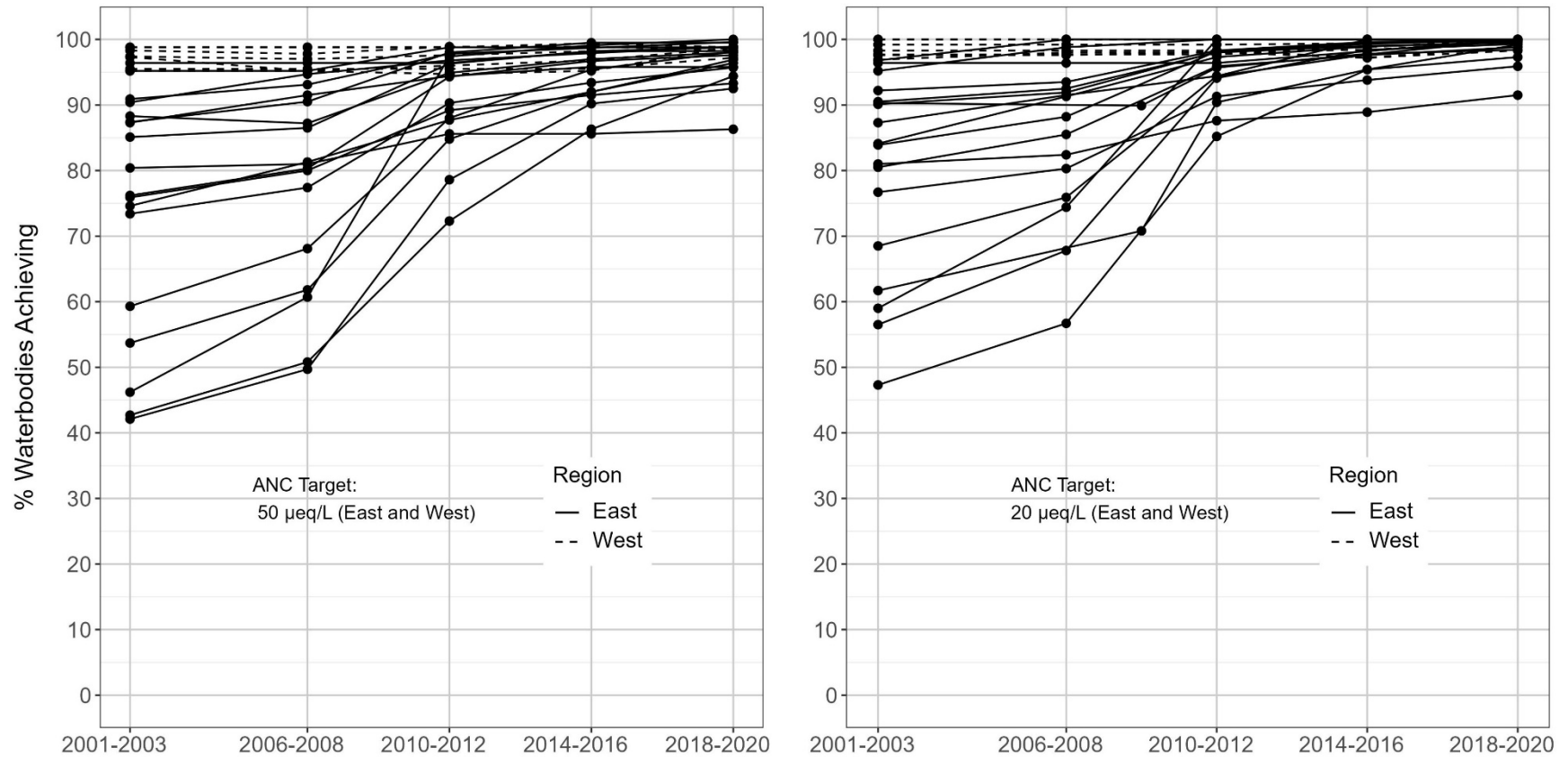


Figure 1. Percent of Waterbodies per Ecoregion Estimated to Achieve ANC at or Above 50 µeq/L (left panel) or 20 µeq/L (right panel). Western Ecoregions have Dashed Lines (Versus Solid Lines for Eastern Ecoregions). Each Line Represents One of the 25 Ecoregions.

E. Proposed Conclusions

In reaching his proposed decision on the current secondary standards for SO_x, N oxides and PM (presented in section II.E.3), the Administrator has taken into account policy-relevant, evidence-based and air quality-, exposure- and risk-based considerations discussed in the PA (summarized in section II.E.1), as well as advice from the CASAC, and public comment on the standard received thus far in the review (section II.E.2). In general, the role of the PA is to help “bridge the gap” between the Agency’s assessment of the current evidence and quantitative analyses of air quality, exposure, and risk, and the judgments required of the Administrator in determining whether it is appropriate to retain or revise the NAAQS. Evidence-based considerations draw upon the EPA’s integrated assessment of the scientific evidence presented in the ISA (summarized in section II.C above) to address key policy-relevant questions in the review. Similarly, the air quality-, exposure- and risk-based considerations draw upon our assessment of air quality, exposure, and associated risk (summarized in section II.D above).

This approach to reviewing the secondary standards is consistent with requirements of the provisions of the CAA related to the review of the NAAQS and with how the EPA and the courts have historically interpreted the CAA. As discussed in section I.A 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. Consistent with the Agency’s approach across all NAAQS reviews, the EPA’s approach to informing these judgments is based on a recognition that the available welfare effects evidence generally reflects a continuum that includes ambient air-related exposures for which scientists generally agree that effects are likely to occur through lower levels at which the

likelihood and magnitude of response become increasingly uncertain. The CAA does not require the Administrator to establish secondary standards at a zero-risk level, but rather at levels that reduces risk sufficiently so as to protect the public welfare from known or anticipated adverse effects.

The proposed decision on the secondary standards for SO_x, N oxides and PM described below is a public welfare policy judgment by the Administrator that draws upon the scientific evidence for welfare effects, quantitative analyses of air quality, exposure, and risks, as available, and judgments about how to consider the uncertainties and limitations that are inherent in the scientific evidence and quantitative analyses. The four basic elements of the NAAQS (i.e., indicator, averaging time, form, and level) have been considered collectively in evaluating the public welfare protection afforded by the current standards. The Administrator's final decision will additionally consider public comments received on this proposed decision.

1. Evidence- and Exposure/Risk-Based Considerations in the Policy Assessment

The PA presents an evaluation of the evidence and quantitative analyses of air quality, exposure and potential risk related to ecological effects of SO_x, N oxides and PM. These ecological effects include both direct effects of the three criteria pollutants on biota, and ecological effects of ecosystem deposition of N and S associated with these pollutants. Because the role of the PA is to provide the broadest array of options for consideration consistent with the scientific information, the PA presents multiple policy options for consideration by the Administrator in this review of the secondary NAAQS for SO_x, N oxides and PM. These options, which are only briefly summarized here, are discussed in detail in section 7.4 of the PA, including with regard to the varying strength of support provided for each by the current evidence and quantitative analyses. For SO_x, the PA options identified include adoption of an

annual average SO₂ standard, averaged over three years, with a level within the range extending below 15 ppb and down to 5 ppb. Based on consideration of the available air quality analyses indicating that such an annual standard could be expected to also provide appropriate control for short-term concentrations, the PA recognizes options that would either revise the existing 3-hour secondary standard to an annual standard or augment it with an annual standard.

For N oxides and PM_{2.5}, the PA recognizes options for retention of the existing standards, without revision, and also options for revision, although with recognition of appreciable associated uncertainty. For N oxides, the PA recognizes the options of retaining the existing secondary NO₂ standard (with its annual average concentration of 53 ppb) or revising the existing standard level to within a range below 53 ppb to as low as 40-35 ppb, in combination with consideration of a form that entails averaging the annual average across three consecutive years. With regard to PM, the PA recognizes options of either retaining the existing suite of secondary standards or revising the current annual secondary PM_{2.5} standard level to within a range below 15 µg/m³ to as low as 12 µg/m³.

The PA additionally considered the potential for establishment of a revised secondary standard or suite of standards with alternate indicator(s) that might target specific chemicals that deposit N and S (e.g., particulate NO₃⁻, SO₄²⁻, NH₄⁺), but recognized there to be a number of associated uncertainties and complications that include uncertainties in relationships between concentrations near sources and in areas of deposition. Based on the currently available data and analyses, the PA did not find there to be advantages or benefits to these alternate indicators over those for the established indicators, while also noting that establishing a standard based on one or more of these indicators would require the establishment of new or updated regulatory monitoring networks and measurement methods that would require additional time and resources

(PA, sections 7.2 and 7.4).

The PA additionally recognizes that, as is the case in NAAQS reviews in general, decisions by the Administrator on the adequacy of existing standards or the appropriateness of new or revised standards will depend 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 the standard also include the interpretation of, and decisions as to the weight to place on, different aspects of the quantitative REA and air quality-deposition information and analyses, and associated uncertainties. Thus, the Administrator's conclusions regarding the secondary standards for SO_x, N oxides and PM will depend in part on public welfare policy judgments, science policy judgments regarding aspects of the evidence and exposure/risk estimates, and judgments about the level of public welfare protection that is requisite under the Clean Air Act.

The subsections below summarize key considerations from the PA. These focus first on consideration of the evidence, as evaluated in the ISA (and supported by the prior ISA and AQCDs), including that newly available in this review, and the extent to which it alters the EPA's overall conclusions regarding ecological effects of SO_x, N oxides and PM, both regarding direct effects on biota and regarding ecological effects of ecosystem deposition of N and S compounds. The PA also considers the available information related to the general approach or framework in which to evaluate public welfare protection of the standard. Additionally, the PA considers the currently available quantitative information regarding environmental exposures likely to occur in areas of the U.S. where the standards are met. In so doing, the PA considers

associated limitations and uncertainties, and the significance of these exposures with regard to the potential for effects, their potential severity and any associated public welfare implications. The PA also considers judgments about the uncertainties inherent in the scientific evidence and quantitative analyses that are integral to consideration of whether the currently available information supports or calls into question the adequacy of the current secondary standards.

a. Direct Effects on Biota

In considering the currently available evidence and quantitative information pertaining to ecological effects of SO_x, N oxides and PM in ambient air, other than those associated with ecosystem deposition of S and N, the PA focused on several aspects. These include the extent to which the newly available information alters our scientific understanding of the ecological effects of SO_x, N oxides and PM in ambient air; the extent to which the currently available information indicates the potential for exposures associated with ecological effects under air quality meeting the existing standards and whether such effects might be of sufficient magnitude, severity, extent and/or frequency such that they might reasonably be judged to be adverse to public welfare; and to what extent important uncertainties identified in past reviews have been reduced and/or whether new uncertainties emerged. These considerations are summarized below, first for SO_x, followed by N oxides and then PM.

(1) Sulfur Oxides

As summarized in section II.C.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 gaseous SO_x — which include SO, SO₂, SO₃, and S₂O — only SO₂ 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 SO₂, documents the effects of SO₂ 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 that the evidence is sufficient to infer a causal relationship between gas-phase SO₂ and injury to vegetation (ISA, section 3.6.1).

In general, direct effects on plants occur at SO₂ exposures higher than a 3-hour average concentration of 0.5 ppm (500 ppb). The evidence derives from a combination of laboratory studies and observational studies. With regard to the sensitive effect of foliar injury, the current ISA finds “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 SO₂ can have acute negative effects on vegetation but does not change conclusions from the 2008 ISA regarding ... the SO₂ 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 SO₂ from those of other pollutants that may have influenced the analyzed effects. Regardless of these uncertainties, the evidence indicates effects are generally associated with air concentrations and durations not expected to occur when the existing standard is met (PA, section 7.1.1; ISA, Appendix 2, section 2.1)

(2) Nitrogen Oxides

The currently available information on direct effects of gaseous N oxides in ambient air is composed predominantly of studies of NO₂ and HNO₃, and also of PAN, with regard to effects on plants and lichens (as summarized in section II.C.1 above). 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, 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, NO₂, and PAN and injury to vegetation (ISA, section IS.4.2).

With regard to NO₂ ambient air exposure concentrations, the newly available information does not alter prior conclusions regarding exposure conditions associated with visible injury and effects on plant photosynthesis or growth. The 1993 AQCD for N oxides concluded that concentrations of NO, NO₂, and PAN in the atmosphere are rarely high enough to have phytotoxic effects on vegetation, and since that document, very little new research has been performed on these phytotoxic effects at concentrations currently observed in the U.S. (ISA, Appendix 3, sections 3.3 and 3.6.2; U.S. EPA, 1993). Further, 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).

Regarding another N oxide compound, HNO₃, in ambient air, the previously available evidence included experimental studies of leaf cuticle damage in tree seedlings, a finding confirmed in a more recent study, and also studies of effects on lichens. Effects of HNO₃ may be related to vapor exposures or direct contact via deposition (PA, section 7.1.2; ISA, Appendix 3, section 3.4). The evidence also includes studies of effects related to historic conditions in the Los Angeles Basin that indicate N oxides, and particularly HNO₃, to be “the main agent of decline of lichen in the Los Angeles [B]asin” (ISA, Appendix 3, p. 3-15). A reassessment in 2008 found that lichen communities have not recovered from the damage evident in the 1970s, although the extent to which this reflects residual impacts of earlier effects is unknown (PA, section 7.1.2;

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 HNO₃ and changes to vegetation” (ISA, section 4.3).

The recently available information for HNO₃ includes effects on tree foliage under controlled 12-hour exposures to 50 ppb HNO₃ (approximately 75 µg/m³) and in longer, 32- or 33-day exposures in which peak HNO₃ concentrations for the “moderate” treatment (30-60 µg/m³) encompassed the range reported in summers during the 1980s in the Los Angeles Basin (ISA, Appendix 3, section 3.4). During that period, NO₂ concentrations in the Basin ranged up to 0.058 ppm, exceeding the secondary standard (PA, section 5.4.2; U.S. EPA, 1987). Effects on lichen photosynthesis have been reported from daily 6.5-hour varying exposures with peaks near 50 ppb (~75 µg/m³) lasting longer than 18 days (ISA, Appendix 6, section 6.2.3.3; Riddell et al., 2012).

In considering the potential for concentrations of N oxides, including HNO₃, that are associated with ecological effects to occur under air quality conditions meeting the current NO₂ standard, the PA noted that air quality at all ambient air monitoring sites in the contiguous U.S. has met the existing secondary NO₂ standard since around 1991 (PA, Figure 2-22). In considering the potential for HNO₃ concentrations of a magnitude sufficient to pose risk of effects to occur under conditions that meet the current NO₂ secondary standard, the PA also considered the magnitude of NO₂ concentrations in the Los Angeles Basin. During the 1970s to 1990s, the Los Angeles metropolitan area experienced NO₂ concentrations in excess of the NO₂ secondary standard (e.g., annual average concentrations up to 0.078 ppm in 1979 and above 0.053 ppm into the early 1990s). At the time of the 2008 reassessment mentioned above, which

reported that impacts documented on lichen communities in the 1970s still remained, NO₂ concentrations were well below the standard (PA, section 7.1.2; ISA, Appendix 3, section 3.4), although the extent to which this finding relates to a lag in recovery or concurrent air pollutant concentrations is unknown. The PA notes that the risk of HNO₃ effects to lichens may be from both direct and deposition-related exposure related to direct contact of the chemical to the lichen surfaces (PA, section 7.1.2).

In summary, the currently available information is somewhat limited with regard to the extent to which it informs conclusions on the potential for ambient air exposures associated with ecological effects under air quality meeting the existing NO₂ secondary standard. More recent studies also indicate variation in eutrophic lichen abundance to be associated with variation in metrics representing N deposition, although the extent to which these associations are influenced by residual impacts of the historic air quality is unclear (ISA, Appendix 6, section 6.2.3.3; PA, sections 5.3.3.2 and 7.1.2). While new uncertainties have not emerged, uncertainties remain in our interpretation of the evidence, including those related to limitations and associated uncertainties of the various study types. 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 may have contributed to the observations. With regard to the risk posed by N oxides, and particularly HNO₃, the evidence summarized in the ISA indicates the potential for effects on lichen species related to air quality occurring during periods when the current secondary standard was not met. The evidence is limited, however, in support of conclusions of effects under conditions meeting the current standard (PA, section 7.1.2).

(3) Particulate Matter

As summarized in section II.C above, the evidence for ecological effects of PM is

consistent with that available in the last review. The causal determinations with regard to ecological effects of PM in the 2013 PM review (2009 PM ISA) and in this review (2020 ISA) focused on effects associated with PM loading (e.g., to leaf surfaces), rather than direct effects of PM suspended 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; 2012 PM ISA, section 9.4).

With regard to direct effects of PM in ambient air, the available information indicates effects occurring only at ambient air concentrations 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 effects of PM under generally lower PM concentrations in ambient air likely to occur under conditions meeting the current secondary standards, and the limited available information does not indicate effects to occur under those conditions (PA, section 7.1.3).

b. Evidence of Ecosystem Effects of S and N Deposition

The evidence base of ecological effects related to atmospheric deposition of N and S compounds 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, as summarized in section II.C.1 above. A long-standing evidence base documents the array of effects of both acidic deposition in aquatic and terrestrial ecosystems and ecosystem N enrichment. The evidence for acid 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 noted in section II.C and II.D above, 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 in 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, and there is 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 of the PA (ISA, Appendix 7, section 7.2.9; 2008 ISA, section 3.3.2.4; Officer et al., 1984). Additionally, the evidence base on the effects of N enrichment in terrestrial ecosystems, primarily in grassland and forested ecosystems, includes evidence that was available in the last review (e.g., 2008 ISA, sections 3.3.3 and 3.3.5).

Some uncertainties associated with the evidence available in the 2012 review remain, and some additional important uncertainties have been identified. In addition to uncertainties related to the specific air quality circumstances associated with effects (e.g., magnitude, duration, and frequency of concentrations associated with effects), there are also uncertainties associated with the effects of N and S deposition expected under changing environmental circumstances. Such uncertainties include atmospheric loading that has declined since 2000, with associated changes to soil and waterbody biogeochemistry and meteorological changes associated with changing climate (ISA, section IS.12; PA section, 7.2.1). The PA also recognizes important uncertainties associated with the various assessment approaches employed by different study types (PA, sections 5.3 and 7.2.1). 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).

Additionally, 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 experiments⁶² include, among others, those related to the potential for effects to occur over longer periods than those assessed in those studies (PA, section 5.3.4.1).

Lastly, studies reporting atmospheric deposition rates associated with effects involve authors' judgments regarding the magnitude of responses considered to be effects, and may also be limited by 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. 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).⁶³ Variability in physical, chemical, and ecological characteristics of ecosystems also contribute uncertainty to such judgments (PA, section 7.2.1).

In sum, a wealth of scientific evidence, spanning many decades, demonstrates effects of

⁶² Addition experiments generally refers to field experiments where compounds (e.g., NO₃⁻ or SO₄²⁻ in acidification experiments) are added (generally annually) to the soil of large forested (for tree studies) areas and the tree measurements (e.g., growth rate) are compared to those in an untreated or control area.

⁶³ In describing critical loads developed from observational studies (or empirical data), the ISA recognizes distinctions from other studies, as seen in the following excerpt (ISA, p. IS-113).

The 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. In contrast, CLs for acidification are typically based on the deposition amount that gives rise to a soil chemical indicator value which is known to cause an adverse biological effect. The link between soil chemical indicator and biological effect is based on empirical evidence (Appendix 5). The relationship between deposition and the biogeochemistry that causes effects on soil chemistry is typically modeled (Appendix 4; section IS.14.2).

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, 1982b, Chapter 7). This evidence base supports conclusions also reached in the 2008 ISA (for the review completed in 2012) 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 effects of N enrichment in both estuarine and freshwater ecosystems, supporting conclusions also 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). 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 (ISA, Table ES-1).

In terrestrial ecosystems, as in the last review, the now expanded evidence base supports determination of a causal relationship between N deposition and alteration of species richness, community composition, and biodiversity (ISA, Table ES-1). 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). Other evidence of effects causally associated with S deposition in wetland and freshwater ecosystems includes that related to chemical transformation and associated toxicity, most specifically alteration of mercury methylation, which was also recognized in the last

review. The other category of effects, not included in the last review, is related to sulfide phytotoxicity and its associated effects in wetland and freshwater ecosystems (ISA, Table ES-1).

Thus, while an array of effects is associated with S and N deposition, information important for quantitative analysis varies across the array. For some categories of effects (e.g., sulfide phytotoxicity) the information is limited and/or quite variable across locations with regard to environmental levels relating to effects, thus hindering analysis. For others, information is limited and/or quite variable, with regard to its linkages to the criteria pollutants. The information with clearest implications to NAAQS decisions pertains to SO_x and S deposition-related ecosystem acidification. While the information regarding effects associated with N loading to ecosystems is extensive, information to support quantitative analysis to inform NAAQS decisions regarding N oxides and PM is not clear, with multiple complicating factors. Such factors include contributions from other, non-criteria pollutants, such as NH₃. As noted in section II.D above, the role of N deposition in aquatic acidification is considered in the REA. With regard to other N deposition-related effects of N oxides and PM, the information does not provide effective support for such analysis, independent of effects from other (non-criteria) pollutants, or, in some cases, from other (non-air) sources.

c. Sulfur Deposition and SO_x

Evidence- and exposure/risk-based considerations discussed in the PA pertaining to S deposition and SO_x in ambient air are summarized in the subsections below. These considerations reflect discussion in the PA, which draws on the available welfare effects evidence described in the current ISA, the 2008 NO_x/SO_x ISA, the 2009 PM ISA, and past AQCDs, as well as information available from quantitative analyses (summarized in Chapters 5 and 6 of the PA), both analyses recently developed and those available from the 2009 REA.

In considering potential public welfare protection from S deposition-related effects in aquatic ecosystems (in light of the aquatic acidification REA results summarized in II.D above), the PA notes as an initial matter, the integral role of watersheds in aquatic ecosystem health (e.g., ISA, Appendix 8, section 8.1 and Appendix 16, section 16.4.2) and the effects of acidic deposition on forested areas in the watersheds that are distinct from effects in water bodies (e.g., reduced tree growth and survival). Further, as discussed in section II.C.2 above, there are an array of benefits of watershed forested areas to the public, including such ecosystem services as silviculture, drinking water supply protection, recreational uses. In light of these public benefits, the PA recognizes the public welfare implications of various effects of acidifying deposition on the natural resources in these areas, with the public welfare significance dependent on the severity and extent of such effects. Given the more extensive quantitative analyses for aquatic acidification in this review, the PA discusses the public welfare implications of S deposition-related effects in aquatic ecosystems with an eye toward their prominence for decision-making in this review (PA, sections 4.5 and 7.2.2.2). In so doing, the PA judges that in focusing on public welfare protection from aquatic acidification-related effects will provide protection from watershed soils, and accordingly, for associated watershed resources.

The PA notes that, as also 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). 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 (PA, section 4.5; ISA, Appendix 14, section 14.3.1). 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 (PA, section 4.5).

The PA recognizes 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 that aquatic acidification resulting in reduced acid buffering capacity can adversely affect waterbodies and associated fisheries, which in addition to any commercial ramifications can have ramifications on recreational enjoyment of affected areas (PA, sections 5.1.1 and 4.5). 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, 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., including in several Class I areas and other national and state parks and forests (PA, section 5.1.2.1).⁶⁴ Two waterbodies in such areas were

⁶⁴ A comparison of Figures 4-4 and 5-6 of the PA indicates multiple Class I areas in ecoregions considered acid sensitive.

included as case studies in the aquatic acidification REA: Shenandoah Valley Area and Rocky Mountain National Park (PA, section 5.1.3.3). 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, it is particularly important in Class I and similarly protected areas. In this review, in considering the potential public welfare significance of aquatic acidification effects of differing levels of S deposition, the PA summarizes the REA ecoregion-scale results in terms of percentages of ecoregions in which differing percentages of waterbodies are estimated to achieve the three acid buffering capacity targets. The PA summarized results in this way to inform identification of S deposition estimates in the context of potential policy options.

The first subsection below (II.E.1.c(1)) focuses on the aquatic acidification REA analyses (summarized in section II.D above), considering first their use of ANC as the indicator of acidification risk, then evaluating the exposure/risk estimates as to what they indicate about acidification risks in freshwater streams and lakes of the contiguous U.S. for S deposition rates estimated to have occurred over the past two decades (much of which is newly assessed in this review),⁶⁵ and lastly identifying important uncertainties associated with the estimates. Section II.E.1.c(2) considers the evidence and quantitative exposure/risk information from a public welfare protection perspective, focusing first on what might be indicated regarding deposition conditions under which waterbodies in acid-sensitive ecoregions might be expected to achieve acid buffering capacity of interest and what the available information indicates pertaining to the consideration of public welfare protection from S deposition related effects in aquatic

⁶⁵ Aquatic acidification risk analyses in the last review considered deposition estimates for 2002 and 2006 derived from CMAQ modeling, 2002 emissions estimates (2009 REA, Appendix 1).

ecosystems. Lastly, section II.E.1.c(2) considers what the published quantitative information regarding S deposition and terrestrial acidification indicates regarding deposition levels of potential concern, along with associated uncertainties in this information. Section II.E.1.c(3) then summarizes considerations in relating SO_x air quality metrics to deposition of S compounds.

(1) Quantitative Information for Ecosystem Risks Associated with S Deposition

As in the last review, we give primary attention to the quantitative assessment of aquatic acidification (including particularly that attributable to S deposition). While noting the uncertainties associated with results of the aquatic acidification REA, as summarized in section II.D.2 above, the PA recognized these results to be informative to the identification of S deposition levels associated with potential for aquatic acidification effects of concern, as summarized below. This assessment of quantitative linkages between S deposition and potential for aquatic acidification is one component of the approach implemented in the PA for informing judgments on the likelihood of occurrence of such effects under differing air quality conditions.

Although the approaches and tools for assessing aquatic acidification have often been applied for S and N deposition in combination, the approach taken in the REA for this review is focused on S deposition. This focus is supported by analyses in the PA indicating the relatively greater contribution of S deposition than N deposition to aquatic acidification risk under the more recent air quality conditions that are the focus of this review (PA, Appendix 5A). As summarized in section II.D above, the aquatic acidification assessment has relied on 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, and the findings from these analyses presented in

the 2009 REA have been considered in this review in the context of more recently available evidence (PA, section 5.3.2.1; 2009 REA, section 4.3).⁶⁶ In the last review, analyses that related estimated atmospheric deposition of acidic N and S compounds (for early 2000s time period) to terrestrial effects, or indicators of terrestrial ecosystem risk, were generally considered to be more uncertain than conceptually similar modeling analyses for aquatic ecosystems. For example, the 2009 REA concluded that “aquatic acidification is clearly the targeted effect area with the highest level of confidence” (2009 REA, section 7.5; 2011 PA, section 1.3). Additionally, the PA for this review notes that 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 in aquatic systems (PA, sections 4.2.3.1 and 4.2.3.2).

As described in sections II.C and II.D above, ANC is an indicator of susceptibility or risk of acidification-related effects in waterbodies, with lower levels indicating relatively higher potential for acidification and related waterbody effects. The support for this relationship is strongest in aquatic systems low in organic material, and the evidence comes predominantly from historically affected waterbodies in the eastern U.S. (e.g., in the Adirondack Mountains) and Canada. In waterbodies with relatively higher levels of dissolved organic material, the presence of organic acid anions contributes to reduced pH, but also creates complexes with dissolved aluminum that protect resident biota against aluminum toxicity (ISA, Appendix 8, section 8.3.6.2; PA, section 7.2.2.1). Accordingly, biota in such systems tolerate lower ANC

⁶⁶ Given findings from the 2009 REA that aquatic acidification provided a more sensitive measure for use in assessing deposition related to ecosystem acidification, and consideration of recent information not likely to result in a different finding, the REA for the current review focused on aquatic acidification.

values (and pH) than biota in waterbodies with low dissolved organic carbon. Thus, while the evidence generally supports the use of ANC as an acidification indicator and for purposes of judging a potential for ecosystem acidification effects generally, the relationship with risk differs depending on the presence of naturally occurring organic acids, which also affects the responsiveness of ANC to acidifying deposition in these areas. For these reasons, in some areas, ANC is less well supported as an indicator for acidic deposition-related effects (and waterbodies are less responsive to changes in acidic deposition) due to dissolved organic material; these areas include the Middle Atlantic Coastal Plain, Southern Coastal Plains, and Atlantic Coastal Pine Barrens ecoregions (PA, section 5.1.2.2).

The PA considers the available evidence to provide strong support for use 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 $\mu\text{eq/L}$ (PA, section 5.1). 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 $\mu\text{eq/L}$ (PA, section 5.1.2.2). The severity of impacts is greatest at the lowest ANC levels. This evidence derives primarily from lakes and streams of the Adirondack Mountains and areas along the Appalachian Mountains. As recognized in the 2012 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, some waterbodies with ANC below 20 $\mu\text{eq/L}$ have been associated with increased probability of low pH events, that, depending on other factors as noted above, may 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 summarizing and considering the acidification risk estimates for the different scales of analysis (national, ecoregion and case study) and using the water quality modeling-based CLs derived for three different ANC targets (20, 30 and 50 $\mu\text{eq/L}$), the PA recognizes 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 in patterns of S deposition. Further, the PA recognizes limitations and uncertainties in the use of ANC as an indicator for model-based risk assessments as noted above (PA, section 7.2.2.1).

The REA national-scale analysis of more than 13,000 waterbody sites in 69 ecoregions demonstrated an appreciable reduction in risk over the 20-year period of analysis (PA, section 5.1.3). For the 2001-2003 period, more than 20% of waterbodies analyzed nationally were estimated to be unable to achieve an ANC of 20 $\mu\text{eq/L}$ or greater based on S deposition estimates (Table 1 above). This percentage declines significantly by the 2010-2012 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 $\mu\text{eq/L}$ and 50 $\mu\text{eq/L}$, respectively (Table 1). The 25 ecoregions included in the ecoregion-scale analyses (i.e., 18 in the East and 7 in the West in which there are at least 50 waterbody sites with CL estimates) are dominated by ecoregions categorized as acid sensitive (PA, Table 5A-5) and excludes the three ecoregions identified above as having natural acidity related to organic acids⁶⁷ (PA, 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

⁶⁷ The natural acidity contributes to a reduced responsiveness to changes in acidic deposition.

the dominance of the acid-sensitive ecoregions among the 25 analyzed in the ecoregion-scale analysis. Specifically, in the most affected ecoregion (Central Appalachians), more than 50% of waterbodies were estimated to be unable to achieve an ANC of 20 $\mu\text{eq/L}$ or greater based on S deposition estimates for the 2001-2003 period; the percentage was close to 60% for an ANC target of 50 $\mu\text{eq/L}$ (Figure 1 above, and PA, Figure 5-13). By the 2018-2020 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 $\mu\text{eq/L}$ and less than 15% of waterbodies in the most affected ecoregion were estimated to be unable to achieve an ANC of 50 $\mu\text{eq/L}$ (Figure 1 above and PA, Figure 5-13).

The PA recognizes uncertainty associated with two overarching aspects of the aquatic acidification REA (PA, section 5.1.4 and Appendix 5A, section 5A.3). The first relates to interpretation of specific thresholds or benchmark concentrations of ANC with regard to aquatic acidification risk. 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 interpretation of ANC thresholds, 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. These other site-specific 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 (PA, sections 5.1.4 and 7.2.2.1 and Appendix 5A, section 5A.3). There are also uncertainties associated with the estimates of S deposition used in the analyses of CL

exceedances, including those for the national- and ecoregion-scale analyses (PA, section 6.3.1, Table 6-13). Consideration of such uncertainties informs the weighing of the findings of the quantitative analyses. For example, there is more uncertainty associated with CLs in areas that are less well studied. Thus, the PA suggests that it is appropriate to put greater emphasis on the more well studied areas and/or less emphasis on estimates for the tails of the distributions (e.g., upper/lower percentiles) of waterbody exceedances within an ecoregion or case study area. This information additionally informs interpretation of the potential risk associated with estimates for the different ANC targets.

With regard to estimation of CLs for the different ANC targets, associated uncertainties, generally related to parameters used in the steady-state CL models, are difficult to characterize and assess. Such uncertainties contribute uncertainty to estimation of the ANC levels that individual waterbodies might be expected to achieve under different rates of S deposition. While the water quality models used for estimating aquatic acidification CL 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 (PA, Appendix 5A, section 5A.3). For example, as recognized in section II.D.2 above, the data to support the site-specific model inputs for some areas are more limited than others, with associated greater uncertainties (PA, sections 4.2.1.3 and 5.1.4).

Most particularly, 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 measurement availability, which varies among waterbodies. The model input associated with estimating base

cation catchment supply is the base cation weathering rate, which the ISA recognizes 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; Li and McNulty, 2007). Although the approach to estimate base-cation supply in the REA (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 magnitude of uncertainty in this estimate is unclear and could be large in some cases. The REA’s quantitative analysis of uncertainty in CL estimates indicates lower uncertainty associated with CLs estimated for sites with more extensive and longer-term water quality datasets and relatively low variability in the runoff measurements, such as CLs for waterbody sites in the eastern U.S., particularly along the Appalachian Mountains, in the Upper Midwest, and in the Rocky Mountains. The analysis found greater uncertainty associated with CLs estimated for sites in the Midwest and South and along the CA to WA coast (PA, Appendix 5A, section 5A.3.1).

(2) General Approach for Considering Public Welfare Protection

In discussing key considerations in judging public welfare protection from S deposition in the context of the review of the secondary standard for SO_x, the PA first focused on the results of the aquatic acidification REA as to what they indicated about deposition conditions under which waterbodies in sensitive ecoregions might be expected to achieve ANC levels of interest. In so doing, the PA focused on the results of the aquatic acidification REA at three scales: national-scale, ecoregion-scale and the more localized case study-scale, giving particular focus to the ecoregion and case-study analyses, which use 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. The PA also

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 recognition of the variation in ANC response reasonably expected across waterbodies in an ecoregion based on both differences in watersheds that can affect sensitivity to S deposition and with different spatial or geographic patterns of S deposition.

At the national scale, as summarized in section II.E.1.c(1) above, unlike the case for the 2000-2002 period, few waterbodies are estimated to be receiving deposition in excess of their CLs for the three ANC targets under recent deposition estimates. For example, 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 $\mu\text{eq/L}$ and 1% for an ANC of 20 $\mu\text{eq/L}$ (Table 1 above). In this time period (2018-2020), median estimates of deposition in all of the 69 ecoregions that are represented in these national-scale percentages (ecoregions with at least one site with a CL estimate) are at or below approximately 4 kg S/ha-yr (PA, Tables 5A-15 and 5A-11).

Based on the array of CL-based analyses, the PA 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 (4,977 sites distributed across three ecoregions), 90% of waterbody sites are estimated to be able to achieve an ANC at or above 20 $\mu\text{eq/L}$ (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 (PA, section 5.1.3.3). For an ANC target at or above 50 $\mu\text{eq/L}$ in the Shenandoah Valley case study, the corresponding deposition estimates are 4.1 and 6.3 kg/ha-yr (PA, Table 5-6). For the other case study areas (White Mountain National

Forest, Northern Minnesota, Sierra Nevada Mountains and Rocky Mountain National Park), there are appreciably fewer waterbody sites for which modeling has been performed to estimate CLs, and accordingly greater uncertainty. Yet, the case study area averages of waterbody CLs for achieving ANC at or above each of the three targets (20, 30 or 50 $\mu\text{eq/L}$) is quite similar across the five case studies, ranging from 9.4 kg/ha-yr for an ANC of 50 $\mu\text{eq/L}$ in Shenandoah Valley Area to 12 kg/ha-yr for an ANC of 20 $\mu\text{eq/L}$ in both Shenandoah and Sierra Nevada Mountains case study areas (PA, Table 5-6).

Findings from the ecoregion-scale analyses of 25 ecoregions (18 East and 7 West), nearly all of which are considered acid sensitive, indicated ranges of deposition estimates associated with high percentages of waterbodies estimated to achieve the three ANC targets that are similar to the case study results immediately above. This was the case when considering the ecoregion-scale analysis results in both of the two ways they were presented: (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.

For example, in the ecoregion-time period combinations presentation, 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 $\mu\text{eq/L}$ 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 (Table 4 below). This summary contrasts with, and indicates appreciably greater acid buffering capacity than, the estimates for S deposition at or below 18 kg/ha-yr (Table 4 below). Further, 70% of waterbody sites in all 18 eastern ecoregions are estimated to achieve an ANC at or above 50 $\mu\text{eq/L}$ with ecoregion median S deposition at or below 9 kg/ha-yr. Although fewer ecoregion-

time period combinations are associated with still lower S deposition estimates, contributing to increased uncertainty, we also note that for the lowest bin that is composed of at least half of the 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 $\mu\text{eq/L}$ in 96% of the combinations and at or above 50 $\mu\text{eq/L}$ in 82% of the combinations (Table 4 below).

In total, the ecoregion-time periods presentation indicates the likelihood of appreciably more waterbodies achieving the acid buffering capacity targets among the combinations with ecoregion median deposition at or below 9 kg/ha-yr (and for the bins for lower values) in eastern ecoregions compared to the estimates of waterbodies achieving acid buffering targets based on the full dataset that includes deposition estimates up to 18 kg/ha-yr. More specifically, this reflects an appreciably greater percentage of waterbodies in more ecoregions achieving ANC at or above 20 $\mu\text{eq/L}$, at or above 30 $\mu\text{eq/L}$, and at or above 50 $\mu\text{eq/L}$ (Table 4 below), with ecoregion median deposition levels at or below 9 kg/ha-yr. Additionally, these percentages increase across the bins for the lower deposition estimates, although they are also based on smaller proportions of the supporting dataset (i.e., fewer ecoregion-time period combinations in each subsequently lower deposition bin).

Table 4. 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								
		$\geq 90\%$ of waterbodies			$\geq 80\%$ of waterbodies			$\geq 70\%$ of waterbodies		
ANC ($\mu\text{eq/L}$) 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 from the more extensive presentation of results in PA, Table 5-5.										

The PA observes that estimates from the temporal trend perspective similarly indicate appreciable percentages of waterbodies per ecoregion being estimated to achieve the acid buffering capacity targets with ecoregion median deposition below a range of approximately 8 to 5 kg/ha-yr. For example, during the latter half of the 20-year period analyzed (i.e., by the 2010-2012 period), by which time all 25 ecoregions are estimated to have more than 70% of waterbodies able to achieve an ANC at or above 50 µeq/L (and at least 85% able to achieve an ANC at or above 20 µeq/L), median deposition in 95% of the ecoregions was below 8 kg S/ha-yr, ranging from 1.3 to 7.3 kg S/ha-yr (PA, Table 7-2 and Figure 7-1). As shown in Table 5 below, 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,⁶⁸ 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 (Table 5). By the 2014-2016 and 2018-2020 periods, 24 of the 25 ecoregions were

⁶⁸ 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 of the PA.

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 (Table 5).

Table 5. 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.

The temporal trends in percentage of waterbodies estimated to achieve the target ANC

levels for each of the 25 individual ecoregions document a large difference between the time periods prior to 2010 and subsequent time periods (Figure 1 above; PA, 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 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 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 for the 25 REA ecoregions (Figure 2; PA, Figure 7-2). The distribution of deposition estimates at waterbody sites assessed in each ecoregion, and particularly the pattern for the higher percentile sites in each ecoregion, illustrates 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 90th percentile deposition estimates ranged from approximately 14 to 17 kg/ha-yr, with maximum values above 20 kg/ha-yr (Figure 2). This contrasts with the deposition estimates during the 2010-2020 period when, among all 25 ecoregions, the medians of the ecoregion 90th percentile deposition estimates ranged from approximately 2 to 5 kg/ha-yr, with all ecoregion 90th percentile estimates below 8 kg/ha-yr (Figure 2). The contrast is less sharp for the ecoregion medians, as the median is a statistic less influenced by changes in the magnitude of values at the upper end of the distribution (Figure 2).

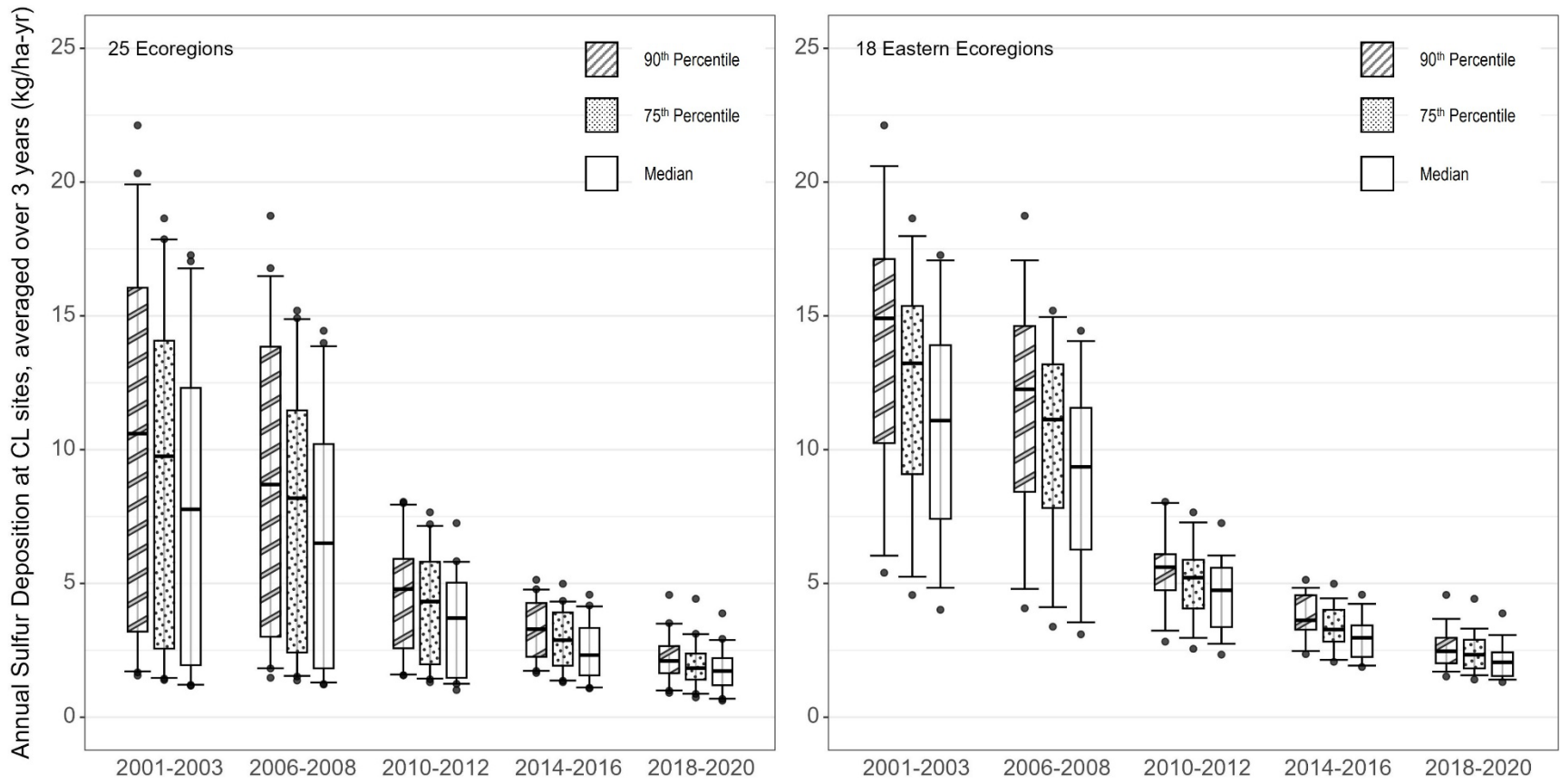


Figure 2. Ecoregion 90th, 75th and 50th Percentile S Deposition Estimates at REA Waterbody Sites Summarized for all 25 Ecoregions (left) and the 18 Eastern Ecoregions (right).

Thus, in considering identification of S deposition levels that may be associated with a desired level of ecosystem protection for an SO_x standard, the PA took note of the increased percentages of waterbodies estimated to achieve more protective ANC levels across the five time periods. The pattern of estimated improving water quality over the 20-year study period is paralleled by the pattern of declining deposition, which is more obvious in the upper percentiles (than the median) of the distribution of values per ecoregion (Figure 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 (2010-2020) 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 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 achieving ANC targets for ecoregion median S deposition at or below about 8 or 9 kg/ha-yr (Table 4). Lastly, the case study CL estimates also indicate appreciable portions of the case study areas that might be expected to attain the three ANC targets with deposition below 9 kg/ha-yr. Thus, in light of these observations, the PA describes S deposition, on an areawide basis, that falls below approximately 10-5 kg/ha-yr, or 8-5 kg/ha-yr (differing slightly depending on the supporting analysis), as being associated with the potential to achieve acid buffering capacity levels of interest in an appreciable portion of sensitive areas.

In considering what the quantitative information regarding S deposition and terrestrial

acidification indicates regarding deposition levels of relatively greater and lesser concern for potential acidification-related effects (and the associated uncertainties), the PA considers 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 (PA, section 5.3). With regard to soil chemistry modeling analyses performed in the last review, the PA notes that the resulting estimates of acidic deposition CLs for three values of the soil acidification indicator, BC:Al ratio, indicated a range generally well above the CL estimates associated with achieving various ANC targets in the aquatic acidification analyses discussed above. The soil acidification CLs were also above all of the ecoregion estimates (across the five time periods from 2001 through 2020) considered in the aquatic acidification analyses (PA, Table 5-7). Thus, the PA concluded that these soil acidification modeling findings indicate that a focus on aquatic acidification might reasonably be expected to also provide protection from soil acidification effects on terrestrial biota. With regard to studies involving S additions to experimental forested areas, the PA notes that although the number of tree species that have been included in such experiments is somewhat limited, the more widely recognized sensitive species (based on field observations) have been included in such studies. Across these studies, the PA observes that effects on the trees analyzed have not been reported with S additions below 20 kg/ha-yr (which is in addition to the atmospheric deposition occurring during the experiment).

The PA also considers the recently available quantitative information regarding S deposition and terrestrial acidification drawn from 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 in regions of the eastern U.S. (PA, section 5.3.2.3 and Appendix 5B, section 5B.3.2). The metrics used 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 west of the Mississippi River, northern New England (e.g., Maine) and southern Georgia and Florida (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 (PA, 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 S/ha-yr (Appendix 5B, section 5B.3.2.3; Horn et al., 2018).

In considering these study observations, the PA notes the history of appreciable acidic deposition in the eastern U.S., with its associated impacts on soil chemistry, that has the potential to be exerting a legacy influence on tree growth and survival more recently (PA, section 5.3.2 and Appendix 5B). Further, the PA notes that 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., PA, sections 2.5.4 and 6.2.1). This similarity in patterns is recognized to have 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, and to contribute uncertainty with regard to interpretation of these studies as to a specific magnitude of deposition that might be expected to elicit specific tree responses, such as those for which associations have been found. The PA notes that, 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 the knowledge that acidification impacts on tree mortality result from cumulative long-term deposition (PA, section 5.3.1 and Appendix 5B).

(3) Relating Air Quality Metrics to S Deposition

In considering what the available information and air quality analyses indicate regarding relationships between air quality metrics and S deposition, the PA evaluated trends over the past two decades as well as a series of analyses of relationships between S deposition and ambient air concentrations of SO₂ (in terms of 3-year averages of the existing SO₂ standard and as an annual average), and between S deposition and ambient air concentrations of other S compounds (e.g., SO₄²⁻ or summed SO₄²⁻ and SO₂) at 27 Class I area sites, as summarized in section II.B above.⁶⁹

⁶⁹ The air quality metrics include one based on the current secondary SO₂ NAAQS, which is the second highest 3-hour daily maximum in a year, as well as an annual average SO₂ air quality metric (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 air quality metrics analyzed, the PA focused primarily on the annual average of SO₂ concentrations, averaged over 3 years, given the focus on control of long-term S deposition and the greater stability of the metric (PA, section 7.2.2.3).

With regard to indicators other than SO₂, lower correlations were observed for collocated total S deposition estimates with indicators of atmospheric S-containing pollutants (particulate SO₄²⁻ and the sum of S in SO₂ and particulate SO₄²⁻) in 27 Class I areas than between S deposition and annual average SO₂ concentrations (averaged over three years) at SLAMS monitors (PA, Figures 6-27 and 6-31 and Table 6-4). Thus, while the data at the Class I area sites (collocated CASTNET and IMPROVE network sites) provide information for S compounds other than SO₂, the analyses based on data from SLAMS are considered particularly relevant given that those sites are primarily in areas of higher SO₂ concentrations near emissions sources and collect FRM/FEM measurements for evaluating ambient air concentrations relative to the existing NAAQS. Information from these monitoring sites is useful in understanding how changes in SO₂ emissions, reflected in ambient air concentrations, may relate to changes in deposition and, correspondingly, what secondary standard options might best relate to ambient air concentrations such that deposition in areas of interest is maintained at or below range of levels identified above (PA, section 7.2.2.3).

Together the air quality and deposition data and analyses in the PA indicate a significant association of S deposition with SO₂ concentrations, with statistically significant correlation coefficients ranging from approximately 0.50 up to 0.70 from the trajectory-based and SLAMS analyses for the five 3-year time periods (during 2001-2020) across all ecoregions. Higher correlations were observed for dry S deposition and at sites in the eastern U.S. (PA, section 7.2.2.3). As summarized in section II.B above, S deposition is generally higher in the east and dry S deposition is generally higher near SO₂ emissions sources. In considering the two types of analyses, relating concentrations to deposition either nearby or in downwind areas, the PA notes that a strength of the analyses for concentrations and deposition estimates at SLAMS locations is

the capturing of near-source deposition, while a strength of the trajectory-based analyses is accounting for the role of transport and transformation in contributing to downwind deposition.

While recognizing the significant correlations between SO₂ concentrations and S deposition, the PA additionally took note of the variability in, and uncertainty associated with relationships between SO₂ concentrations at SLAMS monitors and nearby and/or downwind S deposition. The variability relates to the complexity of the atmospheric chemistry, pollutant transport, and deposition processes (PA, sections 2.1.1 and 2.5). The uncertainty in these relationships relates to a number of factors, including uncertainty in our estimates of S deposition (PA, section 2.5.2) and spatial distribution of monitor sites, including the representation of significant SO₂ emissions sources, as well as elements of the trajectory-based analysis, e.g., inclusion criteria for identifying monitoring sites of influence (PA, section 6.3 and Table 6-13). The PA concluded that it is unclear how much and in what way each of these various uncertainties in the data and analyses, and the inherent variability of the physical and chemical processes involved, might impact the conclusions concerning ambient air SO₂ concentrations related to S deposition estimates at different scales (PA, section 7.2.2.3). In light of such uncertainty and variability, the REA aquatic acidification analyses and discussion of S deposition levels focused on statistics for deposition estimates representing large areas (e.g., at the ecoregion median and 75th or 90th percentile, and case study area average or 70th and 90th percentile CLs). While uncertainty may be greater for relating concentrations to higher points on the distribution of deposition in an ecoregion, the PA recognized that it is the higher deposition estimates, if focused on individual waterbodies, that will contribute most to aquatic acidification risk, and additionally observed that the distribution of S deposition estimates within ecoregions has collapsed in the more recent years of the 20-year analysis period, with 90th percentile

estimates falling much close to the medians than in the first decade of the period (Figure 2 above; PA, Figure 7-2).

In light of the declining trend in S deposition and the corresponding REA estimates of increasing ANC in sensitive ecoregions (as discussed above), the PA considered the annual average SO₂ concentration at SLAMS across five time periods from 2000-2020. In so doing, the PA focused on the most recent time periods analyzed (i.e., since 2010) when the REA indicated appreciably improved levels of acid buffering capability in the waterbodies of the 25 analyzed ecoregions (when ANC targets were met or exceeded in a high percentage of water bodies across a high percentage of ecoregions). This information 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 SO₂ 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 (PA, Figure 7-5, left panel). The distributions of annual average SO₂ 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 (PA, Figure 7-5).

In identifying levels for consideration for a potential annual average SO₂ standard, the PA also gives attention to the SO₂ concentrations at monitoring sites of influence identified in the trajectory-based analyses across different ranges of downwind ecoregion S deposition estimates. In the dataset for all 84 ecoregions in the contiguous U.S., the maximum annual average SO₂ 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,⁷⁰ were all below 15 ppb, and 75% of the monitor sites of influence concentrations were at or below 10 ppb (PA, Figure 7-3).⁷¹ In the subset of data for the 25 REA ecoregions with their upwind monitors, for the bin that includes deposition below 9 down to 6 kg/ha-yr, the concentrations for the metric based on maximum concentration at upwind sites of influence (EAQM-max) range as high as 15 ppb, with more than half below 10 ppb (PA, Figure 7-4, left panel). The EAQM-max concentrations associated with ecoregion median deposition in the lowest bins (S deposition below 6 kg/ha-yr) were all below 10 ppb. 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 is below 13 kg/ha-yr. When the highest EAQM-max concentration is at approximately 11 ppb, or 10 ppb, both the median and 90th percentile deposition are below 9 kg/ha-yr (PA, Figure 7-4).

The PA additionally discusses limitations associated with relating individual monitor SO₂ concentrations to S deposition in the context of the two metrics employed in the trajectory-based analyses. Between these metrics, somewhat stronger correlations were found for the annual average SO₂ weighted EAQM (which provides for proportional weighting of air concentrations from locations projected to contribute more heavily to a particular ecoregion), compared to the EAQM-max, particularly for the first two to three time periods of the 20-year period. This difference is related to the extent to which monitor concentrations can be indicative of atmospheric loading. The weighted EAQM is intended to more closely represent the atmospheric

⁷⁰ The bin for “<9 – 6 kg/ha-yr” is discussed here as it is the bin closest to the deposition target range of 10 or 8 to 5 kg/ha-yr identified above.

⁷¹ Figure 7-3 of the PA presents the pairs of median deposition estimates and associated upwind sites of influence EAQM-max SO₂ concentrations from the trajectory-based analysis in section 6.2.4 of the PA (specifically, the combined datasets presented in PA, Figures 6-40 and 6-41).

loading for the locations (and associated sources) of the contributing (sites of influence) monitors than a single contributing monitor can. However, the weighted metric is not directly translatable to a standard level (which is an upper limit on concentrations in individual locations).

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 correlations for the more recent period with its much lower magnitude of deposition and much smaller range in deposition (PA, section 7.2.2.3).

In its consideration of the trajectory-based analyses to identify a range of annual average SO₂ EAQM-max concentrations that may be associated with an ecoregion median S deposition range from 5 to 10 kg S/ha-yr, the PA recognizes several important considerations. First, monitor concentrations of SO₂ can vary substantially across the U.S., reflecting the distribution of sources, and other factors such as meteorology. This complicates consideration of how the EAQM-max, the maximum contributing monitor identified in the trajectory-based analysis (summarized in section II.B above and described more fully in section 6.2.4 of the PA) relates to S deposition levels in downwind ecosystems. Another consideration is the substantial scatter in the relationship between S deposition estimates and measured SO₂ concentrations with ecoregion median S deposition values below 5 kg/ha-yr. This scatter in the relationship between measured SO₂ concentration and S deposition estimates at these lower deposition levels contributes

increased uncertainty to conclusions regarding potential secondary standard SO₂ metric levels intended to relate to ecoregion median deposition levels at or below 5 kg/ha-yr (PA, section 7.2.2.3).

With regard to consideration of relationships between S deposition and PM_{2.5}, poor correlations were observed for total S deposition estimates with PM_{2.5} at the 27 Class I area sites ($r=0.33$, PA, Figure 6-31), with not much stronger correlations for ecoregion S deposition estimates with PM_{2.5} at upwind sites of influence from the trajectory-based analysis ($r=0.22$ and 0.48 , PA, Table 6-12). While the correlations in the trajectory-based analyses for deposition in eastern ecoregions were much higher ($r=0.83$ and 0.90), the coefficients were negative for deposition in western ecoregions. The PA concluded that the preponderance of western sites in the Class I area dataset (20 of the 27 sites) may be an influence on the low correlation observed for that dataset. Given that the analyses involving total S deposition and ambient air SO₄²⁻ concentrations are at remote locations (Class I areas), distant from sources of SO₂ emissions, and that that relationship is not stronger than that for SO₂ at the SLAMS, which are near sources monitoring SO₂ (the source for atmospheric SO₄²⁻), the PA found that the analyses did not indicate an advantage for an indicator based on SO₄²⁻ measurements (or SO₄²⁻ and SO₂ combined), such as is currently collected at CASTNET sites, or PM_{2.5} mass over options for a potential annual average standard metric focused on SO₂ concentrations (based on FRM/FEMs).⁷²

d. Nitrogen Deposition and N Oxides and PM

The subsections below summarize the evidence and exposure/risk-based considerations

⁷² It is also of note that use of SO₄²⁻ measurements, alone or in combination with SO₂ concentrations, as an indicator of a new standard would entail development of sample collection and analysis FRM/FEMs and of a surveillance network.

of the PA pertaining to N deposition and concentrations of N oxides and PM in ambient air.

These considerations draw on the available welfare effects evidence described in the current ISA (as well as prior ISAs and AQCDs), and discussed in Chapters 4, 5 and 6 of the PA. The focus of these considerations is primarily on N deposition and effects other than aquatic acidification (PA, sections 4.3, 5.2 and 5.3). As recognized in section II.D above, the PA finds S deposition to be the dominant influence on aquatic acidification risk in the 20-year period analyzed (2001-2020), based on the finding that the inclusion of acidic N deposition to the aquatic acidification risk analyses did not appreciably change patterns and percentages of waterbodies estimated to exceed CLs for the three ANC targets (PA, section 5.1.2.4).

In considering potential public welfare protection from N deposition-related effects (in light of the evidence summarized in sections II.C.1 and II.C.3 above), the PA recognizes that the effects of N deposition in both aquatic and terrestrial ecosystems have potential public welfare implications (PA, section 7.2.3.2). 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 broad State, local and national government engagement in activities aimed at assessing and reducing the loading (PA, section 5.2.3). 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, as well as non-use benefits to the public. The impacts of eutrophication relate to the consequence of the rapid and appreciable algal growth it fuels. Decomposition of the plant biomass from the

subsequent algal die-off contributes to reduced waterbody oxygen which, among other things, in turn contributes to fish mortality, and changes in aquatic habitat related to changes in resident plant and animal species (PA, section 4.3; ISA, Appendix 7).

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 primarily 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 HNO₃ effects on lichens (which might be considered to be “direct” effects or the result of deposition onto plant surfaces) are also not clear and might depend on the extent to which they impact whole communities, other biota, or ecosystem structure and function (PA, section 7.2.3.2).

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, the PA recognizes 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, such as N deposition that alters forest ecosystem community structures in ways that appreciably affect use and enjoyment of those

areas by the public (PA, section 7.2.3.2).

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 (including timber), which may be judged and valued differently 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 (PA, section 7.2.3.2).

(1) Quantitative Information for Ecosystem Risks Associated with N Deposition

The PA considers the available information regarding air quality and atmospheric deposition and risk or likelihood of occurrence of ecosystem effects under differing conditions. In so doing, the PA notes the varying directionality of some of the N enrichment-related effects in terrestrial ecosystems, such that some effects 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. The information is also considered with regard to the key limitations and associated uncertainties of this evidence.

Beginning with the appreciable evidence base documenting assessments of N loading to waterbodies across the U.S., the PA notes the waterbody-specific nature of such responses and the relative role played by atmospheric deposition, among other N sources. For example, the

relative contribution to such loading from atmospheric deposition compared to other sources (e.g., agricultural runoff and wastewater discharges) varies among waterbody types and locations, which 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, and aquatic habitat type, among others (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 several 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). 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 (PA, section 5.2.3). 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 II.E.1.d(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 (PA, section 5.2.2). Such seemingly subtle changes include whether P or N is the nutrient limiting phytoplankton growth (and productivity) and

shifts in phytoplankton community composition, for which public welfare implications are less clear (PA, section 7.2.3.1). 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 the organic material released by algae that is 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 (one of the chemicals associated with harmful blooms), 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, the PA recognizes 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 its consideration of the different types of quantitative analysis, the PA recognizes limitations particular to each, and associated uncertainties. Uncertainties associated with the soil acidification modeling analyses in the last review include those 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 (PA, section 5.3.4.1). Uncertainties associated with experimental addition analyses include the extent to which the studies reflect steady-state conditions, as well

as a lack of information regarding historic deposition at the study locations (PA, 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, a variety of factors, including the history of deposition and variability of response across the landscape, 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. (PA, section 5.3.4).

(2) General Approach for Considering Public Welfare Protection

As an initial matter, the PA notes 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 and their biota, structure, and function.

Further, the PA recognizes the contribution to N deposition of atmospheric pollutants

other than the criteria pollutants N oxides and PM, most significantly the contribution of NH₃ (PA, section 6.2.1). This contribution has increased since the last reviews of the NO₂ and PM secondary standards, as seen in Figures 6-17, 6-18 and 6-19 of the PA, reflecting increases in NH₃ emissions over that time period. These trends of increased NH₃ emissions and reduced N deposition coincide with decreasing trends in N oxides emissions and associated contributions of oxidized N to total N deposition (PA, 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 (PA, Figure 6-19). Further, dry deposition of NH₃ as a percentage of total N deposition at CASTNET sites ranges up to a maximum of 65% at the highest site in 2021 (PA, Figure 6-19). The 75th percentile for these sites is greater than 30% (i.e., at 25% of the CASTNET sites, more than 30% of N deposition is from dry deposition of NH₃). This is a noteworthy value given that these sites are generally in the West, with few in the areas of highest NH₃ emissions where the percentage would be expected to be higher still (PA, Figures 6-20 and 2-9).

In light of the contrasting temporal trends for emissions of oxidized and reduced N compounds, the PA observes that the influence of ambient air concentrations of N oxides and PM on N deposition appears to have declined over the past 20 years, complicating consideration of the protection from N deposition-related effects that can be provided by secondary NAAQS for these pollutants. Thus, the PA finds that NH₃, 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, is a complicating factor in considering policy options related to NAAQS for addressing ecological effects related to N deposition (e.g., PA, Figure 6-18 and 6-13).

In considering what the currently available quantitative information regarding terrestrial ecosystem responses to N deposition indicates about levels of N deposition that may be associated with increased concern for adverse effects, the PA focuses first on the evidence for effects of N deposition on trees derived from both experimental addition studies and observational studies of potential relationships between tree growth and survival and metrics for N deposition. With regard to the information available from experimental addition tree studies, the PA recognizes study limitations and associated uncertainties, while noting that the lowest forest N addition that elicited effects was 15 kg N/ha-yr over a 14-year period occurring from 1988-2002 (PA, sections 5.3.2 and 7.2.3.2 and Appendix 5B, Table 5B-1; McNulty et al., 2005). Based on the estimates from the array of observational studies, the PA finds that N deposition with a range of 7 to 12 kg/ha-yr, on a large area basis, may be a reasonable characterization of conditions for which statistical associations have been reported for terrestrial effects, such as reduced tree growth and survival and species richness of herbs and shrubs (PA, sections 5.3.4 and 7.2.3.2).

With regard to observational or gradient studies of N deposition and tree growth and survival (or mortality), the PA gave particular attention to three recently available studies that used the U.S. Forest Service 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. (PA, Appendix 5B, Figure 5B-1) and report associations of tree growth and/or survival metrics with various N deposition metrics for three different time periods. These studies provide 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 (PA, section 5.3.2.3 and Appendix 5B, section 5B.3.2). In considering

information from these studies, the PA notes 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 (PA, sections 5.3.2 and 7.2.3.2 and Appendix 5B). The largest study, which included 71 species with ranges across the U.S., 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 (PA, Appendix 5B, section 5B.3.2.3; Horn et al., 2018). The median deposition values across the sample sites for species with significant positive or negative associations generally ranged from 7 to 12 kg N/ha-yr (PA, section 5.3.2 and Appendix B, section 5B.3.2.3). Among the species for which the association varied from negative to positive across deposition levels, this is the range for those species for which the association was negative at the median deposition value (PA, section 5.3.4).⁷³

With regard to studies of herb and shrub community response, the PA notes a number of recently available studies report on addition experiments (PA, section 5.3.3.1 and Appendix 5B, section 5B.3.1). The lowest rate of N addition, in an addition study, 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 (PA, Appendix 5B, Table 5B-7; Clark and Tilman, 2008). Recent gradient studies of coastal sage scrub in southern

⁷³ This also excluded species for which sample sites were limited to the western U.S. based on recognition by the study authors of greater uncertainty in the west (Horn et al., 2018).

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 (PA, 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 (PA, section 5.3.3.1). Lastly, the PA notes the observational studies that have analyzed variation in lichen community composition in relation to indicators of N deposition (PA, section 5.3.3.2 and Appendix 5B, section 5B.4.2). 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, SO₂, and historical air quality and associated deposition (PA, section 5.3.3.2).

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 shifts in the composition of phytoplankton species communities 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 Bay (ISA, Appendix 7, section 7.2). These projects often use 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 II.C.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, respectively, and atmospheric loading estimated to be occurring in the fourth was below 5 kg/ha-yr (PA, section 7.3). This information, combined with the information from terrestrial studies summarized above, led to the PA identifying 7-12 kg/ha-yr as an appropriate N deposition range on which to focus in

considering policy options (PA, section 7.2.3.2).

(3) Relating Air Quality Metrics to N Deposition Associated with N Oxides and PM

In exploring how well various air quality metrics relate to N deposition, the analyses in Chapter 6 of the PA 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 downwind from the ambient air monitoring sites. The PA finds the analyses utilizing data from monitors using FRM/FEM to collect ambient air concentration data for evaluation with the NAAQS (e.g., to identify violations) to be particularly relevant given that the current standards are judged using design values derived from FRM/FEM measurements at existing SLAMS (PA, section 7.2.3.3). Given their role in monitoring for compliance with the NAAQS, most or many of these monitors are located in areas of relatively higher pollutant concentrations, such as near large sources of NO₂ or PM. Accordingly, the PA notes that information from these monitoring sites can help inform how changes in NO₂ 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 levels of potential concern.

In considering the information and findings of these analyses regarding relationships between N deposition and N oxides and PM in ambient air, the PA considers the current forms and averaging times of the secondary PM and NO₂ NAAQS. For N oxides, the current secondary standard is the annual average of NO₂, 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 quantitative air quality and N deposition analyses in the PA focus on 3-year average metrics (e.g., annual average NO₂ 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 of these air quality–deposition analyses 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, the PA notes that relationships between N deposition and NO₂ and PM air quality are affected by NH₃ emissions and non-N-containing components of PM (PA, section 6.4.2). The PA further notes that 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. (PA, section 6.2.1). Both of these factors are recognized to influence relationships between total N deposition and NO₂ and PM air quality metrics.

For total N deposition estimated for grid cells with collocated SLAMS monitors, the correlations with annual average NO₂ concentrations, averaged over three years, are generally low across all sites and particularly in the East (e.g., correlation coefficients below 0.4), although somewhat higher for sites in the West (PA, Table 6-6). This likely reflects the relatively greater role of NH₃ in N deposition in the East, which for purposes of the analyses in this PA extends across the Midwest (PA, section 6.4.2). For N deposition and NO₂ at upwind monitoring sites of influence, the correlation between estimates of total N deposition (wet plus dry) in eastern ecoregions and annual average NO₂ concentrations at monitor sites of influence (identified via trajectory-based modeling) for the five periods from 2001-2020 is low to moderate (correlation coefficients below 0.4, with the exception of one for EAQM-weighted in 2001-03 at 0.6), with the earlier part of the 20-year period, when NO₂ concentrations were higher and NH₃ emissions were lower (as indicated by Figures 6-6 and 6-5 of the PA) having relatively higher correlation than the later part (PA, Figures 6-6 and 6-4). The correlation is negative or near zero for the western ecoregions (PA, section 6.2.4).

The reductions in NO₂ emissions over the past 20 years have been accompanied by a reduction in deposition of oxidized N (PA, section 6.2.1). However, increases in NH₃ 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 NO₂ emissions and in deposition of oxidized N in the past 10 years there is a trend of increased NH₃ and increased deposition of reduced N (NH₃ and NH₄⁺), most particularly in areas of the Midwest, Texas, Florida and North Carolina (PA, Figures 6-16 and 6-17). The PA finds this to indicate that, while in the earlier years of the assessment period controls on NO₂ emissions may have resulted in reductions in deposition of oxidized N, in more recent years they have much less influence on total N deposition (PA, sections 6.2.1 and 6.4). In terms of ecoregion median statistics, the PA observes the decreasing trend in ecoregion median total N deposition across the period from 2001 through 2012, while taking note that from 2012 onward total N deposition increases, most particularly in ecoregions where most of the total deposition is from reduced N (PA, Figure 7-6).

The PA also considers the impact of increasing deposition of reduced N on the 20-year trend in total N deposition as illustrated by TDep estimates at the 92 CASTNET sites. At these sites, the PA observes that 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% (PA, Figure 6-19). Based on examination of the trends for components of reduced N deposition, the PA notes that the greatest influence on the parallel increase in N deposition percentage composed of reduced N is the increasing role of NH₃ dry deposition. The percentage of total N deposition at the CASTNET sites that is from NH₃ has increased, from a median below 10% in 2000 to a median somewhat above 25% in 2021 (PA, Figure 6-19).

Recognizing limitations in the extent to which CASTNET sites can provide information representative of the U.S. as a whole, the PA also analyzed TDep estimates for the most recent period (2018-2020) with regard to the percent of total N deposition represented by reduced N across the U.S. In areas with ecoregion median total N deposition above 9 kg/ha-yr (PA, Figure 7-7, upper panel), the ecoregion median percentage of total N deposition composed of reduced N is greater than 60% (PA, Figure 7-7, lower panel). The PA further notes that recent (2019-2021) TDep estimates across individual TDep grid cells similarly show that areas of the U.S. where total N deposition is highest, and where it is greater than potential N deposition targets identified in section 7.2.3.2 of the PA, are also the areas with the greatest deposition of NH₃ (PA, Figure 7-8), comprising more than 30% of total N deposition. That is, the PA finds that NH₃ driven deposition is greatest in regions of the U.S. where total deposition is greatest (PA, section 7.2.3.3).

Turning to PM_{2.5}, the PA notes that, as with NO₂ concentrations, the correlation for ecoregion median N deposition and PM_{2.5} concentrations at upwind sites of influence is better for eastern ecoregions than western ecoregions, for which there is no correlation at all (PA, section 6.2.4). For total N deposition and PM_{2.5} concentrations at SLAMS, a low to moderate correlation is observed, also slightly higher at eastern versus western sites (PA, section 6.2.3). In considering the two factors mentioned above (NH₃ emissions and non-N containing components of PM), the PA notes that some NH₃ transforms to NH₄⁺, which is a component of PM_{2.5}, while also noting that, in the areas of greatest N deposition, the portion represented by deposition of gaseous NH₃ generally exceeds 30%. Additionally, while NH₃ emissions have been increasing over the past 20 years, the proportion of PM_{2.5} that is composed of N compounds has declined. The median percentage of PM_{2.5} comprised by N compounds has 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% (PA, section 6.4.2 and Figure 6-56). Further, this percentages varies regionally, with sites in the nine southeast states having less than 10% of PM_{2.5} mass composed of N compounds (PA, Figure 6-56).

In summary, the PA concludes that in recent years, NH₃, 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 of the PA). The PA finds that this situation — of an increasing, and spatially variable, portion of N deposition not being derived from N oxides or PM — complicates assessment of policy options for protection against ecological effects related to N deposition associated with N oxides and PM, and for secondary standards for those pollutants that may be considered to be associated with a desired level of welfare protection. That notwithstanding, recognizing that between the NO₂ primary and secondary NAAQS, the 1-hour primary standard (established in 2010) may be the more controlling on ambient air concentrations, the PA considered analyses of SLAMS air quality data with regard to trends in annual average NO₂ concentrations (PA, Figure 7-9) and relationships between annual average NO₂ concentrations (in a single year and averaged over three years) and design values for the existing 1-hour primary standard (PA, Figure 7-10). In so doing, the PA noted that subsequent to 2011-2012, when ecoregion median N deposition levels in 95% of the eastern ecoregions of the continental U.S.⁷⁴ have generally been at/below 11 kg N/ha-yr, annual average NO₂ concentrations, averaged across three years, have been at/below 35 ppb (PA, Figures 7-6 and 7-9). Further, the SLAMS data indicate that single-year annual average NO₂

⁷⁴As noted in the PA, the eastern designation used throughout PA includes areas generally considered the Great Plains, while the West includes the states of ND, SD, CO, WY, MT, AZ, NM, UT, ID, CA, OR, WA (PA, p. 5-20).

concentrations, averaged over three years, in areas that meet the current 1-hour primary standard have generally been below approximately 35 to 40 ppb (PA, Figure 7-10). The PA recognizes, however, that this information as a whole also suggests the potential for future reductions in N oxide-related N deposition to be negated by increasing reduced N deposition.

Further, the PA notes that 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, but also notes 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 (e.g., organic and elemental carbon) as part of the PM_{2.5} mass (PA, section 7.2.3.3). 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 (PA, section 6.3 and 6.4)

In considering relationships between air quality metrics based on indicators other than those of the existing standards and N deposition (and associated uncertainties), the PA drew on the analyses of relationships for collocated measurements and modeled estimates of N compounds other than NO₂ 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. (PA, sections 6.2.2, 6.3 and 6.4.2). The analyses indicate that total N deposition (TDep) in these rural areas has a moderate correlation with air concentrations of nitric acid and particulate nitrate for the 20-year dataset (2000-2020) (PA, Figure 6-32). The correlations are comparable to the correlation of NO₂ 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. The PA notes that deposition at the western U.S sites is generally less affected by NH₃ (PA, section 6.4.2). Further, the observed

trend of increasing contribution to N deposition of NH₃ 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 PA concludes that 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 NH₃ is prevalent (PA, section 7.2.3.3).

The analyses involving N deposition and N-containing PM components at the 27 Class I area sites yield similar correlation coefficients as those for N deposition (TDep) and PM_{2.5} at SLAMS monitors (PA, section 7.2.3.3 and Figures 6-33, 6-39 [upper panel], and 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 timeframe (PA, Figure 6-33). Thus, the PA concludes that 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} (PA, section 7.4). As a whole, the PA finds that 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 used for the existing standards (PA, section 7.2.3.3). The PA also notes that use of the NO₃⁻ or particulate N measurements analyzed with deposition estimates at the 27 Class I area sites, alone or in combination with NO₂, as an indicator for a new standard would entail development of sample collection and analysis FRM/FEMs⁷⁵ and of a surveillance network.

⁷⁵ For example, sampling challenges have long been recognized for particulate NH₄⁺ (e.g., ISA, Appendix 2, sections 2.4.5; 2008 ISA, section 2.7.3).

2. CASAC Advice and Public Comments

In evaluating the adequacy of the current secondary standards for SO_x, oxides of N, and PM, in addition to evidence and air quality/exposure/risk-based information discussed above, we take note of the advice and recommendations of the CASAC, based on its review of the draft ISA and draft PA, as well as comments from the public. A limited number of public comments have been received in the docket for this review to date, including just a few comments on the draft PA, that primarily focused on technical analyses and information, which were considered in developing the final PA (PA, section 1.4). 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 the CASAC's review of the draft PA.

In reviewing the draft PA, the CASAC first 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.” Accordingly, the CASAC 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.

With regard to protection from effects other than those associated with ecosystem deposition of S and N compounds, the CASAC concluded that the existing SO₂ and NO₂

secondary standards provide adequate protection for direct effects of those pollutants on plants and lichens, providing consensus recommendations that these standards should 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, with their advice divided between a majority opinion and a minority opinion. Advice conveyed from both the majority and minority 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 SO₂ standard with a level in the range of 10 to 15 ppb,⁷⁶ which these members concluded would generally maintain ecoregion median S deposition below 5 kg/ha-yr⁷⁷ based on consideration of the trajectory-based SO₂ analyses (and associated figures) in the draft PA (Sheppard, 2023, Response to Charge Questions, p. 25). They concluded that such a level of S deposition would afford protection for tree and lichen species⁷⁸ and aquatic ecosystems. Regarding aquatic ecosystems, these members cited the ecoregion-scale estimates

⁷⁶ Although the CASAC letter does not specify the form for such a new annual standard, the justification provided for this recommendation cites two figures in the draft PA (Figures 6-17 and 6-18) which presented annual average SO₂ concentrations averaged over three consecutive years (Sheppard, 2023, Response to Charge Questions, p. 25). Therefore, we are interpreting the CASAC majority recommendation to be for an annual standard, averaged over three years.

⁷⁷ Although the CASAC letter does not specify the statistic for the 5 kg/ha-yr value, the PA analyses referenced in citing that value, both the trajectory analyses and the ecoregion-scale summary of aquatic acidification results, focus on ecoregion medians. Therefore, we are interpreting the CASAC advice on this point to pertain to ecoregion means.

⁷⁸ In making this statement, these CASAC members cite two observational data studies with national-scale study areas published after the literature cut-off date for 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 U.S. Forest Service sites from 1990 to 2012. The tree study by Pavlovic et al. (2023) uses 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).

(from the aquatic acidification REA analyses) associated with median S deposition bins for the 90 ecoregion-time period combinations (PA, section 5.1.3.2) 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 $\mu\text{eq/L}$, respectively, in all ecoregion-time period combinations (Sheppard, 2023, Response to Charge Questions, p. 25).⁷⁹ In recommending an annual SO₂ standard with a level in the range of 10 to 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 SO₂ concentrations near industrial sources in 2019, 2020, and 2021,” citing Figure 2-25 of the draft PA⁸⁰ (Sheppard, 2023, Response to Charge Questions, p. 24).

One CASAC member dissented from this recommendation for an annual SO₂ standard⁸¹ and instead recommended adoption of a new 1-hour SO₂ 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 implementation of the existing primary SO₂ standard (established in 2010), indicating the more recent lower deposition to be a

⁷⁹ As seen in Tables 3 and 4 in this preamble, 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 (PA, section 7.2.2.2, Table 7-1).

⁸⁰ The figure cited by the CASAC majority is the prior version of Figure 2-28 in section 2.4.2 of the final PA. The figure presents temporal trend in distribution (box and whiskers) of annual average SO₂ concentrations at SLAMS.

⁸¹ 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 the CASAC (Sheppard, 2023, Response to Charge Questions, p. 23). The former is a Panel formed for this review, while the latter is the standing Committee specified in the CAA.

product of current regulatory requirements (Sheppard, 2023, Appendix A, p. A-2).⁸²

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 NO₂ standard to a level “<10 – 20 ppb” (Sheppard, 2023, Response to Charge Questions, p.24). The justification these members provided was related to their consideration of the relationship presented in the draft PA of median ecosystem N deposition with the weighted⁸³ annual average NO₂ 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 recognized, however, that “when considering all ecoregions, there is no correlation between annual average NO₂ and N deposition” (Sheppard, 2023, Response to Charge Questions, p. 24). Their focus on total N deposition estimates at or below 10 kg/ha-yr appears to relate to consideration of TMDL 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 6.9⁸⁴ kg/ha-yr, respectively, and atmospheric loading estimated in the fourth was below 5 kg/ha-yr (Sheppard, 2023, Response to Charge Questions, pp. 12-14). These members also

⁸² This member stated that the existing primary NAAQS for the three pollutants were significantly more restrictive than the existing secondary standards and provide adequate protection for deposition-related effects (Sheppard, 2023, Appendix A).

⁸³ 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 (PA, section 6.2.4). Values of this metric are not directly translatable to individual monitor concentrations or to potential standard levels.

⁸⁴ The CASAC letter states that the Neuse River Estuary TMDL specified a 30% reduction from the 1991-1995 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).

conclude 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 NO₂ in ambient air” (Sheppard, 2023, Response to Charge Questions, p. 23).

One CASAC member disagreed with revision of the existing annual NO₂ standard and instead recommended adoption of a new 1-hour NO₂ 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 implementation of the existing primary standards for NO₂ and PM (Sheppard, 2023, Appendix A). This member additionally noted that bringing into attainment the areas still out of attainment with the 2013 primary annual PM_{2.5} standard (12.0 µg/m³) will provide further reductions in N deposition. This member also noted his analysis of NO₂ annual and 1-hour design values for the past 10 years (2013-2022) as indicating that the current primary NO₂ standard provides protection for annual average NO₂ 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 PM₁₀ 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 µg/m³. In their justification for this range, these members focus on rates of total N deposition at or below 10 kg/ha-yr and total S deposition at or below 5 kg/ha-yr 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 cited 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 (Horn et al., 2018), which is characterized in the ISA and discussed in sections 5.3.2.3 and 7.2.2.2 of the PA (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), are 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 µg/m³), this group of CASAC members made several statements regarding PM_{2.5} annual concentrations and estimates of S and N deposition for which they cited several figures in the draft PA. Citing figures in the draft PA with TDep deposition estimates and IMPROVE and CASTNET monitoring data, they stated that “[i]n remote areas, IMPROVE PM_{2.5} concentrations in the range of 2-8 µg/m³ 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 cited 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 µg/m³ (Sheppard, 2023, Response to Charge Questions, pp. 23-24). These members additionally cited 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 µg/m³, 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/m³,⁸⁵ and PM_{2.5} EAQM-max values below 8 µg/m³ (Sheppard, 2023, Response to Charge Questions, pp. 23-24). The CASAC also noted 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 earlier in this section.

One CASAC member recommended revision of the annual secondary PM_{2.5} standard to a level of 12 µg/m³ 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 observed 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 concluded this to indicate that the 2013 primary annual PM_{2.5} standard of 12.0 µg/m³ 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 recommended revision of the level to 25 ug/m³ or revision of the indicator and level to deciviews⁸⁶ and 20 to 25, respectively (Sheppard, 2023, Response to Charge Questions, p 25). These members variously cited “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 did not provide further specificity regarding their

⁸⁵ As noted earlier in this section, weighted EAQM values are not directly translatable to concentrations at individual monitors or to potential standard levels.

⁸⁶ Deciviews, units derived from light extinction, are frequently used in the scientific and regulatory literature to assess visibility (U.S. EPA 2019, section 13.2).

reference to lichen species and fog or cloud water. With regard to visibility impairment, these members described the EPA solicitation of comments that occurred with the separate EPA action to reconsider the 2020 decision to retain the existing PM_{2.5} standards as the basis for their recommendations on the secondary 24-hr PM_{2.5} standard (Sheppard, 2023, Response to Charge Questions, p 25; 88 FR 5562-5663, January 27, 2023).⁸⁷ One CASAC member dissented from this view and supported retention of the existing secondary 24-hr PM_{2.5} standard.

Among the CASAC comments on the draft PA⁸⁸ 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. Accordingly, in the final PA, a number of aspects of chapters 4 and 5 were revised from the draft PA; these changes took into account the information emphasized by the CASAC while also referring to the ISA and studies considered in it (PA, section 7.3). More recent studies cited by the CASAC generally concerned 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 (PA, section 7.3).

3. Administrator's Proposed Conclusions

In considering the adequacy of the existing secondary standards for SO_x, N oxides, and

⁸⁷ Protection from impairment of visibility effects was one of the welfare effects within the scope of the PM NAAQS reconsideration rather than the scope of this review (U.S. EPA, 2016, 2017). In that action, the Administrator proposed not to change the 24-hour secondary PM NAAQS for visibility protection and also solicited comment on revising the level of the current secondary 24-hour PM_{2.5} standard to a level as low as 25 µg/m³; in the final action, the Administrator concluded that the current secondary PM standards provide requisite protection against PM-related visibility effects and retained the existing standards without revision (88 FR 5558, January 27, 2023; 89 FR 16202, March 6, 2024).

⁸⁸ 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 of the PA.

PM, and what revisions or alternatives are appropriate, the Administrator has drawn on the ISA conclusions regarding the weight of the evidence for both the direct effects of SO_x, N oxides, and PM in ambient air and for effects associated with ecosystem deposition of N and S compounds, and associated areas of uncertainty; quantitative analyses of aquatic acidification risk and of air quality and deposition estimates, and associated limitations and uncertainties; staff evaluations of the evidence, exposure/risk information, and air quality information in the PA; CASAC advice; and public comments received thus far in the review. In considering the available information in this review, the Administrator recognizes the evidence of direct biological effects associated with elevated short-term concentrations of SO_x and N oxides that formed the basis for the existing secondary SO₂ and NO₂ standards, the evidence of ecological effects of PM in ambient air, primarily associated with loading on vegetation surfaces, and also the extensive evidence of ecological effects associated with atmospheric deposition of N and S compounds into sensitive ecosystems. The Administrator also takes note of the quantitative analyses and policy evaluations documented in the PA that, with CASAC advice, inform his judgments in reaching his proposed decision on the secondary standards for SO_x, N oxides, and PM that provide the requisite protection under the CAA.

In reaching his proposed conclusions for the pollutants included in this review, the Administrator considers first the secondary standard for SO_x and the adequacy of the existing standard for providing protection of the public welfare from direct effects on biota and from ecological effects related to ecosystem deposition of S compounds. In so doing, he considers the evidence regarding direct effects, as described in the ISA and evaluated in the PA, which is focused on SO₂. He takes note of the PA finding that the evidence indicates SO₂ concentrations associated with direct effects to be higher than those allowed by the existing SO₂ secondary

standard (PA sections 5.4.1, 7.1.1 and 7.4). Additionally, he takes note of the CASAC unanimous conclusion that the existing standard provides protection from direct effects of SO_x in ambient air, as summarized in section II.E.2 above. Based on all of these considerations, he judges the existing secondary SO₂ standard to provide the needed protection from direct effects of SO_x. He next turns to consideration of ecological effects related to ecosystem deposition of S compounds.

With regard to S deposition-related effects, as an initial matter, the Administrator recognizes the long-standing evidence of the role of SO_x in ecosystem acidification and related ecological effects. While he additionally notes the ISA determinations of causality for S deposition with two other categories of effects related to mercury methylation and sulfide phytotoxicity (ISA, Table ES-1; PA, section 4.4), he recognizes that quantitative tools and approaches are not well developed for ecological effects associated with atmospheric deposition of S other than ecosystem acidification (PA, section 7.2.2.1). Thus, he gives primary attention to effects related to acidifying deposition, given the robust evidence base and available quantitative tools, as well as the longstanding recognition of impacts in acid-sensitive ecosystems across the U.S. Accordingly, the Administrator takes note of the findings of the aquatic acidification REA and related policy evaluations in the PA. As summarized in the PA, the REA findings include that the range of ecoregion median deposition estimates across the contiguous U.S. analyzed during the 20-year period from 2001 through 2020 extended up to as high as 20 kg S/ha-yr, and that, except for one occasion (in 2011), the existing secondary SO₂ standard was met in all states but Hawaii. Further, other than the design value in 2011, design values for the existing SO₂ standard (second highest 3-hour average in a year) were well below its current level of 500 ppb (PA, section 6.2.1). For example, in the earliest 3-yr period analyzed (2001-03), when virtually

all design values for the existing 3-hour secondary standard were below 400 ppb and the 75th percentile of design values was below 100 ppb (PA, 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 (PA, Figure 6-11). The PA also notes that the magnitude of S deposition estimates at the 90th 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 2; PA, Figure 7-2). The Administrator also takes note of the aquatic acidification risk estimates that indicate that this pattern of S deposition, estimated to have occurred during periods when the existing standard was met (e.g., 2000-2002), is associated with 20% to more than half of waterbody sites in affected eastern ecoregions⁸⁹ being unable to achieve even the lowest of the three acid buffering capacity targets or benchmarks (ANC of 20 µeq/L), and judges such risks to be of public welfare significance.

The Administrator also considers the advice from the CASAC in considering deposition-related effects of S. Although the CASAC provided two sets of advice regarding standards for protecting from such effects, both the majority and the minority of CASAC recommended adoption of a new SO₂ standard for this purpose in light of conclusions that the existing standard did not provide such needed protection. Thus, based on the findings of the REA, associated policy evaluations in the PA with regard to S deposition and acidification-related effects in sensitive ecosystems, and in consideration of advice from the CASAC, the Administrator

⁸⁹ 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 (PA, Table 5-4). Ecoregion median deposition estimates were at or below 2 kg/ha-yr in all eight western ecoregions (PA, Table 5-3).

proposes to judge that the current SO₂ secondary standard is not requisite to protect the public welfare from adverse effects associated with acidic deposition of S compounds in sensitive ecosystems.

Having reached this proposed conclusion that the existing secondary SO₂ standard does not provide the requisite protection of the public welfare from adverse S deposition-related effects, most prominently those associated with aquatic acidification, the Administrator next considers options for a secondary standard that would provide the requisite protection from S deposition-related effects. In so doing, he turns first to the policy evaluations and staff conclusions in the PA, and the quantitative analyses and information described in Chapter 5 of the PA, for purposes of identifying S deposition rates that might be judged to provide an appropriate level of public welfare protection from acidification-related effects. In this context, he takes note of the PA focus on the aquatic acidification risk estimates and the PA recognition of linkages between watershed soils and waterbody acidification, as well as terrestrial effects. He concurs with the PA view that 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. Accordingly, he considers the PA evaluation of the risk estimates in terms of waterbodies estimated to achieve the three acid buffering capacity benchmarks (20, 30 and 50 µeq/L). Further, he concurs with the PA consideration of the ecosystem-scale estimates as appropriate for his purposes in identifying conditions that provide the requisite protection of the public welfare.

In focusing on the ecoregion-scale findings of the aquatic acidification REA, with particular attention to the 18 well studied, acid-sensitive eastern ecoregions, the Administrator considers the PA evaluation of 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 would achieve ANC values at or above the three benchmarks (20, 30 and 50 $\mu\text{eq/L}$), as summarized in Tables 7-1 and 5-5 of the PA. In so doing, he recognizes a number of factors, as described in the PA, which contribute variability and uncertainty to waterbody estimates of ANC and to interpretation of acidification risk associated with different values of ANC (PA, section 5.1.4 and Appendix 5A, section 5A.3).

The Administrator additionally takes note of the approach taken by the CASAC majority in considering the ecoregion-scale risk estimates (summarized in section II.E.2 above). These members considered the summary of results for the ecoregion-scale analysis of ecoregion median deposition bins (in the draft PA⁹⁰) and focused on a level of deposition (at or below 5 kg/ha-yr) estimated to achieve acid buffering capacity at or above the three ANC benchmarks in 80% (for ANC of 20 and 30 $\mu\text{eq/L}$) or 70% (for ANC of 50 $\mu\text{eq/L}$) of waterbodies in all ecoregion-time period combinations⁹¹ (Sheppard, p. 25 of the Response to Charge Questions). As additionally recognized in the PA, the results for ecoregion-time period combinations for median S-deposition in the 18 eastern ecoregions at or below 7 kg/ha-yr also indicate these percentages of waterbodies achieving the three ANC benchmarks (as seen in Tables 7-1 and 5-5 above).⁹² The results for median S deposition at or below 7 kg/ha-yr further indicate that 90% of waterbodies

⁹⁰ While the final PA provides additional presentations of aquatic acidification risk estimates, including those at the ecoregion-scale, the estimates are unchanged from those in the draft PA (PA, section 5.1.3).

⁹¹ The presentation of such percentages in the draft PA (reviewed by the CASAC) were specific to the 90 ecoregion-time period combinations for the 18 eastern ecoregions. Inclusion of the 7 western ecoregions would yield higher percentages, as more than 90% of waterbodies in those ecoregions were estimated to achieve all three ANC concentration in all time periods (PA, Table 5-4).

⁹² Ecoregion median deposition was below 2 kg S/ha-yr in all 35 ecoregion-time period combinations for the eight western ecoregions (PA, Table 5-4).

per ecoregion achieve ANC at or above 20, 30 and 50 $\mu\text{eq/L}$ 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 eastern ecoregions meeting or exceeding the ANC benchmarks declines to 87%, 81% and 72% (as summarized in section II.E.1.c(2) above), and the percentages for all 25 analyzed ecoregions is higher.

The Administrator additionally considers the PA evaluation of 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. As summarized in section II.E.1.b above, based on the observation of appreciably improved acid buffering capacity (i.e., increased ANC) estimates by the third time period (2010-2012), the PA focused on the REA risk and deposition estimates for this and subsequent periods. The S deposition estimated to be occurring in the 2010-2012 time period included ecoregion medians (across 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 5 and Figure 2 above). For this pattern of deposition, more than 70% of waterbodies per ecoregion are estimated to be able to achieve an ANC of 50 $\mu\text{eq/L}$ in all 25 ecoregions (Figure 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 $\mu\text{eq/L}$ (Figure 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 $\mu\text{eq/L}$ 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 $\mu\text{eq/L}$ (Figure 1, right panel).

The Administrator observes that the estimates of acid buffering capacity achievement for the 2010-12 period deposition — achieving the ANC benchmarks in at least 70% to 80% (depending on the specific benchmark) of waterbodies per ecoregion — are consistent with the objectives identified by the CASAC majority (in considering estimates from the ecoregion-scale analysis). The advice from the CASAC majority emphasized ecoregion ANC achievement estimates of 70%, 80% and 80% for ANC benchmarks of 50, 30 and 20 $\mu\text{eq/L}$, respectively. The estimates for the later time period are somewhat higher, with all ecoregions estimated to achieve the ANC benchmarks in at least 80% to 90% (depending on the benchmarks) of waterbodies per ecoregion. In his consideration of these ANC achievement percentages identified by the CASAC, the Administrator notes the variation across the U.S. waterbodies with regard to site-specific factors that affect acid buffering (as summarized in sections II.C.1.b(1) and II.D.1 above and section 5.1.4 of the PA). Based on this and the CASAC majority advice, the Administrator concurs with the PA conclusion that 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.

With regard to the variation in deposition across areas within ecoregions, the Administrator notes the PA recognition 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. He further takes note of the PA observation of an appreciable reduction across the 20-year analysis period in the 90th percentile deposition estimates, as well as the median, for REA sites in each of the 25 ecoregions analyzed. Although the ecoregion 90th 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 (PA, Figure 7-2). In light of this trend, as well as the temporal trend in the REA estimates, the Administrator takes note of the PA findings that 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 benchmarks) might be expected to be met when ecoregion median and upper (90th) percentile deposition estimates at sensitive ecoregions are generally at and below about 5 to 8 kg/ha-yr. He additionally takes note of the PA recognition of uncertainties associated with the deposition estimates at individual waterbody sites and with the associated estimates of aquatic acidification risk (PA, section 5.1.4), and with the PA's consideration of estimates from the case study analyses, which together leads the PA to identify deposition rates at and below about 5 to 8 or 10⁹³ kg/ha-yr as associated with a potential to achieve acid buffering capacity benchmarks in an appreciable portion of acid sensitive areas. Based on all of these considerations, the Administrator focuses on this range of deposition levels in turning his attention to identification of a secondary standard that might be associated with S deposition of such a magnitude.

In considering options for a secondary standard based on consideration of S deposition-related effects, the Administrator takes note of the complexity of identifying a national ambient air quality standard focused on protection of the public welfare from adverse effects associated with national patterns of atmospheric deposition (rather than on protection from direct exposure from patterns of ambient air concentrations of concern). As more specifically described in the

⁹³ 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 (PA, section 7.4).

PA, 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, the Administrator concurs with the PA judgment that 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 SO₂ concentrations at ambient air monitors and S deposition rates in sensitive ecosystems of interest. Based on these considerations in the PA, the Administrator concurs that to achieve a desired level of protection from aquatic acidification effects associated with S deposition in sensitive ecosystems, SO₂ emissions must be controlled at their sources, and that associated NAAQS compliance monitoring is at regulatory SO₂ monitors generally sited near large SO₂ emissions sources.

Further, the Administrator considers findings of the PA analyses of relationships between ambient air concentrations and S deposition estimates, conducted in recognition of the variation across the U.S. in the source locations and magnitude of SO_x emissions, as well as the processes that govern transport and transformation of SO_x to eventual deposition of S compounds. These analyses consider relationships between S deposition estimates and SO₂ concentrations near SO₂ monitors (both at NAAQS regulatory monitors, which are often near large sources of SO₂ emissions, and in remote Class I areas) as well as relationships between ecoregion S deposition estimates and SO₂ concentrations at upwind sites of influence, identified by trajectory analyses to account for the relationship between upwind concentrations near sources and deposition in downwind areas (section II.B above and PA, sections 6.2.2 through 6.2.4.). As evidence of the

influence of SO₂ in ambient air on S deposition, all of these analyses demonstrated there to be an association between SO₂ concentrations and nearby or downwind S deposition (PA, section 7.4). The PA found the correlation coefficients to be 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 (PA, Chapter 6).

With regard to an indicator for a standard to address the effects of S deposition, the Administrator also takes note of the findings of the PA analyses and the parallel trends of SO₂ emissions and S deposition in the U.S. over the past 20 years that indicate the strong influence of SO₂ in ambient air on S deposition (PA, sections 6.4.1 and 7.4). The array of air quality analyses in the PA together illustrate the fact that atmospheric loading is a primary, but not the only, 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. The distribution of monitor SO₂ concentrations is appreciably flatter in the latter 10 years of the analysis period in comparison to the initial years, and S deposition rates during the latter 10 years are appreciably reduced from those in the earlier decade (PA, Figure 7-5). These parallel patterns indicate a broader distribution of concentrations across NAAQS monitors during these years. Additionally, the Administrator notes the PA finding of parallel temporal trends of ecoregion S deposition estimates and the REA aquatic acidification risk estimate across the five time periods analyzed.

In light of all of the linkages connecting SO_x emissions and S deposition-related effects, the Administrator considers the current information with regard to a new or revised standard for SO_x that would be expected to provide protection from aquatic acidification-related risks of S

deposition in sensitive ecoregions. With regard to the indicator for such a standard, he notes the PA findings of support for SO₂ as a good indicator for a secondary standard to address S deposition (PA, sections 6.4.1 and 7.4). This support includes the declining trend of S deposition that is consistent with and parallel to the sharp declines in annual average SO₂ emissions across the 20-year period, as well as the general association of SLAMS with higher annual average SO₂ concentrations with higher local S deposition estimates in addition to the high correlations observed for ecoregion median S deposition with upwind SO₂ monitoring sites of influence in the EAQM analyses. In light of all of these considerations, the Administrator judges SO₂ to be the appropriate indicator for a standard addressing S deposition-related effects.

With regard to the appropriate averaging time and form for such a standard, the Administrator takes note of the PA findings and advice from the CASAC. In the quantitative analyses of air quality and deposition, the PA generally focused on a year's averaging time based on the recognition that longer-term averages (such as over a year) most appropriately relate to deposition and associated ecosystem effects. The PA analyses also used a 3-year average form based on a recognition in the NAAQS program that such a form affords stability to the associated air quality management program that contributes to effective environmental protection. Similarly, in the advice of the CASAC majority on a standard addressing S deposition, these members recommended an annual average standard, and the information considered by the CASAC majority in drawing its conclusion also focused on an annual average SO₂ metric with a form that involved averaging over three consecutive years, implying that to be the recommended form (section II.E.2 above). In consideration of these conclusions of the PA and the CASAC majority, the Administrator focuses on annual average SO₂ concentrations, averaged over three

years, as providing an averaging time and form⁹⁴ that he judges appropriate for providing public welfare protection from adverse effects associated with long-term atmospheric deposition of S compounds.

In turning to consideration of a level for such a standard, as an initial matter, the Administrator again notes the complexity (recognized above) associated with identifying a national ambient air quality standard focused on protection from national patterns of atmospheric deposition rather than on protection from patterns of direct exposure to SO₂. As summarized in the PA, atmospheric deposition rates are a function of atmospheric loading, transformation, and transport, and are not a one-to-one function of concentration at a specific monitoring location. Thus, the maximum concentration at a single upwind site is less important for total loading than the combined impact of all upwind emissions sources. This contributes uncertainty to the identification of the appropriate level for a national standard based on a single maximum concentration that, if occurring at any one or multiple locations, would be expected to constrain areawide deposition rates downwind to the desired level for protection. The atmospheric loading (and deposition) associated with the maximum concentration conceptually represented by a standard level depends on the number and spatial distribution of areas exhibiting that concentration. Reductions in deposition reflect geographically-broad emissions reductions and weighted concentration reductions (e.g., EAQM-weighted) more than reductions in the maximum concentration at individual locations. As shown by the 20-year trends in annual emissions and monitor annual average SO₂ concentrations, the percentage reductions in

⁹⁴ 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).

deposition and emissions are greater than those in the highest monitor concentrations.⁹⁵

Particularly in this case of identifying a standard to provide a pattern of ambient air concentrations that together contribute to deposition across the U.S., it is important to consider the distribution of air concentrations to which the standard will apply. The Administrator takes note of this complexity and associated uncertainty in his identification of a level for an annual average SO₂ standard for S deposition.

In considering an appropriate range of concentrations for a level for such a standard, the Administrator considers the evaluations and associated findings of the PA and advice from the CASAC. In considering the PA analyses and evaluation, the Administrator takes note of the uncertainties associated with potential limitations in the monitoring dataset across the 20-year period (e.g., with regard to the representation of source locations in the earlier years of the monitoring data), in addition to the complexities described above. In so doing, the Administrator considers the two options identified in the PA for a level of an annual average standard, with a 3-year average form. One option identified in the PA would establish a level in the range of somewhat below 15 ppb to a level of 10 ppb, and a second option would establish a standard with a level within the range of 10 to 5 ppb. He additionally recognizes there to be uncertainties in aspects of the aquatic acidification risk modeling that contribute uncertainty to the resulting estimates, and in the significance, of aquatic acidification risk, which he finds to be greater with lower deposition levels (PA, section 5.1.4). Further, the Administrator takes note of the additional and appreciably greater uncertainty recognized in the PA to be associated with

⁹⁵ As recognized in section II.E.1 above, in relating atmospheric loading to individual monitor concentrations, the higher correlations of the EAQM-weighted than the EAQM-max likely reflect the weighting of concentrations across multiple upwind monitors, with the trajectory analysis providing one approach that relates contributions from individual monitor locations to deposition in receiving ecosystems (without explicitly addressing the multiple factors at play).

consideration of a standard level below 10 ppb, including uncertainties in the relationships between S deposition and annual average SO₂ concentrations below 10 ppb (PA, Chapter 6, section 7.4). In general, there is uncertainty in identifying a specific level 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. In this context and based on the PA findings, the Administrator recognizes there to be, on the whole across the various linkages, increased uncertainty for lower SO₂ concentrations and S deposition rates.

The Administrator additionally considers the CASAC majority recommended range of levels for an annual average SO₂ standard to address S deposition-related ecological effects. As described in section II.E.2 above, the majority of the CASAC recommended adoption of an annual SO₂ standard with a level within the range of 10 to 15 ppb. These members indicated that this range of levels was generally associated with S deposition below 5 kg/ha-yr during the 2014-2016 and 2018-2020 trajectory analysis periods in the PA. The CASAC majority further 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 indicated to be associated with relatively high annual average SO₂ concentrations observed in 2019-2021 near a location of industrial sources (Sheppard, Response to Charge Questions, pp. 24-25).

Thus, based on analyses and evaluations in the PA, including judgments related to uncertainties in relating ambient air concentrations to deposition estimates for the purpose of

identifying a standard level associated with a desired level of ecological protection, and based on advice from the CASAC majority, the Administrator judges that a level within the range from 10 to 15 ppb would be appropriate for an annual average SO₂ standard requisite to protect the public welfare from adverse effects related to S deposition.

The Administrator also considered the extent to which a new annual average standard might be expected to control short-term concentrations (e.g., of three hours duration) and accordingly provide protection from direct effects that is currently provided by the existing 3-hour secondary standard. In this context, he notes the analyses and conclusions of the PA with regard to 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 SO₂ standard. These analyses indicate that in areas and periods when the annual SO₂ concentration (annual average, averaged over three years) is below 15 ppb, design values for the existing 3-hour standard are well below the existing secondary standard level of 0.5 ppm SO₂ (PA, Figure 2-29). Based on these findings of the PA, the Administrator proposes that it is appropriate to consider revision of the existing secondary SO₂ standard to an annual standard, with a 3-year average form and a level in the range from 10 to 15 ppb.

The Administrator also takes note of the recommendation from the CASAC minority to establish a 1-hour SO₂ secondary standard, identical to the primary standard, based on its observation that most of the S deposition estimates for the last 10 years are less than 5 kg/ha-yr and judgment that this indicates that the existing 1-hour primary SO₂ standard adequately protects against long-term annual S deposition-related effects. Accordingly, the CASAC minority recommended setting the secondary SO₂ standard equal to the current primary standard (section II.E.2 above; Sheppard, 2023, p. A-2). The Administrator preliminarily concludes, for

the reasons discussed above, that an annual standard is a more appropriate form to address deposition-related effects, but he recognizes that greater weight could be given to the effectiveness of the existing 1-hour primary standard in controlling emissions and associated deposition. In light of these considerations, we solicit comment on this alternate option for revising the secondary SO₂ standard to be identical to the current primary SO₂ standard.

In summary, based on all of the considerations identified above, including the currently available evidence in the ISA, the quantitative and policy evaluations in the PA, and the advice from the CASAC, the Administrator proposes to revise the existing secondary SO₂ standard, to an annual average standard, with a 3-year average form and a level within the range from 10 to 15 ppb as requisite to protect the public welfare. The EPA solicits comment on a lower level for a new annual standard down to 5 ppb, as well as whether the existing 3-hour secondary standard should be retained, in addition to establishing a new annual SO₂ standard. Further, the EPA solicits comment on the option of revising the existing secondary SO₂ standard to be equal to the current primary standard in all respects.

The Administrator additionally considers the available information and the PA evaluations and conclusions regarding the PM standard and S deposition-related effects. In so doing, he takes note of the information indicating varying composition of PM_{2.5} at sites across the U.S. (PA, section 2.4.3), with non-S containing compounds typically comprising more than 70% of the total annual PM_{2.5} mass in the East and even more in the West. Further, he considers the PA findings of air quality analyses that indicate appreciable variation in associations, and generally low correlations, between S deposition and PM_{2.5}, as summarized in section II.B above (PA, sections 6.2.2.3 and 6.2.4.2). In addition, he takes note of the discussion above in support of his decision regarding a revised secondary SO₂ standard, including the atmospheric chemistry

information which indicates the dependency of S deposition on airborne SO_x, as evidenced by the parallel trends of SO₂ emissions and S deposition. Based on all of these considerations, the Administrator judges that protection of sensitive ecosystems from S deposition is more effectively achieved through a revised SO₂ standard than a standard for PM, and a revised PM standard is not warranted to provide protection against the effects of S deposition.

Having reached his proposed decisions with regard to S deposition and SO₂ and PM, the Administrator now turns to consideration of the secondary standards for N oxides and PM with regard to the protection afforded from direct ecological effects and from ecological effects related to ecosystem N deposition. As described below, the Administrator proposes to retain the existing NO₂ and PM standards. In considering the secondary standard for oxides of N and protection from direct effects of N oxides in ambient air, the Administrator notes the 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, and that the currently available information continues to document such effects, as summarized in section II.C.1.a above (ISA, Appendix 3, sections 3.3 and 3.4; PA, sections 4.1 and 5.4.2). With regard to NO₂ and NO, the evidence does not indicate effects associated with ambient air concentrations allowed by the existing standard, as summarized in section II.C.3.c above (PA, section 7.4). Accordingly, the Administrator concurs with the PA conclusion that the evidence related to the direct effects of the N oxides, NO₂ and NO, does not call into question the adequacy of protection provided by the existing standard. With regard to the N oxide, HNO₃, the PA provides additional evaluation in recognition of the evidence of effects associated with air concentrations and associated HNO₃ dry deposition on plant and lichen surfaces, and uncertainty as to the extent to which exposures associated with such effects may be allowed by the existing secondary NO₂ standard (PA

sections 7.1.2 and 5.4.2, and Appendix 5B, section 5B.4). Consistent with the conclusion in the PA, the Administrator judges the limited evidence to lack a clear basis for concluding that such effects might have been elicited by air quality that met the secondary NO₂ standard. Thus, while the Administrator takes note of this discussion in the PA, he additionally recognizes the limitations of the evidence and associated uncertainties and judges them too great to provide support to a revised secondary NO₂ standard. In so doing, he additionally takes note of the unanimous view of the CASAC that the existing secondary NO₂ standard provides protection from direct effects of N oxides (section II.E.2 above).

The Administrator next turns to consideration of the larger information base of effects related to N deposition in ecosystems. In so doing, he recognizes 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 (PA, section 7.2.3). These complexities and challenges are described further below. Some of the 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 other complexities related to available quantitative information and analyses differ. Further, with regard to many aquatic systems that receive N loading from sources other than atmospheric deposition, there is complexity to estimating the portion of N inputs, and associated contribution to effects, derived from atmospheric sources.

It is important to note first that, as a general matter, the Administrator finds there are substantially more significant limitations and uncertainties associated with the evidence base for

ecosystem effects related to N deposition associated with N oxides and PM, and with the available air quality information related to the limited potential for control of N deposition in areas across the U.S., in light of the impacts of other pollutants (i.e., NH₃) on N deposition. 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 NH₃, which is not a criteria pollutant, 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.

Additionally, the Administrator recognizes additional complexities in risk management and policy judgments, including with regard to identifying risk management objectives, such as judging the requisite public welfare protection 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), which, as noted in the PA, also complicates conclusions regarding the

extent to which some ecological effects may be judged adverse to the public welfare (PA, section 7.4). In aquatic systems, identification of appropriate public welfare protection objectives is further complicated by N contributions to these systems from multiple sources other than atmospheric deposition, as well as 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 turning to consideration of the evidence and air quality information related to N deposition, the Administrator takes note of the fact that ecosystem N deposition is influenced by air pollutants other than N oxides, particularly, NH₃, which is not a CAA criteria pollutant (PA, sections 6.1, 6.2.1 and 7.2.3.3). As described in the PA, the extent of this contribution varies appreciably across the U.S. and has increased during the past 20 years.

More specifically, while the PA historical trend analyses document the reductions in N deposition that correspond with reductions in emissions of N oxides, these analyses additionally document the increasing role of NH₃ in N deposition since approximately 2010 and the co-occurring tempering of total N deposition reductions, likely reflecting the countervailing pattern in contributions from NH₃. Further, the areas of highest N deposition appear to correspond to the areas with the greatest deposition of NH₃ (PA, Figure 7-8).⁹⁶ The Administrator concurs with the PA conclusion that this information complicates his consideration of the currently available

⁹⁶ This associated lessening influence of N oxides on total N deposition is also evidenced by the poor correlations between N deposition and annual average NO₂ concentrations (PA, sections 6.2.3 and 6.2.4), most particularly in more recent years and at eastern sites, which may be related to increasing emissions of NH₃ in more recent years and at eastern sites (PA, section 2.2.3 and Figure 6-5).

information with regard to protection from N deposition-related effects that might be afforded by the secondary standard for N oxides; particularly when considering the information since 2010 (and in more localized areas prior to that). That is, while the information regarding recent rates of ecoregion N deposition may in some individual areas (particularly those for which reduced N, specifically NH₃, has a larger role) indicate rates greater than the range of values identified in the PA for consideration (e.g., 7-12 kg/ha-yr based on the considerations in section 7.2.3 of the PA and the benchmark of 10 kg/ha-yr, as conveyed in the advice from the CASAC), the PA notes that the extent to which this occurrence relates to the existing NO₂ secondary standard is unclear. The lack of clarity is both because of uncertainties in relating ambient air NO₂ concentrations to rates of deposition, and because of the increasing contribution of NH₃ to N deposition.

The Administrator notes the PA finding that the temporal trend in ecoregion N deposition differs for ecoregions in which N deposition is driven by reduced N compared to those where reduced N comprises less of the total (e.g., PA, 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 across the full 20-year period, as well as lower N deposition in the second as compared to the first decade (corresponding to the decline in NO₂ emissions), appear to document the influence that NO₂ emissions and concentrations had during this period. However, the influence of N oxides appears to be low in areas of the U.S. where N deposition is currently the highest, and where NH₃ emissions have an influential role (PA, section 7.2.3.3). In light of the PA evaluations of N deposition and relative contribution from reduced and oxidized N compounds, the Administrator concurs with the PA conclusion that, based on the current air quality and deposition information and trends, a secondary standard for N oxides cannot be expected to effectively control total N deposition (PA, section 7.4).

The Administrator additionally considers the two sets of advice from the CASAC regarding an NO₂ annual standard in consideration of N deposition effects (section II.E.2 above). The CASAC majority recommended revision of the existing annual NO₂ standard level to a value “<10 to 20 ppb” (Sheppard, 2023, p. 24). As described in section II.E.2 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 NO₂ metric (annual NO₂ EAQM-weighted), which as noted above is not directly translatable to concentrations at individual monitors or to potential standard levels. 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 II.B above, the correlation coefficients are negative for N deposition with both annual NO₂ EAQMs (PA, Table 6-10). Accordingly, based on the lack of a correlation for N deposition with the EAQMs, as well as the lack of translatability of the EAQM-weighted values to monitor concentrations or standard levels, the PA did not find the information highlighted by the CASAC majority for relating N deposition levels to ambient air concentrations to provide scientific support for their recommended levels. In light of this, the Administrator does not agree with the CASAC majority recommendations on revisions to the annual NO₂ standard.

The minority CASAC member recommended revision of the secondary NO₂ standard to be identical to the primary standard based on their conclusion that the recent N deposition levels meet desired objectives and that the primary standard is currently the controlling standard (Sheppard, 2023, Appendix A). In consideration of this advice, the PA noted that among the NO₂ primary and secondary NAAQS, the 1-hour primary standard (established in 2010) may currently be the controlling standard for ambient air concentrations, and observed that annual

average NO₂ concentrations, averaged over three years, in areas that meet the current 1-hour primary standard, have generally been below approximately 35 to 40 ppb.⁹⁷

In light of this finding, the PA identified a revision option for consideration based on the recent pattern in NO₂ concentrations (i.e., revision to a level below the current level of 53 ppb to as low as 35 to 40 ppb [PA, section 7.4]).

In considering this option as identified in the PA, the Administrator takes note of the PA characterization of the support for this option as “not strong” (PA, section 7.4). He further notes the PA conclusion that while the option may have potential to provide some level of protection from N deposition related to N oxides, there is significant uncertainty as to the level of protection that would be provided. The Administrator notes that this uncertainty relates prominently to the influence of NH₃ on total N deposition separate from that of N oxides, and which in some areas of the U.S., particularly those areas where N deposition is highest, appears to be dominant (PA, section 7.2.3.3). Further, he gives weight to the PA statement that the extent to which the relative roles of these two pollutants (N oxides and NH₃) may change in the future is not known. As evaluated in the PA, 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.

In light of the considerations recognized above (summarized earlier in this section and in section II.E.1 above), the Administrator finds that the existing evidence does not clearly call into

⁹⁷ The air quality information regarding annual average NO₂ concentrations at SLAMS monitors indicates more recent NO₂ concentrations are well below the existing standard level of 53 ppb. As noted in the PA, 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 or below 11 kg N/ha-yr, annual average NO₂ concentrations, averaged across three years, have been at or below 35 ppb (PA, section 7.2.3.3).

question the adequacy of the existing secondary NO₂ standard. In so doing, he additionally notes, as recognized in the PA above, that recent median N deposition estimates are below the N deposition benchmark identified by the CASAC majority of 10 kg/ha-yr in ecoregions for which approximately half or more of recent total N deposition is estimated to be oxidized N, driven by N oxides (PA, section 7.2.3.3). In addition to the substantial uncertainty described above regarding the need for control of N deposition from N oxides that might be provided by a secondary standard for N oxides, the Administrator also takes note of the PA finding that there is substantial uncertainty about the effect of a secondary standard for N-oxides on the control of N deposition such that it is also not clear whether the available information provides a sufficient basis for a revised standard that might be judged to provide the requisite protection. In light of this PA finding, the current information on air quality and N deposition, and all of the above considerations, the Administrator proposes to also judge that the available evidence in this review is sufficient to conclude a revision to the secondary annual NO₂ standard is not warranted. Based on all of these considerations, he proposes to retain the existing secondary NO₂ standard, without revision. The EPA solicits comments on this proposed decision, and also solicits comment on the alternative of revising the level and form of the existing secondary NO₂ standard to a level within the range from 35 to 40 ppb with a 3-year average form.

Lastly, the Administrator turns to consideration of the existing standards for PM. As an initial matter, he takes note of the PA discussion and conclusion that the available information does not 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 (PA, sections 7.1.3 and 7.4). The evidence characterized in the ISA and summarized in the PA indicates such effects to be associated with conditions associated with concentrations much higher than the

existing standards. Thus, the Administrator proposes to conclude that the current evidence does not call into question the adequacy of the existing PM standards with regard to direct effects and deposition of pollutants other than S and N compounds.

With regard to N deposition and PM_{2.5}, the Administrator considers the analyses and evaluations in the PA, as well as advice from the CASAC. As an initial matter, the Administrator takes note of the substantial and significant limitations and uncertainties associated with the evidence base for ecosystem effects related to N deposition associated with PM and with the available air quality information related to the limited potential for control of N deposition in areas across the U.S. in light of the impacts of NH₃ on N deposition, as summarized earlier. For example, he notes limitations and uncertainties that relate to relatively lower correlations in more recent time periods of air quality metrics with N deposition and the variation in PM composition across the U.S. For example, the air quality analyses of relationships found only 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 somewhat higher correlations for the early years of the 20-year period, but with low or no correlation in the later years (PA, Chapter 6 and section 7.2.3.3). The PA also noted 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, and that an appreciable percentage of PM_{2.5} mass does not contribute to N deposition. For example, the highest percentage of PM_{2.5} represented by N compounds at CSN sites in 2020-2022 is 30% and it is less than 10% at an appreciable of sites (PA, section 6.4.2). The PA notes that 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 considering the advice from the CASAC for revision of the existing annual PM_{2.5} secondary standard, the Administrator notes the lack of consensus such that the Committee provided two different recommendations for revising the level of the standard, as summarized in section II.E.2 above: one for a level in the range from 6 to 10 µg/m³ and the second for a level of 12 µg/m³. As summarized in the PA, the specific rationale for the range from 6 to 10 µg/m³ is unclear, with levels within this range described as both relating to N deposition in a preferred range (at or below 10 kg N/ha-yr) and relating to deposition above that range.⁹⁸ The PA noted that this “overlap” illustrates the weakness and variability of relationships of PM_{2.5} with N deposition across the U.S. (PA, section 7.4). Further, the PA notes the low correlation for total N deposition estimates with annual average PM_{2.5} design values in the last 10 years at SLAMS (PA, Table 6-7). The second recommendation, from the CASAC minority, was based on their conclusion that the recent N (and S) deposition levels meet desired targets and that the primary annual PM_{2.5} standard, which has been 12 µg/m³ since 2013, has been the controlling standard for annual PM_{2.5} concentrations (Sheppard, 2023, Appendix A).

Based on the currently available information, taking into account its limitations and associated uncertainties, and in consideration of all of the above, the Administrator proposes to conclude that PM_{2.5} is not an appropriate indicator for a secondary standard intended to provide protection of the public welfare from adverse effects related to N deposition. In reaching this proposed conclusion, the Administrator focuses in particular on the weak correlation between

⁹⁸ 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 µg/m³) refers both to annual average PM_{2.5} concentrations (3-yr averages) ranging from 2 to 8 µg/m³ 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 µg/m³ (at design value sites in areas of N deposition estimates greater than 15 kg/ha-yr), as summarized in section II.E.2 above.

annual average PM_{2.5} design values and N deposition estimates in recent time periods, and additionally notes the PA conclusion that the available evidence, as evaluated in the PA, is reasonably judged insufficient to provide a basis for revising the PM_{2.5} annual standard with regard to effects of N deposition related to PM. Thus, based on consideration of the PA analyses and conclusions, as well as consideration of advice from the CASAC, the Administrator proposes to conclude that no change to the annual secondary PM_{2.5} standard is warranted and he proposes to retain the existing PM_{2.5} secondary standard, without revision. The EPA solicits comment on this proposed decision. Additionally, while recognizing the information and judgments regarding uncertainties that lead him to this proposed decision, the Administrator recognizes that there may be alternate views with regard to whether and to what extent a secondary standard with a PM_{2.5} indicator might be expected to provide control of N deposition. In this context, the Administrator additionally solicits comment on revising the existing standard level to a level of 12 µg/m³, in light of the recommendation and associated rationale provided by the CASAC minority.

With regard to other PM standards, the Administrator concurs with the PA's finding of a lack of information that would call into question the adequacy of protection afforded by the existing PM₁₀ secondary standard for ecological effects, and thus concludes it is appropriate to propose retaining this standard without revision. With regard to the 24-hour PM_{2.5} standard, the Administrator takes note of the PA conclusion that the evidence available in this review, as documented in the ISA, or cited by the CASAC,⁹⁹ does not call into question the adequacy of

⁹⁹ As summarized in section II.E.2 above, the CASAC majority, in its recommendation for revision of the existing standard, did not provide specificity regarding the basis for its references to lichen species and fog or cloud water, and the available evidence as characterized in the ISA does not provide estimates of this deposition or describe associated temporal variability, or specifically describe related effects on biota (ISA, Appendix 2).

protection provided by the 24-hour PM_{2.5} standard from ecological effects (PA, section 7.4). Further, the Administrator considers the comments of the CASAC majority and recommendations for revision of this standard to a lower level or to an indicator of deciviews based on its consideration of short-term fog or cloud-related deposition events, without further specificity, as summarized in section II.E.2 above. In so doing, the Administrator notes the PA finding that, while the available evidence in the ISA recognizes there to be N deposition associated with cloud water or fog, it does not provide estimates of this deposition, describe associated temporal variability, or specify related effects on biota (ISA, Appendix 2; PA, section 7.3). Further, the specific revision options recommended by the CASAC majority were based on visibility considerations although the adequacy of protection provided by the secondary PM_{2.5} standard from visibility effects has been addressed in the reconsideration of the 2020 PM NAAQS decision (89 FR 16202, March 6, 2024) and is not included in this review. The Administrator additionally notes the recommendation from the CASAC minority to retain the existing 24-hour secondary PM_{2.5} standard without revision. Based on all of these considerations, the Administrator proposes to retain the existing 24-hour secondary PM_{2.5} standard, without revision. Further, based on the lack of evidence calling into question the adequacy of the secondary PM₁₀ standards, he also proposes to retain the secondary PM₁₀ standards without revision.

In reaching the conclusions described above regarding protection of the public welfare from ecological effects associated with ecosystem deposition of N and S compounds, the Administrator also takes note of consideration in the PA of the potential for indicators different from those for the current standards that may target specific chemicals that deposit N and S, e.g., NO₃⁻, SO₄²⁻, NH₄⁺ (PA, sections 7.2.2.3, 7.2.3.3 and 7.4). In so doing, however, he recognizes a

number of uncertainties and gaps in the available information important to such consideration. For example, one uncertainty 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 in protected areas. As described in the PA, 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. Based on these considerations, the Administrator judges that the currently available information does not support standards based on such indicators at this time. Additionally, there are not currently approved regulatory monitoring methods for these potential indicators and there are practical considerations associated with establishing new standards with new indicators related to establishment of regulatory measurement methods and surveillance networks, that would impact effective implementation of the standards. Thus, he also recognizes that additional data collection and analysis is needed to develop the required evidence base to inform more comprehensive consideration of such alternatives.

F. Proposed Decision on the Secondary Standards

The Administrator proposes to revise the existing secondary SO₂ standard to be an annual average, averaged over three consecutive years with a level within the range from 10 to 15 ppb. The EPA solicits comments on this proposal, including the averaging time, form and range of levels for the revised standard. The EPA also solicits comments on the option of retaining the existing 3-hour standard, while also establishing such a new annual secondary standard. Additionally, the EPA solicits comment on the second option identified in the PA, for setting the level for a new annual standard in the range from 10 to 5 ppb, and also on the option of revising

the secondary standard to be identical to the existing primary standard in all respects, along with the rationales on which such views are based.

The Administrator proposes to retain the existing secondary standards for N oxides, and the existing suite of secondary standards for PM. The EPA solicits comments on these proposed decisions. The EPA also solicits comment on revising the level and form of the existing secondary NO₂ standard to a level within the range from 40 to 35 ppb and a 3-year average form. Regarding the PM secondary standards, the Administrator also solicits comment on revising the level of the existing annual secondary PM_{2.5} standard to 12 µg/m³.

III. Interpretation of the Secondary SO₂ NAAQS

The EPA is proposing to revise Appendix T to 40 CFR Part 50, Interpretation of the Primary National Ambient Air Quality Standards for Oxides of Sulfur, in order to provide data handling procedures for the proposed annual secondary SO₂ standard. The proposed § 50.21 which sets the averaging period, level, indicator, and form of the proposed annual standard refers to this Appendix T. The proposed revised Appendix T would detail the computations necessary for determining when the proposed annual secondary SO₂ NAAQS is met. The proposed revised Appendix T also would address data reporting, data completeness considerations, and rounding conventions.

A. Background

The general purpose of a data interpretation appendix is to provide the practical details on how to make a comparison between multi-day and possibly multi-monitor ambient air concentration data and the level of the NAAQS, so that determinations of attainment and nonattainment are as objective as possible. Data interpretation guidelines also provide criteria for determining whether there are sufficient data to make a NAAQS level comparison at all. The

regulatory language for the current secondary SO₂ NAAQS, originally adopted in 1971, does not contain detailed data interpretation instructions. This situation contrasts with the primary NO₂, ozone, PM_{2.5}, PM₁₀, lead, and primary SO₂ NAAQS regulations, for which there are detailed data interpretation appendices in 40 CFR part 50 addressing issues that can arise in comparing monitoring data to the NAAQS. The existing Appendix T includes these detailed data interpretation requirements for the primary 1-hour SO₂ NAAQS, thus the proposed revision would provide similar information for the proposed annual secondary SO₂ NAAQS. The EPA has used its experience developing and applying this data interpretation appendix to develop the proposed revisions to the text in Appendix T to address the proposed annual SO₂ standard.

B. Interpretation of the Secondary SO₂ Standard

The purpose of a data interpretation rule for the secondary SO₂ NAAQS is to give effect to the form, level, averaging time, and indicator specified in the proposed regulatory text at 40 CFR 50.21, anticipating and resolving in advance various future situations that could occur. The proposed revised Appendix T provides definitions and requirements that apply to the proposed annual secondary standard for SO₂. The requirements clarify how ambient air data are to be reported, what ambient air data are to be used for comparisons with the SO₂ NAAQS, and how to calculate design values for comparisons with the SO₂ NAAQS. The data already required to be reported by ambient SO₂ monitors for use in calculating design values for the current primary 1-hour SO₂ NAAQS are also sufficient for use in calculating design values for the proposed secondary SO₂ NAAQS.

The proposed revised Appendix T specifies that the annual secondary SO₂ NAAQS would be met at an ambient air quality monitoring site when the valid annual secondary standard design value is less than or equal to [10-15] ppb, depending on the level finalized. The annual

secondary standard design value for an ambient air quality monitoring site is described as the mean of the annual means for three consecutive years, with the annual mean derived as the annual average of daily means, with rounding and data completeness specified as described below. The use of a daily mean value in deriving the design value is consistent with the existing data handling requirements for the current 1-hour primary SO₂ NAAQS.

Data completeness requirements for the proposed annual secondary standard in the proposed revised Appendix T follows past EPA practice for other NAAQS pollutants by requiring that in general at least 75% of the monitoring data that should have resulted from following the planned monitoring schedule in a period must be available for the key air quality statistic from that period to be considered valid. These data completeness requirements are consistent with the current data completeness requirements for the 1-hour primary SO₂ NAAQS in Appendix T, and this proposal would not change those requirements. For the proposed annual secondary SO₂ NAAQS, the key air quality statistics are the annual average of daily mean (24-hour average, midnight-to-midnight) concentrations in three successive years. It is important that daily means are representative of the 24-hour period and that all seasons of the year are well represented. Hence, the 75% requirement is proposed to be applied at the daily and quarterly levels. These completeness requirements, including the calculation of the daily mean, are consistent with existing completeness requirements for the current 1-hour primary SO₂ NAAQS. The EPA invites comment on the proposed completeness requirements in Appendix T.

Recognizing that there may be years with incomplete data, the proposed text provides that a design value derived from incomplete data will nevertheless be considered valid if at least 75 percent of the days in each quarter of each of three consecutive years have at least one reported hourly value, and the 3-year annual average design value calculated according to the

procedures specified in the proposed revised Appendix T is above the level of the secondary annual standard. Additionally, following provisions in the proposed revised Appendix T, a substitution test may be used to demonstrate validity of incomplete design values above the level of the standard by substituting a “low” daily mean value from the same calendar quarter in the 3-year design value period. Similarly, another substitution test may be used to demonstrate validity of incomplete design values below the level of the standard by substituting a “high” daily mean value from the same calendar quarter in the 3-year design value period. These substitution tests are consistent with existing substitution tests for the current 1-hour primary SO₂ NAAQS.

It should be noted that one possible outcome of applying the proposed substitution test is that a year with incomplete data may nevertheless be determined to not have a valid design value and thus to be unusable in making annual secondary NAAQS compliance determinations for that 3-year period. However, the intention of the proposed substitution test is to reduce the frequency of such occurrences. The EPA invites comment on incorporating the proposed substitution test into the final rule.

The EPA is proposing that the Administrator have general discretion to use incomplete data to calculate design values that would be treated as valid for comparison to the NAAQS despite the incompleteness, either at the request of a State or at the Administrator’s own initiative. Similar provisions exist already for the PM_{2.5}, NO₂, lead, and 1-hour primary SO₂ NAAQS. The Administrator would consider monitoring site closures/moves, monitoring diligence, and nearby concentrations in determining whether to use such data.

Regarding rounding conventions for the annual secondary SO₂ NAAQS, the EPA is proposing to be consistent with rounding conventions used for the current 1-hour primary SO₂ NAAQS. Specifically, the EPA proposes that hourly SO₂ measurement data shall be reported to

EPA's regulatory database in units of ppb, to at most one place after the decimal, with additional digits to the right being truncated with no further rounding. Daily mean values and the annual mean of those daily values are not rounded. Further, the annual secondary standard design value is calculated pursuant to the proposed revised Appendix T and then rounded to the nearest whole number or 1 ppb (decimals 0.5 and greater are rounded up to the nearest whole number, and any decimal lower than 0.5 is rounded down to the nearest whole number). The EPA invites comment on the proposed rounding conventions.

IV. Ambient Air Monitoring Network for SO₂

One of the primary purposes of collecting ambient air SO₂ monitoring data is for use in determining whether ambient pollutant concentrations exceed the SO₂ NAAQS. Ambient air SO₂ monitoring data are collected by State, local, and Tribal monitoring agencies, as well as industry and the EPA in some instances, in accordance with the monitoring requirements contained in 40 CFR parts 50, 53, and 58. This section briefly discusses the current status of the SO₂ monitoring network, its adequacy in supporting the existing secondary SO₂ standard, and its support for the proposed revision to the secondary SO₂ NAAQS. Based on a thorough review of the SO₂ monitoring network, the EPA is not proposing any changes to the ambient air monitoring network design requirements.

Ambient air SO₂ monitoring data used for comparison to the NAAQS are typically collected by State, local, and Tribal monitoring agencies ("monitoring agencies"), as well as industry and Federal entities in some situations, in accordance with the monitoring requirements contained in 40 CFR parts 50, 53, and 58. A monitoring network is generally designed to measure and provide relevant air quality data as described and prescribed in 40 CFR part 58. To ensure that the data from the network are accurate and reliable to fulfill their intended purpose,

the monitors in the network must comply with a set of requirements including the use of monitoring methods that EPA has designated as Federal Reference Methods (FRMs) or Federal Equivalent Methods (FEMs) per 40 CFR Part 53, a network design to achieve monitoring objectives, and specific siting criteria, data reporting, quality assurance, and data handling rules or procedures.

When promulgating the existing short-term 1-hour daily maximum primary SO₂ NAAQS in 2010 (75 FR 35520, June 22, 2010), the EPA recognized that monitoring to support the health-based standard required a focus on measuring where SO₂ emissions were elevated and to address population exposure. To that end, the EPA finalized minimum monitoring requirements for ambient air SO₂ that prioritized monitoring resources in areas based on coincidence of elevated SO₂ emissions and population, locking in a significant portion of the existing network at that time as well as introducing new monitors to the network. This approach was based on a hybrid analytical approach that was explained in the preamble to the 2010 primary SO₂ NAAQS review that used both monitoring and modeling to assess compliance with the newly promulgated 1-hour standard.

In 2015, the EPA followed up on that hybrid monitoring and modeling concept to support the new 1-hour primary NAAQS by promulgating the Data Requirements Rule (DRR). The DRR (80 FR 51051, August 21, 2015) required air quality characterization in areas with large sources of SO₂ emissions, specifically taking measures to assess and address the lack of information on SO₂ concentrations around sources or source areas emitting 2,000 tons per year or more. Under the DRR, States had the option to employ monitoring, dispersion modeling, or to take a federally enforceable permit limit to comply. The implementation of the DRR resulted in approximately 78 monitoring sites across the country being focused on collecting data at locations of expected

maximum SO₂ concentrations around sources.

There are approximately 434 ambient air SO₂ monitors currently reporting data to EPA nationwide, with at least one SO₂ monitor in every State, the District of Columbia, and Puerto Rico. The network reflects minimum monitoring requirements promulgated in the 2010 SO₂ Primary NAAQS revision, the requirement to measure SO₂ at all NCore multipollutant monitoring stations, actions taken by monitoring agencies to satisfy the DRR, plus additional monitoring conducted by State, local, and Tribal air agencies on their own prerogative to satisfy other data needs. In the memo in the docket for this action titled “Ambient Air SO₂ Monitoring Network Review and Background” (Watkins et al., 2024), it is indicated that the current SO₂ monitoring network includes a focus on characterizing air quality where SO₂ concentrations are expected to be high in the ambient air. The network provides data needed for implementation of the current primary and secondary SO₂ NAAQS and can also provide data that can be used to support the needs for an annual average standard like the one being proposed in this action.

Based on the EPA’s review of the SO₂ network history, current design and objectives, and data, we believe that the current network is adequate to provide the data needed to implement the proposed secondary SO₂ NAAQS; therefore, modification to the existing SO₂ minimum monitoring requirements is not necessary. As noted in section II.B.1, spatial distribution of SO₂ and sulfate deposition reflect the distribution for SO_x emissions on which the network is largely focused upon. Additionally, as noted in section II.E.3, there is a general association of monitoring sites having higher SO₂ concentrations in areas with higher local sulfur deposition estimates, meaning that a network measuring SO₂ in areas of expected higher concentrations would be expected to capture SO₂ concentrations contributing to areas experiencing higher deposition. We therefore believe that modifications to the existing SO₂

minimum monitoring requirements are not necessary to support implementation of the standard proposed in this action. In further support of this position, the EPA notes that the network is and will continue to be adaptable and can evolve in response to changing data needs, even without the Agency making changes to minimum monitoring requirements. The State, local, and Tribal air agencies that operate most of the network monitors, as well as industry stakeholders, can propose and make adjustments to their pieces of the network when a new need arises, or air quality conditions change. Finally, the EPA has authority through 40 CFR part 58, Appendix D, section 4.4.3, for its Regional Administrators to work with State, local, and Tribal air agencies to require SO₂ monitoring above the minimum monitoring requirements where the network is found to be insufficient to meet its objectives. This means that monitors can be added in an area that has the potential for concentrations that exceed or contribute to an exceedance of the level of the NAAQS.

In summary, the EPA is not proposing any changes to the minimum monitoring requirements as part of this proposal to revise the secondary SO₂ NAAQS because the network is currently adequate, and because the EPA, State, local, Tribal, and industry stakeholders have the authority and ability to adjust monitoring efforts and redirect resources as needed to ensure that the monitoring objectives of the SO₂ network continue to be met. The EPA solicits comment on this proposed determination.

V. Clean Air Act Implementation Considerations for the Proposed Secondary SO₂ Standard

The proposed SO₂ secondary NAAQS, if finalized, would trigger a number of implementation processes which are discussed for informational purposes in this portion of the preamble. The Agency is proposing to retain the secondary NO₂ and PM NAAQS; thus,

discussion of implementation considerations related to those NAAQS is not included in this section.

At the outset, promulgation of a new or revised NAAQS, including finalization of this proposed revision, would trigger a process through which States¹⁰⁰ would make recommendations to the Administrator regarding initial area designations. States also would be required to make a new SIP submission to establish that they meet the necessary structural requirements for such new or revised NAAQS pursuant to CAA section 110(a)(1) and (2), also referred to as the “infrastructure SIP submission” (more on this submission below). Further, if finalized, the SO₂ secondary NAAQS would need to be incorporated into the implementation of applicable air permitting requirements and SIP conformity processes. This section provides background information for understanding the possible implications of the proposed NAAQS changes and describes the EPA’s intentions for providing States any guidance the EPA determines to be needed to assist their implementation efforts, if such proposed changes are finalized. This section also describes existing EPA interpretations of CAA requirements and other EPA guidance relevant to implementation of a new SO₂ secondary NAAQS, if one is finalized.

The EPA is not proposing any regulatory changes to SO₂ implementation as a part of this proposal. Therefore, EPA is not proposing action on such topics in this rulemaking. The public is encouraged to share information regarding implementation needs and considerations. Although this rulemaking is not requesting comment specifically on this topic, information on this topic may be submitted for informational purposes to the docket for this proposed rulemaking. The

¹⁰⁰ This and all subsequent references to “state” are meant to include State, local and Tribal agencies responsible for the implementation of a SO₂ control program.

EPA welcomes the public to provide input to the Agency through comments. However, because these issues are not relevant to the establishment of the proposed secondary NAAQS, and because no specific revisions are proposed for the regulations implementing the proposed secondary NAAQS the EPA does not expect to respond to these comments in the final action on this proposal (nor is it required to do so).

A. Designation of Areas

After the EPA establishes a new or revised NAAQS (primary or secondary), the CAA requires the EPA and States to take steps to ensure that the new or revised NAAQS is met. The first step, known as the initial area designations, involves identifying areas of the country that either meet or do not meet the new or revised NAAQS, along with the nearby areas contributing to NAAQS violations.

Section 107(d)(1) of the CAA states that, “By such date as the Administrator may reasonably require, but not later than 1 year after promulgation of a new or revised national ambient air quality standard for any pollutant under section 109, the Governor of each State shall ... submit to the Administrator a list of all areas (or portions thereof) in the State” and make recommendations for whether the EPA should designate those areas as “nonattainment,” “attainment,” or “unclassifiable.”¹⁰¹ Section 107(d)(1)(A) of the CAA contains definitions of these terms. A nonattainment area is any area that does not meet (or that contributes to ambient air quality in a nearby area that does not meet) a NAAQS; an attainment area is any area (other than an area identified as a nonattainment area) that meets a NAAQS; and an unclassifiable area is any area that cannot be classified on the basis of available information as meeting or not

¹⁰¹ While the CAA says “designating” with respect to the Governor’s letter, in the full context of the CAA section it is clear that the Governor makes a recommendation to which the EPA must respond via a specified process if the EPA does not accept it.

meeting a NAAQS. The CAA provides the EPA discretion to require states to submit their designations recommendations within a reasonable amount of time not exceeding 1 year. The CAA also stipulates that “the Administrator may not require the Governor to submit the required list sooner than 120 days after promulgating a new or revised national ambient air quality standard.” Section 107(d)(1)(B)(i) further provides, “Upon promulgation or revision of a NAAQS, the Administrator shall promulgate the designations of all areas (or portions thereof) ... as expeditiously as practicable, but in no case later than 2 years from the date of promulgation. Such period may be extended for up to one year in the event the Administrator has insufficient information to promulgate the designations.” With respect to the NAAQS setting process, courts have interpreted the term “promulgation” to be signature and widespread dissemination of a final rule.¹⁰²

If the EPA agrees that the state’s designations recommendations are consistent with all relevant CAA requirements, then it may proceed to promulgate the designations for such areas. However, if the EPA disagrees that a state’s recommendation is consistent with all relevant CAA requirements, then the EPA may make modifications to the recommended designations by following the process outlined in the CAA. By no later than 120 days prior to promulgating the final designations, the EPA is required to notify states of any intended modifications to the designations of any areas or portions thereof, including the boundaries of areas, as the EPA may deem necessary. States then have an opportunity to comment on the EPA's intended designations decisions. If a state elects not to provide designations recommendations, then the EPA must timely promulgate the designations that it deems appropriate. While section 107(d) of the CAA specifically addresses the designations process for states, the EPA intends to follow the same

¹⁰² *API v. Costle*, 609 F.2d 20 (D.C. Cir. 1979).

process for Tribes to the extent practicable, pursuant to section 301(d) of the CAA regarding Tribal authority, and the Tribal Authority Rule (63 FR 7254, February 12, 1998). To provide clarity and consistency in doing so, the EPA issued a guidance memorandum to our Regional Offices on working with Tribes during the designations process.¹⁰³

In this action, the EPA is proposing to add an annual average SO₂ secondary standard with a level within the range of 10-15 ppb, averaged over three consecutive years. Consistent with the process used in previous area designations efforts for both primary and secondary standards, the EPA will employ a nationally consistent framework and approach to evaluate each state's designations recommendations, considering air quality and other area-specific facts and circumstances¹⁰⁴ to support area designations and boundaries decisions for the NAAQS. Section 107(d) of the CAA explicitly requires that the EPA designate as nonattainment not only the area that is violating the pertinent standard, but also those nearby areas that contribute to ambient air quality in the violating area. Consistent with past practice, the EPA plans to address issues relevant to area designations more fully in a separate designations-specific memorandum around the time of promulgation of a new SO₂ secondary NAAQS, if the proposal to establish a new standard is finalized. As this section is designed for informational purposes, the public may comment on the process and schedule for the initial area designations and nonattainment

¹⁰³ "Guidance to Regions for Working with Tribes during the National Ambient Air Quality Standards (NAAQS) Designations Process," December 20, 2011, Memorandum from Stephen D. Page to Regional Air Directors, Regions 1-X available at https://www.epa.gov/sites/default/files/2017-02/documents/12-20-11_guidance_to_regions_for_working_with_tribes_naaqs_designations.pdf.

¹⁰⁴ The EPA has historically used nationally consistent area-specific analyses to support nonattainment area boundary recommendations and final boundary determinations by evaluating factors such as air quality data, emissions and emissions-related data (e.g., population density and degree of urbanization, traffic and commuting patterns), meteorology, geography/topography, and jurisdictional boundaries. We expect to follow a similar process when establishing area designations for any new or revised SO₂ secondary NAAQS, if finalized.

boundary setting effort associated with the proposed new SO₂ secondary NAAQS. However, the EPA does not expect to respond to these comments in the final action containing the final decision on the proposed NAAQS (nor is it required to do so).

As in past iterations of establishing a new or revised NAAQS, the EPA intends to make the designations for any new or revised NAAQS based on the most recent 3 years of complete, certified, and valid air quality data and other available information. The EPA intends to use such available air quality data from the current SO₂ monitoring network and other technical information. Monitoring data are currently available from existing FEM and FRM monitors sited and operated in accordance with 40 CFR parts 50 and 58 to determine compliance with the proposed new SO₂ secondary NAAQS, if finalized.

In some areas, State or Tribal air agencies may flag air quality data for certain days in the Air Quality System (AQS) database due to potential impacts from exceptional events (e.g., volcanic activity for SO₂). Air quality monitoring data affected by exceptional events may be excluded from use in identifying a violation at a regulatory ambient air monitoring site if the data meet the criteria for exclusion under EPA's "Treatment of Data Influenced by Exceptional Events" Final Rule (81 FR 68216; October 3, 2016) (Exceptional Events Rule) and codified at 40 CFR 50.1, 50.14, and 51.930. For events affecting initial area designations, including designations under the proposed annual SO₂ secondary NAAQS, if finalized, the air agency is required to follow the exceptional events demonstration submission deadlines that are identified in Table 2 to 40 CFR §50.14(c)(2)(vi)—"Schedule for Initial Notification and Demonstration Submission for Data Influenced by Exceptional Events for Use in Initial Area Designations." The EPA encourages air agencies to work collaboratively with the appropriate EPA Regional office after identifying any exceptional event influencing ambient air quality concentrations in a

way that could affect area designations for the proposed annual SO₂ secondary NAAQS, if finalized.¹⁰⁵

B. Section 110(a)(1) and (2) Infrastructure SIP Requirements

The CAA directs states to address basic SIP requirements to implement, maintain, and enforce the NAAQS. Under CAA sections 110(a)(1) and (2), states are required to have state implementation plans that provide the necessary air quality management infrastructure including, among other things, enforceable emissions limitations, an ambient air monitoring program, an enforcement program, air quality modeling capabilities, and adequate personnel, resources, and legal authority. After the EPA promulgates a new or revised NAAQS, states are required to make a new SIP submission to establish that they meet the necessary structural requirements for such new or revised NAAQS or make changes to do so. The EPA refers to this type of SIP submission as an “infrastructure SIP submission.” Under CAA sections 110(a)(1), all states are required to make these infrastructure SIP submissions within 3 years after promulgation of a new or revised standard, or such shorter deadline as the EPA may impose. Although the CAA authorizes the EPA to set a shorter time for states to make these SIP submissions, the EPA is not proposing to do so. Section 110(b) of the CAA also provides that the EPA may extend the deadline for the “infrastructure” SIP submission for a new secondary NAAQS by up to 18 months beyond the initial 3 years. If the proposed new annual SO₂ secondary NAAQS is finalized, the EPA believes it would be more efficient for states and the EPA if each affected state submits the section 110 infrastructure SIP that addresses the secondary standard within 3 years of promulgation of a new or revised NAAQS, and so is not proposing to apply a shorter deadline. However, the EPA also

¹⁰⁵ Additional information, tools, and resources relating to exceptional events can be found at the EPA’s exceptional events website located at <https://www.epa.gov/air-quality-analysis/final-2016-exceptional-events-rule-supporting-guidance-documents-updated-faqs>

recognizes that individual states may prefer the flexibility to submit the secondary NAAQS infrastructure SIP at a later date, and if requested, the EPA would review such requests on a case-by-case basis as is provided by the EPA's existing regulations implementing CAA section 110(b) at 40 CFR 51.341.

Under CAA section 110(a)(1) and (2), states are required to make SIP submissions that address a number of requirements pertaining to implementation, maintenance, and enforcement of a new or revised NAAQS. The specific subsections in CAA section 110(a)(2) require states to address a number of requirements, as applicable: (A) emissions limits and other control measures; (B) ambient air quality monitoring/data system; (C) programs for enforcement of control measures and for construction or modification of stationary sources; (D)(i) interstate pollution transport and (ii) interstate and international pollution abatement; (E) adequate resources and authority, conflict of interest, and oversight of local governments and regional agencies; (F) stationary source monitoring and reporting; (G) emergency episodes; (H) SIP revisions; (I) plan revisions for nonattainment areas; (J) consultation with government officials, public notification, Prevention of Significant Deterioration (PSD) and visibility protection; (K) air quality modeling and submission of modeling data; (L) permitting fees; and (M) consultation and participation by affected local entities. These requirements apply to all SIP submissions in general, but the EPA has provided specific guidance to states concerning its interpretation of these requirements in the specific context of infrastructure SIP submissions for a new or revised NAAQS.¹⁰⁶

The EPA interprets the CAA such that two elements identified in section 110(a)(2) are

¹⁰⁶ See "Guidance on Infrastructure State Implementation Plan (SIP) Elements under Clean Air Act sections 110(a)(1) and 110(a)(2)" September 2013, Memorandum from Stephen D. Page to Regional Air Directors, Regions 1-10.

not subject to the 3-year submission deadline of section 110(a)(1), and thus states are not required to address them in the context of an infrastructure SIP submission. The elements pertain to part D, in title I of the CAA, which addresses plan requirements for nonattainment areas. Therefore, for the reasons explained below, the following section 110(a)(2) elements are considered by the EPA to be outside the scope of infrastructure SIP actions: (1) the portion of section 110(a)(2)(C), programs for enforcement of control measures and for construction or modification of stationary sources that applies to permit programs applicable in designated nonattainment areas, (known as “nonattainment new source review”) under part D; and (2) section 110(a)(2)(I) in its entirety. The EPA does not expect states to address these two elements pertaining to part D for a new or revised NAAQS in the infrastructure SIP submissions to include regulations or emissions limits developed specifically for attaining the relevant standard as it pertains to areas designated nonattainment for the proposed SO₂ secondary NAAQS, if finalized. States would be required to submit infrastructure SIP submissions for the proposed new SO₂ secondary NAAQS, if finalized, before they are required to submit nonattainment plan SIP submissions to demonstrate attainment with the same NAAQS. As a general matter, states would be required to submit nonattainment plans to provide for attainment and maintenance of the proposed new SO₂ secondary NAAQS, if finalized, within 3 years from the effective date of nonattainment area designations as required under CAA section 172(b). In addition, because this NAAQS, if finalized, would be a secondary standard, section 110(b) of the CAA also provides that the EPA may extend the deadline for the nonattainment plan for up to 18 months beyond the initial 3 years. The EPA reviews and acts upon these later SIP submissions through a separate process. For this reason, the EPA does not expect states to address new nonattainment area emissions controls per section 110(a)(2)(I) in their infrastructure SIP submissions.

Another required infrastructure SIP element is that each state's SIP must contain adequate provisions to prohibit, consistent with the provisions of title I of the CAA, emissions from within the state that will significantly contribute to nonattainment in, or interfere with maintenance by, any other state of the primary or secondary NAAQS.¹⁰⁷ This element is often referred to as the "good neighbor" or "interstate transport" provision.¹⁰⁸ The provision has two prongs: significant contribution to nonattainment (prong 1) and interference with maintenance (prong 2). The EPA and states must give independent significance to prong 1 and prong 2 when evaluating downwind air quality problems under CAA section 110(a)(2)(D)(i)(I).¹⁰⁹ Further, case law has established that the EPA and states must implement requirements to meet interstate transport obligations in alignment with the applicable statutory attainment schedule of the downwind areas impacted by upwind-state emissions.¹¹⁰

The EPA anticipates coordinating with states with respect to the requirements of CAA section 110(a)(2)(D)(i)(I) for implementation of the proposed SO₂ secondary NAAQS, if finalized. We note that states may elect to make SIP submissions that address certain infrastructure SIP elements separately from the others. In recent years, due in part to the complexity of addressing interstate transport obligations, some states have found it efficient to make SIP submissions to address the interstate transport provisions separately from other infrastructure SIP elements.

It is the responsibility of each state to review its air quality management program's

¹⁰⁷ CAA section 110(a)(2)(D)(i)(I)

¹⁰⁸ CAA section 110(a)(2)(D)(i)(II) also addresses certain interstate effects that states must address and thus is also sometimes referred to as relating to "interstate transport."

¹⁰⁹ See *North Carolina v. EPA*, 531 F.3d 896, 909-11 (D.C. Cir. 2008).

¹¹⁰ See *id.* at 911-13. See also *Wisconsin v. EPA*, 938 F.3d 303, 313-20 (D.C. Cir. 2019); *Maryland v. EPA*, 958 F.3d 1185, 1203-04 (D.C. Cir. 2020).

existing SIP provisions in light of each new or revised NAAQS to determine whether any revisions are necessary to implement a new or revised NAAQS. Most states have revised and updated their SIPs in recent years to address requirements associated with other revised NAAQS. For some states, it may be the case that, for a number of infrastructure elements, the state may believe it already has adequate state regulations adopted and approved into the SIP to address a particular requirement with respect to any new or revised NAAQS. For such portions of the state's infrastructure SIP submission, the state could provide an explanation of how its existing SIP provisions are adequate.

If a state determines that existing SIP-approved provisions are adequate in light of the proposed SO₂ secondary NAAQS, if finalized, with respect to a given infrastructure SIP element (or sub-element), then the state could make a SIP submission “certifying” that the existing SIP contains provisions that address those requirements of the specific section 110(a)(2) infrastructure elements.¹¹¹ In the case of such a certification submission, the state would not have to include a copy of the relevant provision (e.g., rule or statute) itself. Rather, the state in its infrastructure SIP submission could provide citations to the SIP-approved state statutes, regulations, or non-regulatory measures, as appropriate, which meet the relevant CAA requirement. Like any other SIP submission, that state could make such a certification only after it has provided reasonable notice and opportunity for public hearing. This “reasonable notice and opportunity for public hearing” requirement for infrastructure SIP submissions is to meet the requirements of CAA sections 110(a) and 110(l). Under the EPA's regulations at 40 CFR part 51, if a public hearing is held, an infrastructure SIP submittal must include a certification by the

¹¹¹ A “certification” approach would not be appropriate for the interstate pollution control requirements of CAA section 110(a)(2)(D)(i).

state that the public hearing was held in accordance with the EPA's procedural requirements for public hearings. See 40 CFR part 51, Appendix V, paragraph 2.1(g), and see 40 CFR 51.102.

In consultation with its EPA Regional Office, a state should follow all applicable EPA regulations governing infrastructure SIP submissions in 40 CFR part 51 - e.g., subpart I (Review of New Sources and Modifications), subpart J (Ambient Air Quality Surveillance), subpart K (Source Surveillance), subpart L (Legal Authority), subpart M (Intergovernmental Consultation), subpart O (Miscellaneous Plan Content Requirements), subpart P (Protection of Visibility), and subpart Q (Reports). For the EPA's general criteria for infrastructure SIP submissions, refer to 40 CFR part 51, Appendix V, Criteria for Determining the Completeness of Plan Submissions. The EPA recommends that states electronically submit their infrastructure SIPs to the EPA through the State Plan Electronic Collaboration System (SPeCS),¹¹² an online system available through the EPA's Central Data Exchange.

C. Prevention of Significant Deterioration and Nonattainment New Source Review Programs for the Proposed Secondary SO₂ Standard

The CAA, at parts C and D of title I, contains preconstruction review and permitting programs applicable to new major stationary sources and major modifications of existing major sources. The preconstruction review of each new major stationary source and major modification applies on a pollutant-specific basis, and the requirements that apply for each pollutant depend on whether the area in which the source is situated is designated as attainment (or unclassifiable) or nonattainment for that pollutant. In areas designated attainment or unclassifiable for a pollutant, the PSD requirements under part C apply to construction at major sources. In areas designated nonattainment for a pollutant, the Nonattainment New Source Review (NNSR)

¹¹² <https://cdx.epa.gov/>

requirements under part D apply to major source construction. Collectively, those two sets of permit requirements are commonly referred to as the “major New Source Review” or “major NSR” programs.

The EPA is not proposing any changes to the NSR program regulations as part of this proposal to implement the proposed SO₂ secondary NAAQS, if finalized. Under the PSD program, at the effective date of a final new or revised NAAQS, the applicant must demonstrate that any new construction or major modification and associated source emissions increase triggering PSD requirements does not cause or contribute to violation of that new or revised NAAQS. The EPA has models, guidance, and other tools for making this showing. The EPA anticipates that sources and reviewing authorities will be able to use most of these existing tools to demonstrate compliance with the secondary SO₂ standard, if finalized as proposed. However, some adjustment and updates to these tools may be appropriate. The EPA is also considering an alternative compliance demonstration approach (described in section V.D. of this action) that the Agency may support using to make this PSD permitting demonstration. Considering these topics, the EPA has developed a separate technical document (Tillerson et al., 2024),¹¹³ which provides a technical justification for how a demonstration of compliance with the 1-hour primary SO₂ standard can suffice to demonstrate compliance with the proposed new SO₂ secondary standard, if such a standard is finalized.

The statutory requirements for a PSD permit program set forth under part C of title I of the CAA (sections 160 through 169) are implemented through the EPA’s PSD regulations found at 40 CFR 51.166 (minimum requirements for an approvable PSD SIP) and 40 CFR 52.21 (PSD

¹¹³ This technical memo (Tillerson et al., 2024) is available in the docket for this NAAQS review (EPA-HQ-OAR-2014-0128).

permitting program for permits issued under the EPA's federal permitting authority). Whenever a proposed new major source or major modification triggers PSD requirements for SO₂, either 40 CFR 52.21 or state regulations based on 40 CFR 51.166 will apply for undesignated areas and for areas that are designated as attainment or as unclassifiable for the proposed SO₂ secondary NAAQS, if such standard is finalized.

For PSD, a "major stationary source" is one with the potential to emit 250 tons per year (tpy) or more of any regulated NSR pollutant, unless the new or modified source is classified under a list of 28 source categories contained in the statutory definition of "major emitting facility" in section 169(1) of the CAA. For those 28 source categories, a "major stationary source" is one with the potential to emit 100 tpy or more of any regulated NSR pollutant. A "major modification" is a physical change or a change in the method of operation of an existing major stationary source that results, first, in a significant emissions increase of a regulated NSR pollutant and, second, in a significant net emissions increase of that pollutant. *See* 40 CFR 51.166(b)(2)(i), 40 CFR 52.21(b)(2)(i). The EPA PSD regulations define the term "regulated NSR pollutant" to include any pollutant for which a NAAQS has been promulgated and any pollutant identified in the EPA regulations as a constituent or precursor to such pollutant. *See* 40 CFR 51.166(b)(49), 40 CFR 52.21(b)(50). Thus, the PSD program currently requires the review and control of emissions of SO₂, as applicable. Among other things, for each regulated NSR pollutant emitted or increased in a significant amount, the PSD program requires a new major stationary source or a major modification to apply the "best available control technology" (BACT) and to conduct an air quality impact analysis to demonstrate that the proposed major stationary source or major modification will not cause or contribute to a violation of any

NAAQS or PSD increment.¹¹⁴ *See* CAA section 165(a)(3)–(4), 40 CFR 51.166(j)–(k), 40 CFR 52.21(j)–(k). The PSD requirements may also include, in appropriate cases, an analysis of potential adverse impacts on Class I areas. *See* CAA section 162(a) and 165, 40 CFR 51.166(p); 40 CFR 52.21(p)).¹¹⁵

With regard to nonattainment NSR, the EPA’s regulations for the NNSR programs are contained in 40 CFR 51.165, 52.24 and 40 CFR part 51, Appendix S. Specifically, the EPA developed minimum program requirements for a NNSR program that is approvable in a SIP, and those requirements, which include requirements for SO₂, are contained in 40 CFR 51.165. In addition, 40 CFR part 51, Appendix S contains requirements constituting an interim NNSR program. This program enables NNSR permitting in nonattainment areas by states that lack a SIP-approved NNSR permitting program (or a program that does not apply to the relevant pollutant) during the time between the date of the relevant designation and the date that the EPA approves into the SIP a NNSR program. *See* 40 CFR part 51, Appendix S, Part I; 40 CFR 52.24(k). Any new NNSR requirements for SO₂ associated with the proposed secondary standard, should it be finalized, would not become applicable until the effective date of any

¹¹⁴ By establishing the maximum allowable level of ambient air pollutant concentration increase in a particular area, an increment defines “significant deterioration” of air quality in that area. Increments are defined by the CAA as maximum allowable increases in ambient air concentrations above a baseline concentration and are specified in the PSD regulations by pollutant and area classification (Class I, II and III). 40 CFR 51.166(c), 40 CFR 52.21(c); 75 FR 64864; October 20, 2010; The EPA has developed the Guideline on Air Quality Models and other documents to, among other things, provide methods and guidance for demonstrating compliance the NAAQS and PSD increments including the annual SO₂ standard. *See* 40 CFR part 51, Appendix W; 82 FR 5182, January 17, 2017.

¹¹⁵ Congress established certain Class I areas in section 162(a) of the CAA, including international parks, national wilderness areas, and national parks that meet certain criteria. Such Class I areas, known as mandatory Federal Class I areas, are afforded special protection under the CAA. In addition, states and Tribal governments may establish Class I areas within their own political jurisdictions to provide similar special air quality protection.

nonattainment designation for the final standard.

As noted above, the EPA intends to review and consider the appropriateness of existing PSD compliance demonstration tools for implementation of any new secondary SO₂ NAAQS. In addition, as described below and in section V.D. of this document, the EPA acknowledges that there may be added burdens associated with making the required PSD air quality impact demonstration for the annual secondary standard if finalized, and the EPA may develop an alternative compliance demonstration based upon Tillerson et al. (2024) if the secondary SO₂ NAAQS is finalized as proposed. Under such an alternative compliance demonstration, sources and reviewing authorities would be able to sufficiently demonstrate compliance with the proposed new SO₂ secondary standard by demonstrating compliance with the primary 1-hour standard. Section V.D. of this document includes further discussion of an alternative compliance demonstration approach and the technical justification that sources and permitting authorities may apply in permitting actions.

D. Alternative PSD Compliance Demonstration Approach for the Proposed Secondary SO₂ Standard

If the new secondary SO₂ NAAQS is finalized as proposed, the EPA would plan to calculate design values for the new secondary NAAQS using the procedures described in section III of this preamble, relying upon ambient air SO₂ measurement data. The PSD program requires that new or modified stationary sources complete a compliance demonstration using air quality modeling or other methods to demonstrate that their proposed emissions increases will not cause or contribute to a violation of any NAAQS, including this secondary SO₂ NAAQS, if finalized. See 40 CFR 51.166(k), (m) and 40 CFR 52.21(k), (m). Under 40 CFR 51.166(l) and 40 CFR 52.21(l), all PSD demonstrations for purposes of determining whether a new or modified source will cause or contribute to a NAAQS violation, including a violation of the secondary NAAQS

for SO₂, if finalized, must be based upon air quality models, databases, and other requirements specified 40 CFR part 51, Appendix W.

Under section 9.2.3 of Appendix W, the EPA recommends a multi-stage approach to making the required demonstration of compliance with the NAAQS, which enables a streamlined demonstration in many cases using PSD screening tools. If a cumulative impact assessment is necessary, due to the source-oriented nature of the current monitoring network for SO₂, there is some uncertainty as to whether sources may be able to rely on existing monitoring data to adequately represent background for their PSD compliance demonstrations. Although the current SO₂ monitoring network is primarily geared to measure in areas of high SO₂ emissions proximate to populations and to sources, it has a limited number of monitors away from emissions areas that are needed to provide the information necessary for area specific estimates of background concentrations. Therefore, there may be situations where prospective PSD sources could be required to collect new data in order to determine the representative background concentrations of annual SO₂.

Because of the added burdens that may result for applicants and permitting authorities from these considerations, the EPA is considering alternative approaches to enable prospective PSD sources to more readily demonstrate that they will not cause or contribute to a violation of the proposed secondary SO₂ NAAQS, if finalized as proposed. The EPA believes that it is reasonable to allow the use of an alternative demonstration approach where such an approach is technically justified. The EPA is providing notice of the potential for an alternative PSD compliance demonstration approach discussed in this section and based upon the technical analysis detailed in Tillerson et al. (2024) included in the docket. The public is encouraged to share information on this alternative compliance demonstration approach. The EPA may

consider information provided by the public in developing any future guidance on this approach for the new secondary SO₂ NAAQS. The EPA is not proposing this alternative compliance demonstration approach for the proposed secondary standard nor is the EPA taking any action to implement this alternative compliance demonstration approach in this rulemaking. Consequently, the EPA would not be obligated to respond to any comments received on this topic as part of the final rulemaking.

The Agency believes that following an alternative compliance demonstration approach could aid implementation of the PSD permitting program after enactment of the proposed secondary SO₂ NAAQS, if finalized. To support consideration of alternative approaches that could be used by prospective PSD sources, the EPA conducted a two-pronged technical analysis of the relationships between the proposed secondary standard and the existing 1-hour SO₂ primary NAAQS (*See* Tillerson et al., 2024). The first prong of the analysis addressed aspects of a PSD source impact analysis by evaluating whether an individual source's impact resulting in a small increase in 1-hour SO₂ concentration, at the level of the significant impact level (SIL) for the primary SO₂ NAAQS, would produce a comparably small increase in the annual SO₂ concentration. This analysis included modeled estimates of SO₂ for a range of source categories and scenarios. It indicated that small increases in 1-hour SO₂ concentrations caused by individual sources produce similarly small changes in the annual SO₂ concentrations. The second prong of the analysis addressed aspects of a PSD cumulative impact analysis indicating that a demonstration showing attainment of the 1-hour SO₂ standard is expected to also show attainment of the proposed secondary SO₂ standard. This analysis was based on 2017 to 2022 air quality data and compared the air quality that would meet the current 1-hour SO₂ standard (with its level of 75 ppb in conjunction with a 99th percentile averaged over 3 years) with air quality

that would meet the proposed secondary SO₂ standard (with a level of 10-15 ppb in conjunction with an annual mean averaged over three years). As shown in Tillerson et al. (2024), this analysis indicated that all areas for which existing monitoring data showed attainment of the 1-hour SO₂ standards would also likely be in attainment of the proposed secondary SO₂ standard. The EPA believes that this technical analysis is robust and that its conclusions can be applied across the United States.

Based on this technical analysis, the EPA currently believes that there is sufficient evidence that, for the purposes of making a demonstration under the PSD program that a new or modified source will not cause or contribute to a violation of the proposed secondary SO₂ NAAQS, if finalized, a persuasive demonstration that the source will not cause or contribute to a violation of the 1-hour SO₂ NAAQS could serve as a suitable alternate compliance demonstration. As such, many or all sources undergoing PSD review for SO₂ would be able to rely upon their analysis demonstrating that they will not cause or contribute to a violation of the 1-hour SO₂ NAAQS to also demonstrate that they will not cause or contribute to a violation of the proposed secondary SO₂ NAAQS, if finalized. This alternative compliance demonstration approach would thus serve to streamline air quality analyses in a manner consistent with the CAA and NSR regulations. Using this approach would result in a source not needing to provide a separate and distinct analysis to demonstrate compliance with the proposed secondary SO₂ standard, if finalized. The EPA believes this alternative compliance demonstration approach could fulfill PSD requirements for individual sources in PSD areas for the proposed secondary SO₂ NAAQS, if finalized. This approach would apply in both areas that would not yet have been designated as nonattainment for the new secondary SO₂ NAAQS, if finalized, and those that would be ultimately designated as attainment or unclassifiable areas. The EPA will continue to

evaluate this potential approach and may consider it in guidance addressing implementation of the proposed secondary SO₂ NAAQS, if finalized, separate from this rulemaking setting the standard itself.

E. Transportation Conformity Program

Transportation conformity is required under CAA section 176(c) (42 U.S.C. 7506(c)) to ensure that federally supported highway and transit activities are consistent with (“conform to”) the purpose of the SIP. Transportation conformity applies to areas that are designated as nonattainment areas and to nonattainment areas that have been redesignated to attainment with an approved CAA section 175A maintenance plan (i.e., maintenance areas) for transportation-related criteria pollutants: carbon monoxide, ozone, NO₂, PM_{2.5}, and PM₁₀. Motor vehicles are not significant sources of SO₂, and thus transportation conformity does not apply to any SO₂ NAAQS (40 CFR 93.102(b)(1)), existing or proposed.¹¹⁶ Therefore, the EPA is not proposing any changes to the transportation conformity rule (40 CFR 51.390 and 40 CFR part 93, subpart A) for the proposed SO₂ secondary NAAQS.

F. General Conformity Program

The General Conformity program requirements at 40 CFR part 93 subpart B establish criteria and procedures for determining conformity as required under CAA section 176(c),¹¹⁷

¹¹⁶ See “VII. Description of the Proposal” in “Criteria and Procedures for Determining Conformity to State or Federal Implementation Plans of Transportation Plans, Programs, and Projects funded or Approved Under Title 23 U.S.C. or the Federal Transit Act.” (58 FR 3768, January 11, 1993). EPA finalized the original transportation conformity regulations on November 24, 1993 (58 FR 62188). The rule has subsequently been revised and the current provisions of the transportation conformity rule are found at 40 CFR part 93, subpart A.

¹¹⁷ Under CAA section 176(c)(1), Federal agencies have the affirmative responsibility to assure their actions achieve conformity to the purpose of an implementation plan, where the term “conformity to an implementation plan” is defined at CAA sections 176(c)(1)(A) and 176(c)(1)(B). Under CAA section 176(c)(4) the EPA is required to establish criteria and procedures for determining conformity.

which prohibits a Federal agency from taking an action that would interfere with the ability of a state or Tribe to attain or maintain the NAAQS. General Conformity applies only to Federal activities not defined as transportation plans, programs, or projects under 40 CFR 93.102. The program requirements apply to emissions of all six criteria pollutants and their precursors per 40 CFR 93.153(b)(1) and (b)(2), but only to the extent the emissions can be characterized as “direct emissions” or “indirect emissions” as defined under 40 CFR 93.152.

The General Conformity program applies to activities that cause emissions of the criteria or precursor pollutants to originate within designated nonattainment areas¹¹⁸ or redesignated attainment areas that operate under approved CAA section 175A maintenance plans (*i.e.*, maintenance areas).

The EPA is not proposing changes to the General Conformity regulations in this proposed rulemaking. However, in the future, the EPA intends to review the need to issue or revise guidance describing how the current conformity regulations apply in nonattainment and maintenance areas for any new or revised NAAQS, as needed.

VI. Statutory and Executive Order Reviews

Additional information about these statutes and Executive Orders can be found at <https://www.epa.gov/laws-regulations/laws-and-executive-orders>.

A. Executive Order 12866: Regulatory Planning and Review and Executive Order 14094: Modernizing Regulatory Review

This action is a “significant regulatory action” as defined in Executive Order 12866, as amended by Executive Order 14094. Accordingly, EPA submitted this action to the Office of

¹¹⁸ Applicability of the General Conformity program to any newly designated nonattainment area for a specific NAAQS begins one year following the effective date of the final nonattainment designation, as allowed under CAA section 176(c)(6) and 40 CFR 93.153(k).

Management and Budget (OMB) for Executive Order 12866 review. Documentation of any changes made in response to the Executive Order 12866 review is available in the docket. The EPA prepared an analysis to determine if additional emission reductions would be needed to meet the proposed revision of the secondary SO₂ NAAQS. This analysis is contained in the document “Air Quality Analyses Using Sulfur Dioxide (SO₂) Air Quality Data,” which is available in the docket for this NAAQS review (ID No. EPA-HQ-OAR-2014-0128). The analysis concluded that no additional emissions reductions beyond any needed to meet the current 1-hour primary SO₂ NAAQS would be expected to be necessary to meet the proposed annual secondary SO₂ NAAQS, resulting in no costs or benefits associated with pollution controls for this proposed NAAQS revision, if finalized. Accordingly, no regulatory impact analysis has been prepared for this proposal.

B. Paperwork Reduction Act (PRA)

This action does not impose an information collection burden under the PRA. There are no information collection requirements directly associated with a proposed decision to revise or retain a NAAQS under section 109 of the CAA.

C. Regulatory Flexibility Act (RFA)

I certify that this action will not have a significant economic impact on a substantial number of small entities under the RFA. This action will not impose any requirements on small entities. Rather, this proposed rule establishes national standards for allowable annual average concentrations of SO₂ in ambient air as required by section 109 of the CAA. See also *American Trucking Associations v. EPA*, 175 F.3d 1027, 1044-45 (D.C. Cir. 1999) (NAAQS do not have significant impacts upon small entities because NAAQS themselves impose no regulations upon small entities), rev'd in part on other grounds, *Whitman v. American Trucking Associations*, 531

U.S. 457 (2001).

D. Unfunded Mandates Reform Act (UMRA)

This action does not contain any unfunded mandate as described in the Unfunded Mandates Reform Act (UMRA), 2 U.S.C. 1531—1538, and does not significantly or uniquely affect small governments. Furthermore, as indicated previously, in setting a NAAQS the EPA cannot consider the economic or technological feasibility of attaining ambient air quality standards, although such factors may be considered to a degree in the development of state plans to implement the standards. See also *American Trucking Associations v. EPA*, 175 F. 3d at 1043 (noting that because the EPA is precluded from considering costs of implementation in establishing NAAQS, preparation of the RIA pursuant to the Unfunded Mandates Reform Act would not furnish any information that the court could consider in reviewing the NAAQS).

The EPA acknowledges, however, that if corresponding revisions to associated SIP requirements and air quality surveillance requirements are proposed at a later time, those revisions might result in such effects. Any such effects would be addressed as appropriate if and when such revisions are proposed.

E. Executive Order 13132: Federalism

This action does not have federalism implications. It will not have substantial direct effects on the states, on the relationship between the national government and the states, or on the distribution of power and responsibilities among the various levels of government. However, the EPA recognizes that states will have a substantial interest in this action and any future revisions to associated requirements.

F. Executive Order 13175: Consultation and Coordination with Indian Tribal Governments

This action does not have Tribal implications, as specified in Executive Order 13175. It

does not have a substantial direct effect on one or more Indian Tribes as Tribes are not obligated to adopt or implement any NAAQS. Thus, Executive Order 13175 does not apply to this action. However, consistent with the *EPA Policy on Consultation and Coordination with Indian Tribes*, the EPA will offer government-to-government consultation with Tribes as requested.

G. Executive Order 13045: Protection of Children from Environmental Health Risks and Safety Risks

EPA interprets Executive Order 13045 as applying only to those regulatory actions that concern environmental health or safety risks that EPA has reason to believe may disproportionately affect children, per the definition of “covered regulatory action” in section 2-202 of the Executive Order.

Therefore, this action is not subject to Executive Order 13045 because it does not concern an environmental health risk or safety risk. Since this action does not concern human health, EPA’s Policy on Children’s Health also does not apply.

H. Executive Order 13211: Actions Concerning Regulations that Significantly Affect Energy Supply, Distribution or Use

This action is not a “significant energy action” because it is not likely to have a significant adverse effect on the supply, distribution, or use of energy. The purpose of this action is to propose to revise the existing secondary SO₂ standard, and also to propose to retain the current secondary standards for NO₂, PM_{2.5} and PM₁₀. The action does not prescribe specific pollution control strategies by which these ambient air standards and monitoring revisions will be met. Such strategies will be developed by states on a case-by-case basis, and the EPA cannot predict whether the control options selected by states will include regulations on energy suppliers, distributors, or users. Thus, the EPA concludes that this proposal does not constitute a significant energy action as defined in Executive Order 13211.

I. National Technology Transfer and Advancement Act (NTTAA)

This action involves technical standards. The EPA is proposing to use the existing indicator, SO₂, for measurements in support of this action. To the extent feasible, the EPA employs a Performance-Based Measurement System (PBMS), which does not require the use of specific, prescribed analytic methods. The PBMS is defined as a set of processes wherein the data quality needs, mandates or limitations of a program or project are specified and serve as criteria for selecting appropriate methods to meet those needs in a cost-effective manner. It is intended to be more flexible and cost effective for the regulated community; it is also intended to encourage innovation in analytical technology and improved data quality. Though the FRM for the NAAQS indicators defines the particular specifications for ambient air monitors, there is some variability with regard to how monitors can measure the pollutants, including SO₂. Therefore, it is not practically possible to fully define the FRM in performance terms to account for this possible or realized variability in measurement principles of operation. Nevertheless, our approach in the past has resulted in multiple brands of monitors being approved as FRM for SO₂, and we expect this to continue. Also, the FRMs described in 40 CFR part 50 and the equivalency criteria described in 40 CFR part 53, constitute a performance-based measurement system for SO₂, since methods that meet the field testing and performance criteria can be approved as FEMs. The EPA is not precluding the use of any other method, whether it constitutes a voluntary consensus standard or not, as long as it meets the specified performance criteria and is approved as an FRM or FEM.

J. Executive Order 12898: Federal Actions to Address Environmental Justice in Minority Populations and Low-Income Populations and Executive Order 14096: Revitalizing our Nation's Commitment to Environmental Justice for All

The EPA believes that the human health and environmental conditions that exist prior to

this action do not result in disproportionate and adverse effects on communities with Environmental Justice (EJ) concerns. As discussed in sections II.D and II.E above, and Chapters 5 and 7 of the PA, the acid buffering capacity of waterbodies in key acid-sensitive ecoregions in recent years is estimated to meet protection targets in high percentages. As discussed in section II.C.2 above, impacts on acid-sensitive waterbodies, if sufficiently severe, would have the potential to impact the public welfare through impacts to fisheries. Although recent conditions do not indicate such a level of severity, to the extent local communities relied on such fisheries disproportionately to their representation in the population, such effects of the past (e.g., effects associated with acidification risks of 20 or more years ago) would have had the potential for disproportionate impacts. Recent conditions do not indicate risk of aquatic acidification to such a level of severity and the available information for recent acid buffering capacity levels does not include evidence of disproportionate and adverse impacts on communities with EJ concerns. As the proposed action is to establish a new, more stringent standard to protect acid-sensitive waterbodies to recent levels and protect against recurrence of acidification effects from the past, for which the potential for disproportionate and adverse effects on local communities is unknown, the EPA believes that this action is not likely to result in new disproportionate and adverse effects on communities with EJ concerns. The information supporting this Executive Order review is contained in the PA for this review and sections II.C, II.D and II.E of this document.

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List of Subjects in 40 CFR Part 50

Environmental protection, Air pollution control, Carbon monoxide, Lead, Nitrogen dioxide, Ozone, Particulate matter, Sulfur oxides.

Dated:

Michael S. Regan,
Administrator.

For the reasons set forth in the preamble, the Environmental Protection Agency proposes to amend chapter I of title 40 of the Code of Federal Regulations as follows:

PART 50—NATIONAL PRIMARY AND SECONDARY AMBIENT AIR QUALITY STANDARDS

1. The authority citation for part 50 continues to read as follows:

Authority: 42 U.S.C. 7401, *et seq.*

2. Add § 50.21 to read as follows:

§ 50.21 National secondary ambient air quality standards for sulfur oxides (sulfur dioxide).

(a) The national secondary ambient annual air quality standard for oxides of sulfur is [10-15] parts per billion (ppb), measured in the ambient air as sulfur dioxide (SO₂) by a reference method based on Appendix A-1 and Appendix A-2 of this part, or by a Federal Equivalent Method (FEM) designated in accordance with part 53 of this chapter.

(b) The secondary annual standard is met when the 3-year average of the annual SO₂ concentration is less than or equal to [10 – 15] ppb, as determined in accordance with Appendix T of this part.

3. Appendix T to Part 50 is revised to read as follows:

Appendix T to Part 50 – Interpretation of the Primary and Secondary National Ambient Air Quality Standards for Oxides of Sulfur (Sulfur Dioxide)

1. General

(a) This appendix explains the data handling conventions and computations necessary for determining when the primary and secondary national ambient air quality standards for Oxides of Sulfur as measured by Sulfur Dioxide (“SO₂ NAAQS”) specified in § 50.17 are met at an

ambient air quality monitoring site. Sulfur dioxide (SO₂) is measured in the ambient air by a Federal reference method (FRM) based on appendix A or A-1 to this part or by a Federal equivalent method (FEM) designated in accordance with part 53 of this chapter. Data handling and computation procedures to be used in making comparisons between reported SO₂ concentrations and the levels of the SO₂ NAAQS are specified in the following sections.

(b) Decisions to exclude, retain, or make adjustments to the data affected by exceptional events, including natural events, are made according to the requirements and process deadlines specified in §§ 50.1, 50.14 and 51.930 of this chapter.

(c) The terms used in this appendix are defined as follows:

Daily maximum 1-hour values for SO₂ refers to the maximum 1-hour SO₂ concentration values measured from midnight to midnight (local standard time) that are used in NAAQS computations.

Daily mean values for SO₂ refers to the 24-hour average of 1-hour SO₂ concentration values measured from midnight to midnight (local standard time) that are used in NAAQS computations.

Design values are the metrics (*i.e.*, statistics) that are compared to the NAAQS levels to determine compliance, calculated as specified in section 5 of this appendix. The design value for the primary 1-hour NAAQS is the 3-year average of annual 99th percentile daily maximum 1-hour values for a monitoring site (referred to as the “1-hour primary standard design value”). The design value for the secondary annual NAAQS is the 3-year average of the annual mean of daily mean values for a monitoring site (referred to as the “annual secondary standard”).

99th percentile daily maximum 1-hour value is the value below which nominally 99 percent of all daily maximum 1-hour concentration values fall, using the ranking and selection method specified in section 5.1 of this appendix.

Annual mean refers to the annual average of all the daily mean values as defined in section 5.2 of this appendix.

Pollutant Occurrence Code (POC) refers to a numerical code (1, 2, 3, etc.) used to distinguish the data from two or more monitors for the same parameter at a single monitoring site.

Quarter refers to a calendar quarter.

Year refers to a calendar year.

2. Requirements for Data Used for Comparisons With the SO₂ NAAQS and Data

Reporting Considerations

(a) All valid FRM/FEM SO₂ hourly data required to be submitted to EPA's Air Quality System (AQS), or otherwise available to EPA, meeting the requirements of part 58 of this chapter including appendices A, C, and E shall be used in design value calculations. Multi-hour average concentration values collected by wet chemistry methods shall not be used.

(b) Data from two or more monitors from the same year at the same site reported to EPA under distinct Pollutant Occurrence Codes shall not be combined in an attempt to meet data completeness requirements. The Administrator will combine annual 99th percentile daily maximum concentration values from different monitors in different years, selected as described here, for the purpose of developing a valid 1-hour primary standard design value. If more than one of the monitors meets the completeness requirement for all four quarters of a year, the steps specified in section 5.1(a) of this appendix shall be applied to the data from the monitor with the

highest average of the four quarterly completeness values to derive a valid annual 99th percentile daily maximum concentration. If no monitor is complete for all four quarters in a year, the steps specified in sections 3.1(c) and 5.1(a) of this appendix shall be applied to the data from the monitor with the highest average of the four quarterly completeness values in an attempt to derive a valid annual 99th percentile daily maximum concentration. Similarly, the Administrator will combine annual means from different monitors in different years, selected as described here, for the purpose of developing a valid annual secondary standard design value. If more than one of the monitors meets the completeness requirement for all four quarters of a year, the steps specified in section 5.2(a) of this appendix shall be applied to the data from the monitor with the highest average of the four quarterly completeness values to derive a valid annual mean. If no monitor is complete for all four quarters in a year, the steps specified in sections 3.2(c) and 5.2(a) of this appendix shall be applied to the data from the monitor with the highest average of the four quarterly completeness values in an attempt to derive a valid annual mean. This paragraph does not prohibit a monitoring agency from making a local designation of one physical monitor as the primary monitor for a Pollutant Occurrence Code and substituting the 1-hour data from a second physical monitor whenever a valid concentration value is not obtained from the primary monitor; if a monitoring agency substitutes data in this manner, each substituted value must be accompanied by an AQS qualifier code indicating that substitution with a value from a second physical monitor has taken place.

(c) Hourly SO₂ measurement data shall be reported to AQS in units of parts per billion (ppb), to at most one place after the decimal, with additional digits to the right being truncated with no further rounding.

3. Comparisons With the NAAQS

3.1 Comparisons With the 1-Hour Primary SO₂ NAAQS

(a) The 1-hour primary SO₂ NAAQS is met at an ambient air quality monitoring site when the valid 1-hour primary standard design value is less than or equal to 75 parts per billion (ppb).

(b) An SO₂ 1-hour primary standard design value is valid if it encompasses three consecutive calendar years of complete data. A year meets data completeness requirements when all four quarters are complete. A quarter is complete when at least 75 percent of the sampling days for each quarter have complete data. A sampling day has complete data if 75 percent of the hourly concentration values, including State-flagged data affected by exceptional events which have been approved for exclusion by the Administrator, are reported.

(c) In the case of one, two, or three years that do not meet the completeness requirements of section 3.1(b) of this appendix and thus would normally not be useable for the calculation of a valid 3-year 1-hour primary standard design value, the 3-year 1-hour primary standard design value shall nevertheless be considered valid if one of the following conditions is true.

(i) At least 75 percent of the days in each quarter of each of three consecutive years have at least one reported hourly value, and the design value calculated according to the procedures specified in section 5.1 is above the level of the primary 1-hour standard.

(ii)

(A) A 1-hour primary standard design value that is equal to or below the level of the NAAQS can be validated if the substitution test in section 3.1(c)(ii)(B) results in a “test design value” that is below the level of the NAAQS. The test substitutes actual “high” reported daily maximum 1-hour values from the same site at about the same time of the year (specifically, in the same calendar quarter) for unknown values that were not successfully measured. *Note* that the test is merely diagnostic in nature, intended to

confirm that there is a very high likelihood that the original design value (the one with less than 75 percent data capture of hours by day and of days by quarter) reflects the true under-NAAQS-level status for that 3-year period; the result of this data substitution test (the “test design value,” as defined in section 3.1(c)(ii)(B)) is not considered the actual design value. For this test, substitution is permitted only if there are at least 200 days across the three matching quarters of the three years under consideration (which is about 75 percent of all possible daily values in those three quarters) for which 75 percent of the hours in the day, including State-flagged data affected by exceptional events which have been approved for exclusion by the Administrator, have reported concentrations.

However, maximum 1-hour values from days with less than 75 percent of the hours reported shall also be considered in identifying the high value to be used for substitution.

(B) The substitution test is as follows: Data substitution will be performed in all quarter periods that have less than 75 percent data capture but at least 50 percent data capture, including State-flagged data affected by exceptional events which have been approved for exclusion by the Administrator; if any quarter has less than 50 percent data capture then this substitution test cannot be used. Identify for each quarter (e.g., January–March) the highest reported daily maximum 1-hour value for that quarter, excluding State-flagged data affected by exceptional events which have been approved for exclusion by the Administrator, looking across those three months of all three years under consideration. All daily maximum 1-hour values from all days in the quarter period shall be considered when identifying this highest value, including days with less than 75 percent data capture. If after substituting the highest reported daily maximum 1-hour value for a quarter for as much of the missing daily data in the matching deficient

quarter(s) as is needed to make them 100 percent complete, the procedure in section 5 yields a recalculated 3-year 1-hour standard “test design value” less than or equal to the level of the standard, then the 1-hour primary standard design value is deemed to have passed the diagnostic test and is valid, and the level of the standard is deemed to have been met in that 3-year period. As noted in section 3.1(c)(i), in such a case, the 3-year design value based on the data actually reported, not the “test design value,” shall be used as the valid design value.

(iii)

(A) A 1-hour primary standard design value that is above the level of the NAAQS can be validated if the substitution test in section 3.1(c)(iii)(B) results in a “test design value” that is above the level of the NAAQS. The test substitutes actual “low” reported daily maximum 1-hour values from the same site at about the same time of the year (specifically, in the same three months of the calendar) for unknown hourly values that were not successfully measured. Note that the test is merely diagnostic in nature, intended to confirm that there is a very high likelihood that the original design value (the one with less than 75 percent data capture of hours by day and of days by quarter) reflects the true above-NAAQS-level status for that 3-year period; the result of this data substitution test (the “test design value,” as defined in section 3.1(c)(iii)(B)) is not considered the actual design value. For this test, substitution is permitted only if there are a minimum number of available daily data points from which to identify the low quarter-specific daily maximum 1-hour values, specifically if there are at least 200 days across the three matching quarters of the three years under consideration (which is about 75 percent of all possible daily values in those three quarters) for which 75 percent of the hours in the day have reported

concentrations. Only days with at least 75 percent of the hours reported shall be considered in identifying the low value to be used for substitution.

(B) The substitution test is as follows: Data substitution will be performed in all quarter periods that have less than 75 percent data capture. Identify for each quarter (e.g., January–March) the lowest reported daily maximum 1-hour value for that quarter, looking across those three months of all three years under consideration. All daily maximum 1-hour values from all days with at least 75 percent capture in the quarter period shall be considered when identifying this lowest value. If after substituting the lowest reported daily maximum 1-hour value for a quarter for as much of the missing daily data in the matching deficient quarter(s) as is needed to make them 75 percent complete, the procedure in section 5.1 yields a recalculated 3-year 1-hour standard “test design value” above the level of the standard, then the 1-hour primary standard design value is deemed to have passed the diagnostic test and is valid, and the level of the standard is deemed to have been exceeded in that 3-year period. As noted in section 3.1(c)(i), in such a case, the 3-year design value based on the data actually reported, not the “test design value”, shall be used as the valid design value.

(d) A 1-hour primary standard design value based on data that do not meet the completeness criteria stated in 3.1(b) and also do not satisfy section 3.1(c), may also be considered valid with the approval of, or at the initiative of, the Administrator, who may consider factors such as monitoring site closures/moves, monitoring diligence, the consistency and levels of the valid concentration measurements that are available, and nearby concentrations in determining whether to use such data.

(e) The procedures for calculating the 1-hour primary standard design values are given in section 5.1 of this appendix.

3.2 Comparisons With the Annual Secondary SO₂ NAAQS

(a) The annual secondary SO₂ NAAQS is met at an ambient air quality monitoring site when the valid annual secondary standard design value is less than or equal to [10-15] parts per billion (ppb).

(b) An SO₂ annual secondary standard design value is valid if it encompasses three consecutive calendar years of complete data. A year meets data completeness requirements when all four quarters are complete. A quarter is complete when at least 75 percent of the sampling days for each quarter have complete data. A sampling day has complete data if 75 percent of the hourly concentration values, including State-flagged data affected by exceptional events which have been approved for exclusion by the Administrator, are reported.

(c) In the case of one, two, or three years that do not meet the completeness requirements of section 3.2(b) of this appendix and thus would normally not be useable for the calculation of a valid 3-year annual secondary standard design value, the 3-year annual secondary standard design value shall nevertheless be considered valid if one of the following conditions is true.

(i) At least 75 percent of the days in each quarter of each of three consecutive years have at least one reported hourly value, and the design value calculated according to the procedures specified in section 5.2 is above the level of the secondary annual standard.

(ii)

(A) An annual secondary standard design value that is equal to or below the level of the NAAQS can be validated if the substitution test in section 3.2(c)(ii)(B) results in a “test design value” that is below the level of the NAAQS. The test substitutes actual “high”

reported daily mean values from the same site at about the same time of the year (specifically, in the same calendar quarter) for unknown or incomplete (less than 75 percent of hours reported) daily mean values. *Note* that the test is merely diagnostic in nature, intended to confirm that there is a very high likelihood that the original design value (the one with less than 75 percent data capture of hours by day and of days by quarter) reflects the true under-NAAQS-level status for that 3-year period; the result of this data substitution test (the “test design value,” as defined in section 3.2(c)(ii)(B)) is not considered the actual design value. For this test, substitution is permitted only if there are at least 200 days across the three matching quarters of the three years under consideration (which is about 75 percent of all possible daily values in those three quarters) for which 75 percent of the hours in the day, including State-flagged data affected by exceptional events which have been approved for exclusion by the Administrator, have reported concentrations. However, daily mean values from days with less than 75 percent of the hours reported shall also be considered in identifying the high daily mean value to be used for substitution.

(B) The substitution test is as follows: Data substitution will be performed in all quarter periods that have less than 75 percent data capture but at least 50 percent data capture, including State-flagged data affected by exceptional events which have been approved for exclusion by the Administrator; if any quarter has less than 50 percent data capture then this substitution test cannot be used. Identify for each quarter (e.g., January–March) the highest reported daily mean value for that quarter, excluding State-flagged data affected by exceptional events which have been approved for exclusion by the Administrator, looking across those three months of all three years under consideration.

All daily mean values from all days in the quarter period shall be considered when identifying this highest value, including days with less than 75 percent data capture. If after substituting the highest daily mean value for a quarter for as much of the missing daily data in the matching deficient quarter(s) as is needed to make them 100 percent complete, the procedure in section 5 yields a recalculated 3-year annual standard “test design value” less than or equal to the level of the standard, then the annual secondary standard design value is deemed to have passed the diagnostic test and is valid, and the level of the standard is deemed to have been met in that 3-year period. As noted in section 3.2(c)(i), in such a case, the 3-year design value based on the data actually reported, not the “test design value,” shall be used as the valid design value.

(iii)

(A) An annual secondary standard design value that is above the level of the NAAQS can be validated if the substitution test in section 3.2(c)(iii)(B) results in a “test design value” that is above the level of the NAAQS. The test substitutes actual “low” reported daily mean values from the same site at about the same time of the year (specifically, in the same three months of the calendar) for unknown or incomplete (less than 75 percent of hours reported) daily mean values. Note that the test is merely diagnostic in nature, intended to confirm that there is a very high likelihood that the original design value (the one with less than 75 percent data capture of hours by day and of days by quarter) reflects the true above-NAAQS-level status for that 3-year period; the result of this data substitution test (the “test design value,” as defined in section 3.2(c)(iii)(B)) is not considered the actual design value. For this test, substitution is permitted only if there are a minimum number of valid daily mean values from which to identify the low quarter-

specific daily mean values, specifically if there are at least 200 days across the three matching quarters of the three years under consideration (which is about 75 percent of all possible daily values in those three quarters) for which 75 percent of the hours in the day have reported concentrations. Only days with at least 75 percent of the hours reported shall be considered in identifying the low daily mean value to be used for substitution.

(B) The substitution test is as follows: Data substitution will be performed in all quarter periods that have less than 75 percent data capture. Identify for each quarter (e.g., January–March) the lowest reported daily mean value for that quarter, looking across those three months of all three years under consideration. All daily mean values from all days with at least 75 percent capture in the quarter period shall be considered when identifying this lowest value. If after substituting the lowest reported daily mean value for a quarter for as much of the missing daily data in the matching deficient quarter(s) as is needed to make them 75 percent complete, the procedure in section 5.2 yields a recalculated 3-year annual standard “test design value” above the level of the standard, then the annual secondary standard design value is deemed to have passed the diagnostic test and is valid, and the level of the standard is deemed to have been exceeded in that 3-year period. As noted in section 3.2(c)(i), in such a case, the 3-year design value based on the data actually reported, not the “test design value,” shall be used as the valid design value.

(d) An annual secondary standard design value based on data that do not meet the completeness criteria stated in 3.2(b) and also do not satisfy section 3.2(c), may also be considered valid with the approval of, or at the initiative of, the Administrator, who may consider factors such as monitoring site closures/moves, monitoring diligence, the consistency and levels of the valid

concentration measurements that are available, and nearby concentrations in determining whether to use such data.

(e) The procedures for calculating the annual secondary standard design values are given in section 5.2 of this appendix.

4. Rounding Conventions

4.1 Rounding Conventions for the 1-Hour Primary SO₂ NAAQS

(a) Hourly SO₂ measurement data shall be reported to AQS in units of parts per billion (ppb), to at most one place after the decimal, with additional digits to the right being truncated with no further rounding.

(b) Daily maximum 1-hour values and, therefore, the annual 99th percentile of those daily values are not rounded.

(c) The 1-hour primary standard design value is calculated pursuant to section 5.1 and then rounded to the nearest whole number or 1 ppb (decimals 0.5 and greater are rounded up to the nearest whole number, and any decimal lower than 0.5 is rounded down to the nearest whole number).

4.2 Rounding Conventions for the Annual Secondary SO₂ NAAQS

(a) Hourly SO₂ measurement data shall be reported to AQS in units of parts per billion (ppb), to at most one place after the decimal, with additional digits to the right being truncated with no further rounding.

(b) Daily mean values and the annual mean of those daily values are not rounded.

(c) The annual secondary standard design value is calculated pursuant to section 5.2 and then rounded to the nearest whole number or 1 ppb (decimals 0.5 and greater are rounded up to the

nearest whole number, and any decimal lower than 0.5 is rounded down to the nearest whole number).

5. Calculation Procedures

5.1 Calculation Procedures for the 1-Hour Primary SO₂ NAAQS

(a) *Procedure for identifying annual 99th percentile values.* When the data for a particular ambient air quality monitoring site and year meet the data completeness requirements in section 3.1(b), or if one of the conditions of section 3.1(c) is met, or if the Administrator exercises the discretionary authority in section 3.1(d), identification of annual 99th percentile value is accomplished as follows.

(i) The annual 99th percentile value for a year is the higher of the two values resulting from the following two procedures.

(A) *Procedure 1.* For the year, determine the number of days with at least 75 percent of the hourly values reported.

(1) For the year, determine the number of days with at least 75 percent of the hourly values reported including State-flagged data affected by exceptional events which have been approved for exclusion by the Administrator.

(2) For the year, from only the days with at least 75 percent of the hourly values reported, select from each day the maximum hourly value excluding State-flagged data affected by exceptional events which have been approved for exclusion by the Administrator.

(3) Sort all these daily maximum hourly values from a particular site and year by descending value. (For example: (x[1], x[2], x[3], * * *, x[n]). In this case, x[1] is the largest number and x[n] is the smallest value.) The 99th percentile is

determined from this sorted series of daily values which is ordered from the highest to the lowest number. Using the left column of Table 1, determine the appropriate range (*i.e.*, row) for the annual number of days with valid data for year y (cn_y). The corresponding “ n ” value in the right column identifies the rank of the annual 99th percentile value in the descending sorted list of daily site values for year y . Thus, $P_{0.99, y} =$ the n th largest value.

(B) *Procedure 2.* For the year, determine the number of days with at least one hourly value reported.

(1) For the year, determine the number of days with at least one hourly value reported including State-flagged data affected by exceptional events which have been approved for exclusion by the Administrator.

(2) For the year, from all the days with at least one hourly value reported, select from each day the maximum hourly value excluding State-flagged data affected by exceptional events which have been approved for exclusion by the Administrator.

(3) Sort all these daily maximum values from a particular site and year by descending value. (For example: ($x[1]$, $x[2]$, $x[3]$, * * *, $x[n]$). In this case, $x[1]$ is the largest number and $x[n]$ is the smallest value.) The 99th percentile is determined from this sorted series of daily values which is ordered from the highest to the lowest number. Using the left column of Table 1, determine the appropriate range (*i.e.*, row) for the annual number of days with valid data for year y (cn_y). The corresponding “ n ” value in the right column identifies the rank

of the annual 99th percentile value in the descending sorted list of daily site values for year y. Thus, $P_{0.99,y}$ = the nth largest value.

(b) The 1-hour primary standard design value for an ambient air quality monitoring site is mean of the three annual 99th percentile values, rounded according to the conventions in section 4.1.

Table 1 of Appendix T of Part 50

Annual number of days with valid data for year “y” ($c_{n,y}$)	$P_{0.99,y}$ is the nth maximum value of the year, where n is the listed number
1–100	1
101–200	2
201–300	3
301–366	4

5.2 Calculation Procedures for the Annual Secondary SO₂ NAAQS

(a) When the data for a site and year meet the data completeness requirements in section 3.2(b) of this appendix, or if the Administrator exercises the discretionary authority in section 3.2(c), the annual mean is simply the arithmetic average of all the daily mean values.

(b) The annual secondary standard design value for an ambient air quality monitoring site is the mean of the annual means for three consecutive years, rounded according to the conventions in section 4.2.