Review of the Secondary Standards for Ecological Effects of Oxides of Nitrogen, Oxides of Sulfur, and Particulate Matter: Risk and Exposure Assessment Planning Document

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Review of the Secondary Standards for Ecological Effects of Oxides of Nitrogen, Oxides of Sulfur, and Particulate Matter: Risk and Exposure Assessment Planning Document

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Office of Air Quality Planning and Standards
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LIST OF ACRONYMS AND ABBREVIATIONS

AAI	Aquatic Acidification Index
Al	Aluminum
AMoN	Ammonia Monitoring Network
ANC	Acid Neutralizing Capacity
AQ	Air quality
ASSETS EI	Assessment of Estuarine Trophic Status Eutrophication Index
Bc/Al	Base cation/Aluminum
BCS	Base Cation Surplus
BCw	Base Cation weathering
C	Carbon
CAA	Clean Air Act
CALFIRE	State of California fire occurrence, prevention and fire-fighting expenditures
CAMx	Comprehensive Air Quality Model with eXtensions
CASAC	Clean Air Scientific Advisory Committee
CASTNet	Clean Air Status and Trends Network
CH_4	Methane
CL	Critical Load
CLAD	Critical Loads of Atmospheric Deposition Science Committee
CMAQ	Community Multiscale Air Quality Model
CONUS	Contiguous United States
CO_2	Carbon dioxide
CSN	Chemical Speciation Networks
CSS	Coastal Sage Scrub
CV	Coefficient of Variation
DOC	Dissolved Organic Carbon
EMAP	Environmental Monitoring and Assessment Program
EPA	Environmental Protection Agency
FAB	First-order Acid Balance
FASOM	Forest and Agriculture Sector Optimization Model
FIA	Forest Inventory and Analysis
FR	Federal Register
FRM	Federal Reference Method
GIS	Geographic Information Systems
GLNC	Georeferenced Lake Nutrient Chemistry database
HNO_3	nitric acid

IMPROVE	Interagency Monitoring of Protected Visual Environments
IRIS	Internet Research Information Series
IRP	Integrated Review Plan
ISA	Integrated Science Assessment
LTM	Long-Term Monitoring
MAGIC	Model of Acidification of Groundwater In Catchments
MeHg	Methylmercury
MCF	Mixed Conifer Forest
$\mu g/m^3$	micrograms per cubic meter
N	Nitrogen
NAAQS	National Ambient Air Quality Standard
NADP	National Atmospheric Deposition Program
NARS	National Aquatic Resource Surveys
NCCA	National Coastal Condition Assessment
NCLD	National Critical Loads Database
NCore	National Core network
NEI	National Emissions Inventory
NH_3	Ammonia
NH_4^+	Ammonium
NH_4NO_3	Ammonium nitrate
NLA	National Lakes Assessment
NOAA	National Oceanic and Atmospheric Administration
NO	Nitric Oxide
NO_2	Nitrogen Dioxide
NO_3^-	Nitrate
NO_x	Oxides of Nitrogen
NOy	Sum of oxidized N in the gas phase and particle nitrate
N_2O	Nitrous Oxide
NPS	National Park Service
NRSA	National River and Stream Assessment
NTN	National Trends Network
NWCA	National Wetland Condition Assessment
OAQPS	Office of Air Quality Planning and Standards
ppb	parts per billion
ppm	parts per million
PA	Policy Assessment

PAN	Peroxyacetyl Nitrate
PM	Particulate Matter
$PM_{2.5}$	particles with a diameter less than or equal to 2.5 µg
PM_{10}	particles with a diameter less than or equal to 10 µg
PRISM	Parameter-elevation Regression on Independent Slopes Model
REA	Risk and Exposure Assessment
S	Sulfur
SAV	Submerged Aquatic Vegetation
SLT	State and Local agencies and Tribes
SMB	Simple Mass Balance
SO_2	Sulfur dioxide
SO_3	Sulfur trioxide
SO_x	Oxides of Sulfur
SO_4^{2-}	Sulfate
SPARROW	SPAtially Referenced Regressions on Watershed
SRB	Sulfur-reducing bacteria
SRP	Sulfur-reducing prokaryotes
SSWC	Steady State Water Chemistry
TDEP	Total Deposition
TIME	Temporally Integrated Monitoring of Ecosystems
USFS	U.S. Forest Service
USGS	U.S. Geological Survey
VOC	Volatile organic compounds
WHO	World Health Organization

EXECUTIVE SUMMARY

The U.S. Environmental Protection Agency (EPA) is conducting a review of the air quality criteria and the associated secondary (welfare-based) national ambient air quality standards (NAAQS) for oxides of nitrogen, oxides of sulfur and particulate matter (PM). This review is focused on the contribution of oxides of nitrogen, oxides of sulfur, and PM to ecological effects and particularly the contribution of these pollutants to such effects through atmospheric deposition. Based on analysis of the information available in this review regarding support for a quantitative risk and exposure assessment (REA) to inform the review, this document outlines a plan, including scope and methods, for conducting a REA, and is intended to facilitate consultation with the Clean Air Scientific Advisory Committee (CASAC), as well as an opportunity for public participation. The information considered includes newly available scientific evidence, new and/or improved data, methods, and tools and other information or data supporting a quantitative REA particularly those assessed in the second draft *Integrated Science Assessment for Oxides of Nitrogen, Oxides of Sulfur and Particulate Matter – Ecological Criteria* (ISA).

The current review of the secondary standards for oxides of nitrogen, oxides of sulfur and PM considers secondary standards for these three pollutants together with regard to protection against adverse ecological effects on public welfare. This review differs from the review completed in 2012 in that the current review includes consideration of the secondary PM standards, in addition to the secondary standards for oxides of nitrogen and sulfur. Given the contribution of nitrogen compounds to PM, including but not limited to those related to oxides of nitrogen, the current review provides for an expanded and more integrated consideration of N deposition and the current related air quality information.

Since the last review, the scientific evidence for all effect categories has been expanded, especially for terrestrial nitrogen enrichment effects. New critical loads data and exposure-response functions for nitrogen enrichment and acidification effects on terrestrial and freshwater ecosystems are now available that together provide a basis for analysis of ecological effects of atmospheric deposition across the nation. This new information contributes to the stronger weight of evidence and expanded causality determinations in the second draft ISA and enables an evaluation of growth and mortality effects, as well as species or community composition changes, that was not available at the time of the last review. In addition, new air quality data suggest that the spatial variability and distribution of atmospheric deposition has changed in recent years. New techniques are also available for combining these measurements and modeling outputs to estimate total deposition with lower uncertainty. Thus, given the new scientific information and data available since the last review, we believe there is support for a new and updated assessment of risk and exposure to inform the current review.

EPA's proposed plans for the risk and exposure assessment include quantitative analyses of terrestrial and aquatic effects using critical loads and exposure-response functions. These analyses will be conducted at a national-scale and within select case study areas. To estimate deposition for this assessment, we intend to develop new estimates of total N and S deposition that will utilize the methodologies developed by the National Atmospheric Deposition Program (NADP) Total Deposition (TDEP) Science Committee and combine the most recently available measured ambient air concentration and wet deposition data with modeled deposition velocity and dry deposition data. To assess ecological risk under current conditions at a national scale, we intend to compare these TDEP estimates to national critical loads and exposure-response datasets for aquatic and terrestrial acidification and N enrichment. This will inform our understanding of sensitivity and risk for acidification and nitrogen enrichment on a national scale for multiple species.

In addition, to assess ecological risk when just meeting the current standards and any potential alternative standards, as appropriate, we propose to evaluate the impacts of changing ambient concentrations and atmospheric deposition on aquatic and terrestrial acidification and N enrichment in selected study area locations. To do this, we intend to evaluate the relationship between emissions, ambient concentrations and atmospheric deposition of N and S based on data from air quality models and ambient measurements and to use this information to adjust air quality in the study area locations to reflect just meeting the appropriate air quality scenarios. These atmospheric deposition estimates would then be compared to critical loads and exposure-response datasets for aquatic and terrestrial acidification and N enrichment that are most applicable for the individual study area locations. Together, the national-scale and study area assessment information would be used to inform decisions regarding potential adverse effects to public welfare.

1 INTRODUCTION

The U.S. Environmental Protection Agency (EPA) is conducting a review of the air quality criteria and the associated secondary (welfare-based) national ambient air quality standards (NAAQS) for oxides of nitrogen, oxides of sulfur, and particulate matter (PM). The purpose of this planning document (titled *Review of the Secondary National Ambient Air Quality Standards for Ecological Effects of Oxides of Nitrogen, Oxides of Sulfur and Particulate Matter: Risk and Exposure Assessment Planning Document* – hereafter referred to as the REA Planning Document) is to describe EPA's consideration of the extent to which newly available scientific evidence, tools or methodologies, and/or information warrant the conduct of a quantitative risk and exposure assessment (REA) that might inform this review, and may expand our characterization of exposure and risk estimates provided by the assessments conducted for the last review. Based on these considerations, and as described below, we plan to develop a new REA to inform the current review of the secondary NAAQS for oxides of nitrogen, oxides of sulfur, and PM. Accordingly, this document's additional purpose is to describe the general plan, including scope and methods for conducting the REA.

This review of the secondary (welfare-based) NAAQS for oxides of nitrogen, [1] oxides of sulfur[2] and PM,[3] is focused on the contribution of these pollutants to ecological effects and particularly the contribution of these pollutants to such effects through atmospheric deposition.[4] In so doing, we recognize that oxides of nitrogen, oxides of sulfur, and PM contribute to

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[1] In this document, the term oxides of nitrogen refers to all forms of oxidized nitrogen (N) compounds, including nitric oxide (NO), nitrogen dioxide (NO_2), and all other oxidized N-containing compounds formed from NO and NO_2. This follows usages in the Clean Air Act Section 108(c): "Such criteria [for oxides of nitrogen] shall include a discussion of nitric and nitrous acids, nitrites, nitrates, nitrosamines, and other carcinogenic and potentially carcinogenic derivatives of oxides of nitrogen." By contrast, within air pollution research and control communities, the terms "oxides of nitrogen" and "nitrogen oxides" are restricted to refer only to the sum of NO and NO_2, and this sum is commonly abbreviated as NO_x. The category label used by this community for the sum of all forms of oxidized nitrogen compounds including those listed in Section 108(c) is total oxidized nitrogen (NO_y).

[2] Oxides of sulfur are defined here to include sulfur monoxide (SO), sulfur dioxide (SO_2), sulfur trioxide (SO_3), disulfur monoxide (S_2O), and sulfate (SO_4^{2-}), however, SO, SO_3, and S_2O are present at much lower ambient levels than SO_2 and SO_4^{2-} .

[3] PM is the generic term for a broad class of chemically and physically diverse substances that exist as discrete liquid and/or solid particles over a wide range of sizes. Particles may be emitted directly from anthropogenic and natural sources, or formed in the atmosphere by transformations of gaseous emissions such as SO_2, nitrogen oxides (NO_x), ammonia (NH_3) and volatile organic compounds (VOC). The chemical and physical properties of PM vary greatly with time, region, meteorology, and source category.

[4] In addition, these air pollutants contribute to effects on vegetation, soils, and biota both through direct exposure to the pollutant in air, and indirect exposure after deposition.

ecological effects together, through deposition of N and/or S to the surface of vegetation, soils or water bodies. In recognition of these linkages, addressing the pollutants together to assess ecological effects enables a comprehensive look at the nature and interactions of the pollutants.

The current secondary NAAQS for oxides of nitrogen and oxides of sulfur were established to protect against direct damage to vegetation by exposure to gas-phase oxides of nitrogen and oxides of sulfur. The secondary standard for oxides of nitrogen is an annual average not to exceed 0.053 parts per million (ppm) NO_2. The secondary standard for oxides of sulfur is a 3-hour average of 0.5 ppm SO_2, not to be exceeded more than once per year. The secondary PM standards were established to provide protection against a variety of PM-associated welfare effects, including effects on vegetation as well as visibility impairment and materials damage (e.g., soiling, corrosion).[5] The annual secondary $PM_{2.5}$ standard is set at a level of 15 $\mu g/m^3$, with an annual arithmetic mean averaged over three years, and a secondary 24-hour $PM_{2.5}$ standard is set at a level of 35 $\mu g/m^3$, as the 98th percentile of the 24-hour average, averaged over 3 years. The annual secondary PM_{10} standard is an annual arithmetic mean, averaged over three years with a level of 50 $\mu g/m^3$, and the secondary 24-hour PM_{10} standard is a 24-hour average of 150 $\mu g/m^3$, not to be exceeded more than once per year on average over a three-year period.

Welfare effects associated with PM that are not ecologically related (and therefore not included in this review), such as visibility impairment, climate effects and materials damage, and the health effects of PM (including particulate transformation products of oxides of nitrogen and oxides of sulfur) are being considered as part of a separate review of the NAAQS for PM (U.S. EPA, 2016). The health effects of oxides of nitrogen were considered in a separate assessment that was completed recently as part of the review of the primary (health-based) NAAQS for oxides of nitrogen (83 FR 17226). Similarly, the health effects of oxides of sulfur are currently being considered in a separate assessment as part of the review of the primary NAAQS for oxides of sulfur (83 FR 26752). In addition, NH_3 is not a criteria pollutant but is a precursor to PM (ammonium sulfate (NH_4SO_4) and ammonium nitrate (NH_4NO_3)) and is considered in this review to the extent that it contributes to atmospheric transformations and loading to ecosystems.[6]

This REA Planning Document presents an evaluation of information related to ecological effects that is newly available in the second draft of the *Integrated Science Assessment for Oxides of Nitrogen, Oxides of Sulfur, and Particulate Matter— Ecological Criteria* (U.S. EPA,

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[5] The secondary PM standards were most recently reviewed in 2012 (78 FR 3086, January 15, 2018).

[6] The scientific and technical information and analyses in this review are expected to inform our understanding of the contribution of NH_3 to total N deposition and N-related ecological effects, as well as on the role of NH_3 in the formation of PM (which is a criteria pollutant).

2017a; hereafter referred to as the second draft ISA). Advances in modeling tools and techniques and air quality data that have become available since the last review are also considered. This document is intended to facilitate consultation with the Clean Air Scientific Advisory Committee (CASAC), as well as an opportunity for public participation, on the evaluation of the potential support in the newly available scientific evidence (including existing and historical air quality patterns and trends), new and/or improved data, methods, and tools, for conducting updated quantitative assessments, and on the plan for such analyses, as warranted. Additionally, this document evaluates the extent to which new information, tools or methodologies, will address or improve our consideration of important limitations or uncertainties associated with the analyses from the last review (summarized in chapter 2). Based on these considerations and our preliminary conclusions on the extent to which updated quantitative analyses of ecological risks and/or exposures are warranted in the current review, this document presents general plans for such analyses.

1.1 BACKGROUND

Sections 108 and 109 of the CAA govern the establishment and periodic review 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 air pollutants that in his/her “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… [the Administrator] plans to issue air quality criteria…” CAA section 108(a)(1). The NAAQS are established for these pollutants. The CAA requires that NAAQS are to be based on air quality criteria, which 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 that may be expected from the presence of [the] pollutant in the ambient air…” CAA section 108(a)(2). Under CAA section 109 [42 U.S.C. 7409], the EPA Administrator is to propose, promulgate, and periodically review, at five-year intervals, “primary” (health-based) and “secondary” (welfare-based)[7] NAAQS for such pollutants for which air quality criteria are issued. Section 109(b)(2) of the CAA directs that a secondary standard is to “specify a level of air quality the attainment and maintenance of which, in the judgment of the Administrator, based on such criteria, is requisite

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[7] Section 302(h) of the CAA provides that all language referring to effects on welfare includes but is not limited to, “…effects on soils, water, crops, vegetation, man-made 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…”

to protect the public welfare from any known or anticipated adverse effects associated with the presence of [the] pollutant in the ambient air."[8] Based on periodic reviews of the air quality criteria and standards, the Administrator is to make revisions in the criteria and standards, and promulgate any new standards, as may be appropriate. The CAA also requires that an independent scientific review committee review the air quality criteria and standards and recommend to the Administrator any new standards and revisions of existing air quality criteria and standards as may be appropriate, a function now performed by the CASAC.

The overall plan for this review was presented in the Integrated Review Plan (IRP) (U.S. EPA, 2017b), which discusses the preparation of key documents in the NAAQS review process including an ISA, an REA, as warranted, and a Policy Assessment (PA). In general terms, the ISA provides a critical assessment of the latest available scientific information upon which the NAAQS are to be based. The purpose of the REA in a secondary standards review is to estimate risk and exposure to public welfare associated with the current standards and, if appropriate, evaluate potential improvements in public welfare that could be achieved from meeting potential alternate standard(s). The PA evaluates the policy implications of the information contained in the ISA and of any policy-relevant quantitative analyses, such as a quantitative REA, that were performed for the review. Based on that evaluation, the PA presents conclusions regarding standard-setting options for the Administrator to consider in reaching decisions on the NAAQS.[9] The general NAAQS review process is illustrated in Figure 1-1, below.

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[8] Section 109(b)(1) [42 U.S.C. 7409] of the CAA defines a primary standard as one "the attainment and maintenance of which in the judgment of the Administrator, based on such criteria and allowing an adequate margin of safety, are requisite to protect the public health."

[9] Review of the NAAQS involves consideration of the four basic elements of a standard: indicator, averaging time, form, and level. The indicator defines the pollutant to be measured in the ambient air in order to determine compliance with the standard. The averaging time defines the time-period over which air quality measurements are to be obtained and averaged or cumulated. 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. The level of a standard defines the air quality concentration used (i.e., an ambient air concentration of the indicator pollutant).

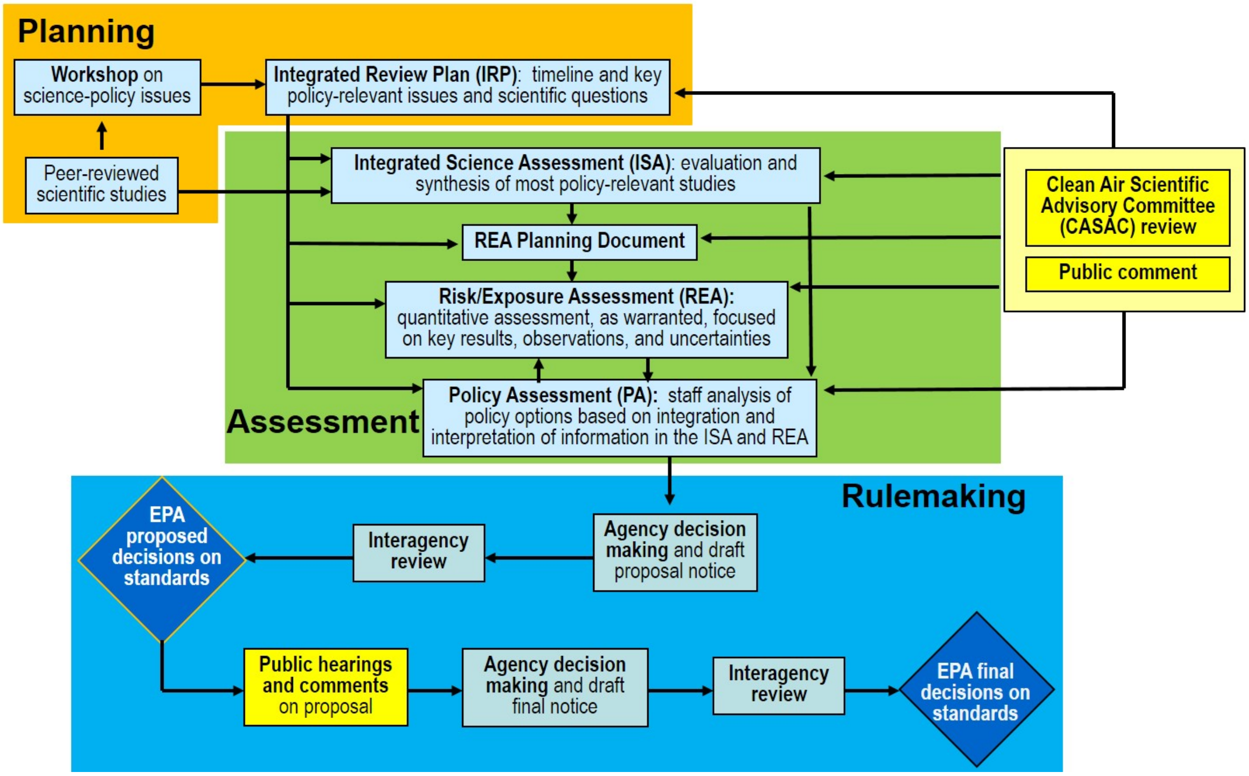

Figure 1-1. General NAAQS Review Process

1.2 EXPOSURE AND RISK ANALYSES FOR OXIDES OF NITROGEN, OXIDES OF SULFUR AND PM

This REA Planning Document serves to provide support for conducting any new or updated quantitative assessments in the current review. Conclusions regarding such support are based on our consideration of the available scientific evidence; the available technical information, tools, and methods; and judgments as to the likelihood that quantitative assessments will add substantially to our understanding of risk or exposure related to ecological effects, beyond the insights gained from the assessments conducted in the last review. Specifically, this consideration also includes an evaluation of the newly available data, tools and methods and whether they would be expected to reduce previously identified uncertainties or limitations from the last review.

1.2.1 Context for Analyses

The most recent review of the secondary NO_2 and SO_2 NAAQS was completed in 2012. Technical analyses for that review focused on 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 current secondary NO_2 and SO_2 standards were developed to protect against, and (2) effects associated with nitrogen and sulfur deposition to sensitive aquatic and terrestrial ecosystems (77

FR 20218, April 3, 2012). The REA for the 2012 review presented assessments of aquatic and terrestrial acidification-related and nutrient enrichment-related effects. Based on the REA findings, the PA gave primary attention to aquatic acidification effects from deposition of nitrogen and sulfur, and developed an aquatic acidification index (AAI)— an equation that relied upon ecosystem and air quality modeling to estimate the ecosystem's natural ability to buffer acidic deposition from ambient nitrogen and sulfur deposition.

In the Agency's consideration of the scientific and technical information and analyses with regard to the potential use in establishing a new standard based on aquatic acidification, several important uncertainties and data limitations were recognized, both those related to characterizing the relationships between N and S deposition and ecological effects generally, and those specifically relating to the development of an AAI. Thus, while recognizing the scientifically-supported conceptual basis for an AAI-based standard,[10] the EPA also recognized current limitations in relevant data and uncertainties that, together, were concluded to be too great to support a standard that would meet the requirements of the Act (77 FR 20218, April 3, 2012).

The current review of the secondary NO_2 and SO_2 standards differs from the review completed in 2012 in that the current review also includes consideration of the secondary PM standards. Specifically, the Agency is considering secondary standards for these three pollutants together with regard to protection against adverse ecological effects on public welfare and particularly such effects related to atmospheric deposition. Given the contribution of nitrogen compounds to PM, including but not limited to those related to oxides of nitrogen, the current review provides for an expanded and more integrated consideration of N deposition and the current related air quality information.

1.2.2 Key Technical Issues

Because NO_x, SO_x and PM are deposited from the ambient air into ecosystems where they have the potential to affect individual organisms and communities, considerations in this review will include potential impacts on the public welfare from alterations in structure and function of ecosystems. Important considerations for the review, which the REA will inform, include:

- **Source contributions and loading.** It is important to consider the emissions sources, chemical species and contributions to overall loading to ecosystems. The ability to evaluate modeled data as well as the ability to characterize components of ambient air

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[10] The Administrator recognized that while an AAI-based standard was innovative and unique, the structure of the proposed standard was well-grounded in the science underlying the relationships between ambient concentrations of oxides of nitrogen and sulfur and the aquatic acidification related to deposition of nitrogen and sulfur associated with such ambient concentrations (77 FR 20251, April 3, 2012).

and atmospheric deposition are important to understanding the potential effects of the pollutants on ecosystems. In some systems atmospheric deposition is not the only source of loadings and N and S can be introduced into the system from other sources (e.g., surface water runoff). Additionally, the composition of emissions in a given area will have an effect on what types of chemical species are contributing to atmospheric deposition. Using analytical tools and data to help understand what pollutants are contributing to the overall effect will be important, particularly in the case of reduced nitrogen impacts[11].

- **Historic air quality impacts on deposition-related effects.** Historic air quality and associated deposition can have an appreciable impact on ecosystems in many areas and will be an important consideration in understanding the role of current air quality versus historic air quality on deposition-related effects. For example, in areas where deposition levels have been historically high there may be terrestrial systems where the soil is saturated or aquatic systems where improvements to water quality are impeded by the area's geology (e.g., underlying bedrock). In such cases, the continued role of this historic deposition in the area's deposition-related impacts may mean that we would not expect the deposition-related risk and exposures to be as responsive to changes in atmospheric deposition, and that there would be a substantial delay prior to any such response.
- **Timescale of effects and potential for recovery.** Another consideration for this review is to understand the timescale for the effects being considered. For example, the response time for trees to stressors is very different than for fish populations or herbaceous plants. This affects both the exposure period necessary for effects to become adverse, as well as the uncertainty associated with the ability of the REA to assess these impacts on public welfare. In addition, while information may be limited, understanding the scientific evidence indicating the potential for recovery or improvement is also important to consider.
- **Spatial scale of effects.** It is important to consider the spatial scale, magnitude, and associated variability, of some ecological effects. The effects of N, S and PM deposition on aquatic and terrestrial ecosystems occur over time and are a result of atmospheric reactions and transport. The evaluation of environmental responses to these pollutants, accordingly, will need to consider the variability of environmental characteristics of ecosystems across the nation, including those related to ecosystem susceptibility and to the relative importance of individual effects (such as acidification or N enrichment).
- **Impacts to public welfare.** Importance to the public welfare is an important consideration to the review. For example, potential impacts in areas with special federal protections, and lands set aside by states, tribes, and public interest groups to provide similar benefits to the public welfare, may be of particular importance. The relevance of such areas to consideration of effects on the public welfare has been recognized in past NAAQS decisions (e.g., 80 FR 65292, October 26, 2015). Such areas include Class I

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[11] NH_3 is not a criteria pollutant but is a precursor to PM and is considered in this review to the extent that it contributes to atmospheric transformations and loading to ecosystems.

areas[12] which are federally mandated to preserve certain air quality related values. Class I areas and other parks have been afforded special federal protection to preserve services that provide for the enjoyment of these resources for current and future generations.

1.2.3 Conceptual Model

In each NAAQS review, selection of the approach most appropriate for the characterization of risks is influenced by the nature and strength of the evidence for the subject pollutants. Depending on the type of evidence available, analyses may include quantitative risk assessments based on exposure-response, or ambient air concentration/deposition-response relationships. Figure 1-2 below illustrates our conceptual model for evaluating ecological risk and exposure of oxides of nitrogen, oxides of sulfur and PM in this review, including exposure from direct pathways and through atmospheric deposition. Included in Figure 1-2, below is a basic conceptual diagram highlighting the relevant pathways of exposure for the pollutants in this review.

As described in chapter 4, the REA for this review will include an evaluation of the relationship between emissions of the three criteria pollutants (and their precursors) and ambient air concentrations, as well as their contribution to direct as well as deposition-related exposures to biota, soils, sediments and surface waters. While the review focuses on atmospheric inputs, non-atmospheric inputs of the pollutants that may occur through runoff of the pollutants from other human activities, such as agriculture and industrial processes, will also be considered as part of the total loading of the pollutant to the ecosystem. The ecological effects of these inputs (e.g., nutrient enrichment) will then be evaluated based on the biogeochemical and/or ecological responses of “receptors” [13] (e.g., plants, wildlife) within ecosystems.

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[12] Areas designated as Class I include all national 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 six thousand 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 Act.

[13] A receptor is defined as a biological element that is impacted by the conditions created by atmospheric deposition.

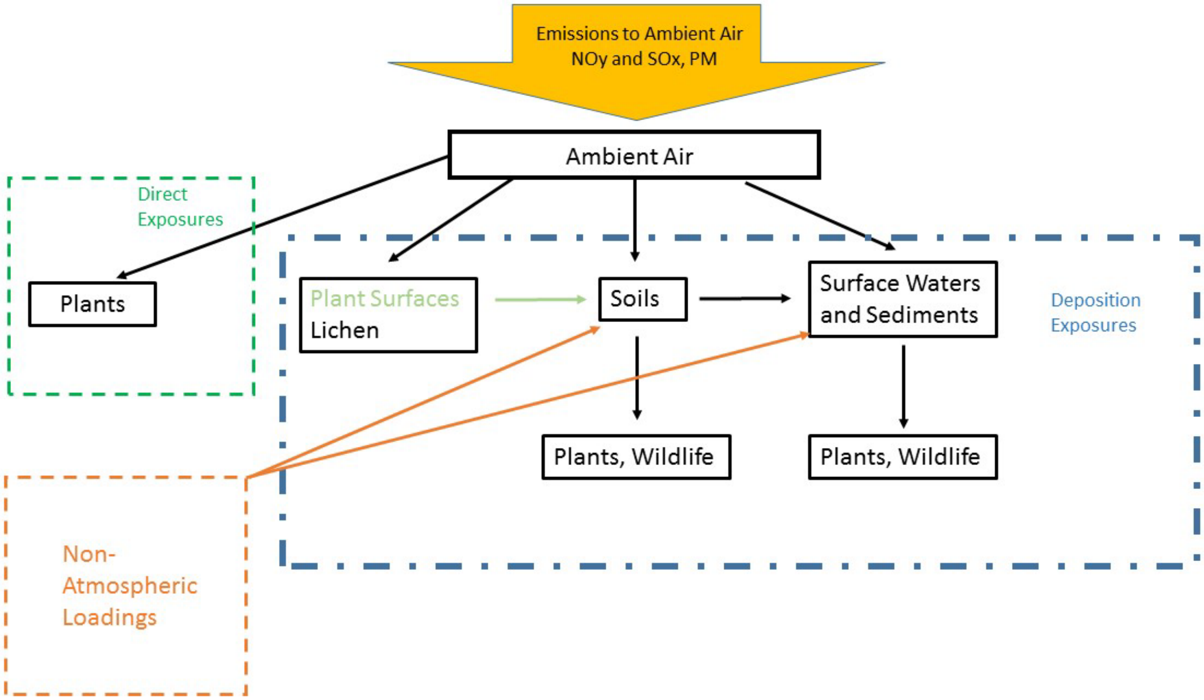

Figure 1-2. Conceptual Model.

1.3 ORGANIZATION OF THIS DOCUMENT

The remainder of this document presents our evaluations and preliminary conclusions regarding the degree to which available evidence and information address important uncertainties and the support for updated or new quantitative analyses in the current review. Chapter 2 provides background information regarding the assessments conducted in the prior reviews, evaluating deposition-related effects of oxides of nitrogen, oxides of sulfur, and PM as well as direct effects of oxides of nitrogen and oxides of sulfur. Chapter 2 also includes information regarding any uncertainties that were identified in the last reviews. Chapter 3 includes consideration of information newly available in this review, including scientific evidence and data for various ecological effects. This chapter also includes consideration and conclusions regarding whether this new information is expected to reduce uncertainties or limitations from the prior review and inform updated or new quantitative assessments for this review. Chapter 4 presents an overview of the REA analysis approach, summarizes approaches for characterizing air quality, and describes the plan for quantitatively evaluating ecological effects in various ecosystems.

2 OVERVIEW OF PREVIOUS ASSESSMENTS

This chapter summarizes the analyses and limitations and/or uncertainties related to ecological risk and exposure as assessed in the two previous reviews: the secondary NAAQS review of oxides of nitrogen and sulfur; and the review of particulate matter. More detailed information on the body of evidence and causal determinations for these reviews can be found in the *Integrated Science Assessment for Oxides of Nitrogen and Sulfur - Ecological Criteria* (U.S. EPA, 2008; hereafter referred to as the 2008 NOx/SOx ISA) as well as the *Integrated Science Assessment for Particulate Matter* (U.S. EPA, 2009; hereafter referred to as the 2009 PM ISA). More detailed descriptions of the assessment approaches in the previous oxides of nitrogen and sulfur joint review can be found in the *Risk and Exposure Assessment for Review of the Secondary National Ambient Air Quality Standards for Oxides of Nitrogen and Oxides of Sulfur: Final* (U.S. EPA, 2009; hereafter referred to as 2009 NOx/SOx REA) as well as the *Policy Assessment for the Review of the Secondary National Ambient Air Quality Standards for Oxides of Nitrogen and Oxides of Sulfur: Final* (U.S. EPA, 2011a; hereafter referred to as the 2011 NOx/SOx PA). More details about the previous PM review can be found in the *Particulate Matter Urban-Focused Visibility Assessment - Final Document* (U.S. EPA, 2010a)[14] and in the *Policy Assessment for the Review of the Particulate Matter National Ambient Air Quality Standards* (U.S. EPA, 2011b).

In each NAAQS review, selection of the approach most appropriate for the characterization of risk is influenced by the nature and strength of the evidence for the subject pollutants. This chapter summarizes the assessment approach used in the last review of NO_x and SO_x, which focused on evaluating the protection provided by secondary standards for oxides of nitrogen and sulfur for two general types of effects: (1) direct effects on vegetation associated with exposure to gaseous oxides of nitrogen and sulfur in the ambient air, which are the effects that the current NO_2 and SO_2 secondary standards were set to protect against; and, (2) effects associated with the deposition of nitrogen (N) and sulfur (S) to aquatic and terrestrial ecosystems. This chapter also summarizes the assessment approach used in the last PM review to evaluate direct and indirect effects of non-N and non-S deposited particles (including metals and organics).

2.1 AIR QUALITY

The 2009 NOx/SOx REA examined the path from emissions to air concentrations, providing characterizations of (1) major emissions sources of NO_x, NH_3 and SO_2, (2)

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[14] Note that this review does not cover visibility.

atmospheric concentrations of NO_y and SO_2, and (3) estimates of deposition of total N and S across the U.S. and for specific case study areas (see 2009 NOx/SOx REA, sections 3.2 and 3.3). Data from the 2002 National Emission Inventory (U.S. EPA, 2006) were assessed in the 2009 NOx/SOx REA to understand the major emissions sources of N and S across the U.S., and used as inputs to the Community Multiscale Air Quality (CMAQ) model to provide estimates of atmospheric concentrations and deposition of N and S in the U.S. Spatial fields representing 2002 annual average N and S deposition were also created by extracting the dry deposition predictions from the CMAQ model simulations and combining them with the wet deposition measurements from the National Atmospheric Deposition Program (NADP) National Trends Network (NTN).

Based on these analyses, the REA concluded that there was significant variation both spatially and seasonally across the country with the eastern U.S. receiving much greater deposition of both N and S than the states west of the Mississippi river. CMAQ simulations of scenarios with emission reductions of NO_x, SO_x, and NH_3 were used to examine the impact of these emissions to specific ecosystems. At the time of the REA analysis, oxidized N was the largest contributor to deposition, and the 2009 NOx/SOx REA found that a 50% reduction in NO_x emissions led to a 30% - 40% reduction in N deposition across most of the Eastern US. In locations with more NH_3 emissions, such as portions of the Potomac River Estuary and the Neuse River Estuary, the 50% NH_3 emission reduction led to a 40% - 50% reduction in N deposition. For SO_2 emissions, a 50% reduction resulted in a nearly 50% reduction in S deposition. The 2009 NOx/SOx REA concluded that most of the N and S deposition can be attributed to emissions of NO_x, NH_3, and SO_2. However, the REA also recognized that not all loadings of N and S compounds to freshwater, estuarine, and wetland ecosystems are due to atmospheric deposition. Other inputs, such as runoff from agricultural soils and point-source discharges, also contribute to acidification and N enrichment. Thus, some ecosystems may be solely impacted by atmospheric deposition (e.g., high elevation lakes), while ecological effects attributed to N and S in other systems might be largely due to non-atmospheric sources (e.g., high order streams). Sources to total loading of N and S was discussed more in the 2011 NOx/SOx PA in consideration of the standard-setting process.

Building upon these 2009 NOx/SOx REA analyses, the 2011 NOx/SOx PA further explored the relationship between atmospheric concentrations and deposition of N and S. In doing so, the 2011 NOx/SOx PA introduced the concept of a “transference ratio” and defined it as the ratio of deposition to ambient atmospheric concentrations. The transference ratios were calculated in the PA by dividing the 2002 CMAQ model outputs of the annual average of deposition of NO_y or SO_x by the annual average of ambient air concentrations of NO_y or SO_x, respectively. These annual average deposition and ambient air concentration values were estimated for every CMAQ model grid cell (e.g., 12 km by 12 km) and then averaged across areas of ecological similarity (i.e., spanning 100 km or more, as defined by level III ecoregions). The transference ratios were then used to characterize how changes in air quality concentrations of NO_y or SO_x related to changes in deposition of NO_y or SO_x in each of these ecological areas.

Limitations and uncertainties related to linking atmospheric concentrations to deposition are summarized in the 2011 NOx/SOx PA in Table 7-3, and the 2009 NOx/SOx REA in Section 3.6. Frequently identified issues include lack of spatial coverage and reference methods for concentration measurements, CMAQ modeling uncertainties, especially with respect to emission estimates, and the inherent difficulty in measuring dry deposition.

Table 2-1. Noted Uncertainties from the REA and PA in the Prior Review

Source of uncertainty	Description of uncertainty from the previous NOx/SOx review
Measurements of atmospheric concentrations of NO_y	A Federal Reference Method (FRM) for NO_y instruments was not available and existing techniques were known to have a negative bias. The spatial coverage of existing NO_y measurements was not adequate for assessing effects outside of urban areas.
Measurements of atmospheric concentrations of SO_x	A lack of adequate spatial coverage was the primary concern for $SO_2 + SO_4^{2-}$ observations.
Atmospheric deposition of NO_y, derived from NTN measurements of wet deposition and CMAQ estimates of dry deposition	Quantifying uncertainty in CMAQ estimates of dry deposition was hampered by a lack of dry deposition measurements.
Atmospheric deposition of SO_x, derived from NTN measurements of wet deposition and CMAQ estimates of dry deposition	There was general consensus that the overall mass balance of S was treated well in CMAQ. As with all dry deposition estimates, technologies for direct measurements were not available.
Atmospheric deposition of NH_x, derived from NTN measurements of wet deposition and CMAQ estimates of dry deposition	A lack of both NH_3 and ammonium (NH_4^+) ambient observations made it difficult to characterize uncertainty in NH_x deposition. As with all dry deposition estimates, technologies for direct measurements were not available routinely.
Deposition Transference Ratios, calculated using CMAQ	Transference ratios were not evaluated in a traditional model to observation context. Uncertainty was attributed to the information (e,g., NH_3 emissions) driving these calculations and availability of observations to evaluate model behavior.
Emissions of NO_x, SO_x, and NH_3	Uncertainty depends on the emission source. Emissions from electricity generating facilities are directly measured and have low uncertainty; however, emission estimates of livestock and fertilized fields do not capture all of the variability in these sources.

2.2 DIRECT EFFECTS

The 2008 NOx/SOx ISA determined that there was a causal relationship between exposure to nitric acid (HNO_3) and changes in vegetation, and a causal relationship between exposure to NO, NO_2, peroxyacetyl nitrate (PAN), and SO_2 and injury to vegetation. It was also noted that oxides of N and S gases have different degrees of phytotoxic effects on vegetation. In addition, while there was relatively little information about the direct effects of HNO_3 vapor on vegetation in previous reviews, research on the decline of sensitive lichen species was highlighted in the 2008 NOx/SOx ISA. In the mixed conifer forest of the San Bernardino Mountains, HNO_3 has been estimated to provide 60% of all dry deposited nitrogen, and it has been suspected as the cause of a dramatic decline in lichen species (see 2009 NOx/SOx REA, Section 6.4.3). At high concentrations over the short-term, HNO_3 can damage vascular plants such as seedlings of ponderosa pine (*Pinus ponderosa*) and California black oak (*Quercus kelloggii*) (see 2009 NOx/SOx REA, Section 6.4.3). The 2009 NOx/SOx REA noted that more research was needed to determine long-term exposure effects at lower concentrations. Additionally, at the time of the last review, there were no tools available for use in quantitative assessments of direct gaseous effects. For these reasons, the 2009 NOx/SOx REA included a qualitative discussion of direct gaseous effects but no quantitative analyses were conducted.

2.3 NON-N AND S EFFECTS OF PM

Direct and indirect ecological effects associated with deposited PM components (including metals and organics) were evaluated in the 2009 PM ISA. Direct deposition-related effects include alteration of leaf processes from deposition of PM ("dust") to vegetative surfaces. Indirect deposition-related effects encompass physiological responses associated with uptake of PM components and alterations to ecosystem structure and function. Evidence reviewed in the 2009 PM ISA was sufficient to determine that there is a likely causal relationship between PM deposition and a variety of effects on individual organisms and ecosystems.

In the REA for the prior PM review, issued in 2009,[15] the EPA determined that despite this likely causal relationship data was insufficient to support quantitative assessments for non-N and S deposition-related PM effects. After a careful evaluation of the evidence, the EPA determined that data needed to conduct quantitative assessments for ecological welfare effects in the last review were not available. A qualitative assessment was included in an Appendix of the REA (see 2009 PM REA, Appendix A).

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[15] U.S. EPA, Risk and Exposure Assessment for Review of the Secondary National Ambient Air Quality Standards for Oxides of Nitrogen and Oxides of Sulfur, September 2009, EPA-452/R-09-008a.

2.4 DEPOSITION-RELATED EFFECTS OF N AND S

Deposition-related ecological effects are those related to the deposition of mixtures of N- and S-containing compounds onto aquatic or terrestrial surfaces (e.g., vegetation, soil, or waterbodies). The ways in which ambient air pollution gets assimilated into ecosystems is a complex process, with many geochemical and biological factors contributing to the resulting deposition loading. The 2009 NOx/SOx REA assessed the ecological effects associated with deposition of N and S, focusing on four main targeted ecosystem effects on terrestrial and aquatic systems: (1) aquatic acidification[16]; (2) terrestrial acidification; (3) aquatic nitrogen enrichment[17]; and (4) terrestrial nitrogen enrichment[18,19]. In conducting these analyses, the 2009 NOx/SOx REA evaluated the relationships between atmospheric concentrations, deposition (wet and dry), biologically relevant exposures, targeted ecosystem effects, and, to the extent possible, associated ecosystem services. In doing so, the 2009 NOx/SOx REA recognized a lack of broad-scale data and that deposition-related effects were not evenly distributed across the U.S. Accordingly, the 2009 NOx/SOx REA used a case study approach for the quantitative assessments, building from the scientific information presented in the 2008 NOx/SOx ISA as well as the identification of ecosystems that are sensitive to N and/or S deposition.

To show the impacts of ecological effects on public welfare, the 2009 NOx/SOx REA and 2011 NOx/SOx PA qualitatively and quantitatively associated changes in ecological effects with changes in their ecological benefits and ecosystem services or welfare effects.

The sections that follow summarize the analysis of deposition-related effects as presented in the prior reviews, including (1) the relationship between emissions, air concentrations, and deposition; (2) the qualitative and quantitative approaches used to assess deposition-related ecological effects and related ecosystem services, where relevant; and (3) key limitations or uncertainties.

2.4.1 Aquatic Acidification

With respect to aquatic acidification, the 2008 NOx/SOx ISA for the prior review of oxides of nitrogen and sulfur determined that the evidence was sufficient to infer a causal

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[16] These analyses were then expanded upon in the 2011 NOx/SOx PA which focused on outlining an approach for a standard which focused on the ecological effects associated with acidifying deposition of oxides of nitrogen and sulfur in aquatic ecosystems. (see 2011 NOx/SOx PA, Chapter 7).

[17] Referred to as “nutrient enrichment” in the 2009 NOx/SOx REA. This effect includes eutrophication.

[18] Referred to as “nutrient enrichment” in the 2009 NOx/SOx REA.

[19] The REA also qualitatively addressed the influence of sulfur oxide deposition on MeHg production; nitrous oxide effects on climate; nitrogen effects on primary productivity and biogenic greenhouse gas fluxes; and phytotoxic effects on plants.

relationship between N and S deposition and changes in biogeochemistry and changes in biota in freshwater ecosystems.

The 2009 NOx/SOx REA conducted a case study evaluating acidifying effects on streams in the Shenandoah region of the southern Appalachian Mountains of Virginia (hereafter referred to as the Shenandoah), and lakes in the Adirondack Mountains of New York (hereafter referred to as the Adirondack). These areas were chosen because they were among the most severely acid-impacted regions in North America at the time the case study was conducted and had substantially high levels of N and S deposition known to have acidified a large number of waterbodies in both regions.

2.4.1.1 Assessment Approach

For the case studies, the impacts of acidification on waterbodies were assessed by (1) examining the amount and trends of N and S deposition using NADP data, (2) relating water quality condition to known biological impacts from acidification, (3) examining past, present, and future water quality conditions using long-term monitoring data and the Model of Acidification of Groundwaters in Catchments (MAGIC) biogeochemical model,[20] and (4) calculating critical loads (CLs) that relate deposition levels with water quality conditions. N and S deposition from all NADP monitors within the case study areas showed acidifying deposition had declined substantially since 1990, but still remained high relative to other regions in the United States and historical levels.

Both case studies used sulfate (SO_4^{2-}) concentrations, nitrate (NO_3^-) concentrations, and acid neutralizing capacity (ANC)[21] as the chemical indicators of deposition-driven acidification. ANC was particularly of interest, given its well understood relationship between deposition, water quality, and biological conditions, as well as its common use in modeling platforms (see 2009 NOx/SOx REA, section 4.2.1). Status of current conditions and trends in $SO4^{2-}$, NO_3^-, ANC concentrations measured in surface water were used to characterize links to the effects of acidifying deposition on the acid-base chemistry of the studied waterbodies, and to determine if conditions of the waterbodies were improving and recovering or were still degrading. These trends were analyzed along with monitoring data from the EPA-administered Temporally Integrated Monitoring of Ecosystems (TIME)/Long-Term Monitoring (LTM) programs along with survey data from EPA's Environmental Monitoring and Assessment Program (EMAP) for

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[20] MAGIC is a lumped-parameter model of intermediate complexity, developed to predict the long-term effects of acidic deposition on surface water chemistry (Cosby et al. 1985a,b,c, 2001). The model simulates soil solution and surface water chemistry to predict average concentrations of the major ions.

[21] ANC was defined in the 2009 NOx/SOx REA as "a key indicator of the ability of water to neutralize the acid or acidifying inputs it receives. This ability depends largely on associated biogeophysical characteristics, such as underlying geology, base cation concentrations, and weathering rates." (see "Key Terms" in U.S. EPA 2009)

the years 1990 to 2006. At the time the case studies were completed, the trends analysis found that the SO_4^{2-} concentrations were strongly declining in nearly all lakes in the Adirondack while ANC in many lakes were improving. The results of NO_3^- concentrations were mixed. For the Shenandoah case study, SO_4^{2-}, NO_3^-, and ANC concentrations were all variable with some sites improving while many were still declining.

MAGIC was used to produce estimates of past, present, and future water chemistry in 60 streams in the Shenandoah case study area and 44 lakes in the Adirondack case study area. Furthermore, MAGIC was used to evaluate the associated risk and uncertainty of the current levels of acidification given the pre-acidification water quality and the levels of uncertainty in the input parameters to the model. The MAGIC model output for each waterbody was summarized into five ANC levels that relate to biological impacts of acidification. The five ANC levels correspond to the aquatic status categories: *Acute Concern, Severe Concern, Elevated Concern, Moderate Concern,* and *Low Concern.* For each of these levels, the expected ecological effects were identified (see 2009 NOx/SOx REA, Table 4.2-1).

Additionally, after considering the five ANC levels that correspond to aquatic status categories, ANC "limits" of biological protection were selected: 0 μeq/L (acidic), 20 μeq/L (minimal protection), 50 μeq/L (moderate protection), and 100 μeq/L (full protection). The MAGIC modeling results showed that SO_4^{2-} and NO_3^- concentrations in the waterbodies at the time of the case studies were still well above pre-historical conditions. In addition, ANC levels were much lower, as compared to pre-historical levels, fostering ecological impacts. Lastly, the MAGIC model indicated that recovery of ANC was unlikely, assuming no changes in emissions between 2002 and 2050.

The 2009 NOx/SOx REA also calculated CLs[22] for waterbodies in each of the case study areas in order to assess whether current deposition of N and S was high enough to cause ecological effects. For these case studies, the calculated CLs used ANC limits of 0, 20, 50, and 100 μeq/L to define the biological risk to biota. From the 169 modeled lakes and 60 streams in the Shenandoah and Adirondack case study areas, respectively, the number and percentage of waterbodies that receive acidifying deposition above their CLs for a given ANC limit of 0, 20, 50, and 100 μeq/L were determined. These case studies indicated that large number of waterbodies were still being impacted by N and S deposition. Between 18 to 58 percent of modeled lakes in the Adirondacks and 52 to 93 percent of modeled streams in Shenandoah had exceeded their critical load in 2002 for a given ANC level of 0 to 100. (see 2009 NOx/SOx REA, section 4.2.4.2).

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[22] A critical load is formally defined as a quantitative estimate of exposure to one or more pollutants below which significant harmful effects on specified sensitive elements of the environment do not occur according to present knowledge (Nilsson and Grennfelt 1988, UNECE 2004).

Building from the 2009 NOx/SOx REA, the 2011 NOx/SOx PA focused on a potential new standard that could protect against ecological effects associated with acidifying deposition of N and S in aquatic ecosystems. As mentioned in Chapter 1 of this document, the 2011 NOx/SOx PA developed an approach to account for the geographical differences in sensitivity to acidification across the United States, called the Aquatic Acidification Index (AAI). In general, the AAI was meant to allow for consistent protection of ecosystems across the U.S. by selecting a single ANC value for each ecoregions in the U.S., and calculating how much acidifying deposition from N and S an ecosystem could accept before crossing the selected ANC limit, accounting for differences in ecosystem sensitivity.

The PA recognized that both dynamic (e.g., MAGIC) and steady state [e.g., Steady state water chemistry (SSWC)] models calculate ANC and can be used to calculate CLs, and discussed these models in the context of answering the question as to which modelling approach would be appropriate for development of a nationally applicable standard (see 2001 PA, section 7.2.2). The 2011 NOx/SOx PA additionally noted that information provided by steady state modeling of aquatic acidification would be sufficient to develop and analyze alternative NAAQS and the kind of protection they would afford. In addition, the 2011 NOx/SOx PA noted that the First-order Acid Balance (FAB) model includes more explicit modeling of N processes including soil immobilization, denitrification, in-lake retention of N and S, as well as lake size. Hence, the PA used a combination of the SSWC and the FAB model to inform development of the form of the standard (the AAI).

Recognizing the spatial variability across the U.S. of the factors in the AAI equation, the PA suggested that AAI values were meant to be calculated specifically for each ecologically relevant region (i.e., Ecoregion III (Omernik, 1987)). With regard to a level for the AAI, the PA concluded consideration should be given to a ANC threshold within the range of 20 to 75 μeq/L noting that a target ANC value of 20 μeq/L would be a reasonable lower end of this range, so as to protect against chronic acidification-related adverse impacts which have been characterized as severe on fish populations at ANC values below this level. Further, a target ANC value of 75 μeq/L would be a reasonable upper end of this range in recognition that the potential for additional protection at higher ANC values is substantially more uncertain in light of evidence that acidification-related effects are far less responsive to increases in ANC above this value (see 2011 NOx/SOx PA, section 7.7).

Ecosystem services related to aquatic acidification were discussed in the 2009 NOx/SOx REA and the 2011 NOx/SOx PA. The 2009 NOx/SOx REA included qualitative discussions of recreational fishing, which was identified as the service most relevant to aquatic acidification by freshwater lakes and streams (see 2009 NOx/SOx REA, section 4.2.1). The 2011 NOx/SOx PA included a quantitative assessment of the recreational fishing benefits to New York residents of reducing deposition of anthropogenic N and S for the Adirondacks case study area, but also recognized that there are numerous other ecosystem services that may be related to the ecological effects of acidification (see 2011 NOx/SOx PA, section 4.4).

2.4.1.2 Key Uncertainties/Limitations

An evaluation of the uncertainty in the parameters for the steady state critical load model was performed using a Monte Carlo approach in the 2009 NOx/SOx REA.[23] This probabilistic framework specifically assessed the degree of confidence in the exceedance values and the coefficient of variation (CV) of the critical load and exceedance values. The results suggested that the estimates of CLs and exceedances for the case study areas were robust. It was noted in the 2009 NOx/SOx REA, however, that this analysis may understate the actual uncertainty because some of the range and distribution types of critical load model parameters were not well understood for aquatic systems in the U.S. at the time.

An evaluation of the uncertainty for the water quality estimates and parameters used in the MAGIC model was also performed in the 2009 NOx/SOx REA. These uncertainty estimates were derived by running multiple calibrations of each site using the “optimization” tool and procedure as part of the MAGIC model. Direct comparison of simulated versus observed water chemistry values were compared to determine the uncertainty and variability in the MAGIC model output. Average water chemistry (SO_4^{2-}, NO_3^-, and ANC) simulated versus observed values during the calibration period (i.e., reference year) were compared for all modeled sites. The 2009 NOx/SOx REA found that the simulated and observed water quality values were in close agreement. (see 2009 NOx/SOx REA, section 4.2.8).

No formal analysis of the uncertainty in the AAI was performed in the 2011 NOx/SOx PA. However, the 2011 NOx/SOx PA noted that uncertainty and natural variability existed in all of the components of the AAI (for uncertainty analyses conducted for the 2011 NOx/SOx PA, see Appendix G). In addition, the 2011 NOx/SOx PA noted that there was no apparent directional bias in the uncertainty regarding the biological, chemical and physical processes incorporated in the AAI. Lastly, the 2011 NOx/SOx PA noted that the estimates for ecosystem services generally were believed to be biased low, meaning the monetary and non-monetary value of reaching a target level of protection is underestimated. However, quantification of these values was recognized as perhaps the most uncertain of all aspects considered.

Additionally, no formal analysis of uncertainty was performed for ecosystem services in the 2011 NOx/SOx/PA.

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[23] For a summary of the Monte Carlo approach used in the 2009 NOx/SOx REA, see section 4.2.8.1.

2.4.2 Aquatic Nitrogen Enrichment

With respect to aquatic N enrichment, the 2008 NOx/SOx ISA determined that the evidence was sufficient to infer a causal relationship between N deposition and the following:

- Biogeochemical cycling of N in freshwater and estuarine ecosystems
- Alteration to the biogeochemical cycling of carbon (C) in freshwater, estuarine, and near coastal marine ecosystems; and
- Alteration of species richness, species composition and biodiversity in freshwater and estuarine ecosystems

The 2009 NOx/SOx REA conducted a case study for estuaries in the Chesapeake Bay and the Pamlico Sound areas. Both areas were selected primarily based on the availability of data. Because the Chesapeake Bay and Pamlico Sound are fed by multiple river systems, the case study was scaled to one main stem river for each system: the Potomac River/Potomac Estuary (in the Washington, DC metropolitan area) and the Neuse River/Neuse River Estuary (in North Carolina).

2.4.2.1 Assessment Approach

The 2009 NOx/SOx REA focused the case study on aquatic nitrogen enrichment of estuaries and used the National Oceanic and Atmospheric Administration (NOAA)'s Assessment of Estuarine Trophic Status Eutrophication Index, commonly referred to as ASSETS EI (Bricker et al., 2007), as the ecological indicator for the case study. ASSETS EI is an estimation of the likelihood that an estuary is experiencing eutrophication or will experience eutrophication in the future based on five chemical and/or biological indicators: chlorophyll *a*, macroalgae, dissolved oxygen, nuisance/toxic algal blooms, and submerged aquatic vegetation (SAV) (Bricker et al., 2007).

Specifically, the analysis in this case study sought to determine the change in N loading required to improve the ASSETS EI from its current level set in the 2002 current condition analysis.[24] To create response curves for the ASSETS EI based on changes in N loads to an estuary, the case study used the U.S. Geological Survey's (USGS's) SPAtially Referenced Regressions on Watershed attributes (SPARROW) model to calculate the N loads.[25]

The case study found that a decrease of 78 percent of atmospheric N deposition would be required to improve the eutrophication index category ASSETS EI Score for the Potomac

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[24] Current conditions were evaluated using 2002 CMAQ model year and NADP monitoring data.

[25] SPARROW relies on a nonlinear regression formulation to relate water quality measurements throughout the watershed of interest to attributes of the watershed. SPARROW can be used to predict total N loads at the outlet of the watershed that result from changes in the total N atmospheric deposition loads.

River/Potomac Estuary from *Bad* to *Poor* (see 2009 NOx/SOx REA, section 5.2.7.1), and that the Neuse River/Neuse River Estuary ASSETS EI score could not be improved from *Bad* to *Poor* with only decreases in the 2002 atmospheric N deposition load to the watershed (see 2009 NOx/SOx REA, section 5.2.7.2). The 2009 NOx/SOx REA found that even if all atmospheric N deposition inputs were eliminated (100% decrease), the total annual N load to the Neuse River Estuary would only decrease by 4%. This small effect is because atmospheric deposition of nitrogen to the Neuse River watershed are small relative to other sources, such as run-off and agricultural sources.

The 2009 NOx/SOx REA included qualitative and quantitative assessments of ecosystem services related to aquatic nutrient enrichment in the Potomac River/Potomac Estuary and Neuse River/Neuse River Estuary (see 2009 NOx/SOx REA, section 5.2.1). The 2009 NOx/SOx REA evaluated several cultural ecosystem services, including recreational fishing, boating, and beach use. In addition, aesthetic and nonuse values were evaluated; the impacts on recreational fishing (e.g., closings, decreased species richness) to eutrophication symptoms through monitoring data were quantitatively linked; other recreational activities and aesthetic and non-use services to eutrophication symptoms were quantitatively related through user surveys and valuation literature; and the current commercial fishing markets were described. The 2011 NOx/SOx PA projected the quantitative change in the provision of these services based on the changes in water quality related to a policy scenario that eliminated the deposition of anthropogenic N to the Chesapeake Bay. The PA also included a hedonic study for aesthetic benefits related to improved water quality for near-shore residents. (see 2011 NOx/SOx PA, section 4.4.5)

2.4.2.2 Key Uncertainties/Limitations

The 2009 NOx/SOx REA noted potential uncertainties in the inputs and outputs of the SPARROW model, as well as sensitivity of the SPARROW model. The 2009 NOx/SOx REA also noted uncertainties in the inputs to the ASSETS EI, given the numerous data requirements and sources required to conduct a full ASSETS EI analysis.

The 2009 NOx/SOx REA determined that the small effect of decreasing atmospheric deposition in the Neuse River watershed is because the other N sources within the watershed are more influential than atmospheric deposition to the total nitrogen loadings to the Neuse River Estuary as estimated with the SPARROW model. The 2009 NOx/SOx REA noted that future application of the methods to case study areas where atmospheric deposition plays a larger role in the N loading to an estuary will likely provide more tangible results.

The 2011 NOx/SOx PA noted that the relative lack of empirical models and valuation studies imposed obstacles to the estimation of ecosystem services affected by N deposition resulting in underestimation of the monetary and non-monetary value of changes in ecosystem service provision. (see 2011 NOx/SOx PA, section 4.4.5)

2.4.3 Terrestrial Acidification

With respect to terrestrial acidification, the 2008 NOx/SOx ISA determined that the evidence was sufficient to infer a causal relationship between N and S deposition and changes in biogeochemistry and biota.

At the time of the 2009 NOx/SOx REA, *Picea rubens* (red spruce) and *Acer saccharum* (sugar maple) were the best studied species in North America with regard to impacts of acidification from N and S deposition to terrestrial systems. Hence, the 2009 NOx/SOx REA conducted a case study to evaluate the effects of acidifying deposition on sugar maple and red spruce physiological condition leading to impacts on tree growth and/or mortality in the Kane Experimental Forest and Hubbard Brook Experimental Forest, located in the Allegheny Plateau region in Pennsylvania and New Hampshire's White Mountains, respectively.

2.4.3.1 Assessment Approach

In the case study areas, critical load calculations were applied to multiple areas within 24 states for sugar maple and in 8 states for red spruce—site locations within each state were determined by the U.S. Forest Service's Forest Inventory and Analysis (FIA) database permanent sampling plots. The 2009 NOx/SOx REA noted that the Simple Mass Balance (SMB) model was a commonly used tool for evaluating soil acidification. The SMB model specifically examines a long-term, steady-state balance of base cation, chloride, and nutrient inputs, "sinks," and outputs within an ecosystem, and base cation equilibrium is assumed to equal the system's critical load for ecological effects. The 2009 NOx/SOx REA also noted that the base cation/aluminum (Bc/Al) ratio is a good indicator for soil acidification and relates well to the calcium/aluminum (Ca/Al) ratio in the soil solution. (see 2009 NOx/SOx REA, section 4.3.1.1) Hence, the 2009 NOx/SOx REA quantitative assessment included the use of the SMB model to calculate CLs for soil acidification, with the Bc/Al ratio as the indicator.

It should be noted that in order to properly use the SMB model, which is expressed as an equation, the user must first choose a critical level or levels for the Bc/Al ratio. For purposes of the 2009 NOx/SOx REA, data published by Sverdrup and Warfvringe (1993) was used to make decisions regarding the Bc/Al ratio as well as percent estimates for negative tree responses. Bc/Al values of 0.6 and 1.2 were selected based on a 50% and 75% chance of negative tree response (i.e., >20% reduced growth) for sugar maple and red spruce. A Bc/Al level of 10 was also chosen to represent the lowest impact (greatest level of protection) to tree growth. At the time of the 2009 NOx/SOx REA, it was considered the most conservative value used in studies that had calculated CLs in the U.S. and Canada. (see 2009 NOx/SOx REA, section 4.3.4).

One important input into the SMB model is base cation weathering (BCw) rates, which the 2009 NOx/SOx REA estimated using a clay-substrate method. At the time of the REA, this method was one of the most-commonly used methods for estimating this parameter.

Case study results suggested that the health of at least a portion of the sugar maple and red spruce growing in the U.S. may have been compromised with acidifying total N and S deposition in 2002. We concluded that the pattern of case study results suggests that N and S acidifying deposition in the sugar maple and red spruce forest areas studied were very close to, if not greater than, the CLs for those areas and both ecosystems are likely to be sensitive to any future changes in the levels of deposition.

The 2009 NOx/SOx REA also identified several ecosystem services that a deterioration of sugar maple and red spruce tree health could negatively impact. These include provisioning (use of trees for timber and maple syrup), cultural (endangered and threatened species habitat), regulating (soil stabilization and erosion control, water regulation, and climate regulation) and recreational ecosystem services (fall color viewing). The 2009 NOx/SOx REA used preference studies for the southern Appalachians to capture willingness to pay (WTP) for forest improvements that specifically addressed non-use values. Most services were qualitatively evaluated due to lack of data. However, the 2009 NOx/SOx REA did include a pilot study using the Forest and Agriculture Sector Optimization Model (FASOM) to estimate changes in timber harvest for red spruce and sugar maple. Uncertainties in model parameterization, however, were deemed too high to allow utilization of this model (see 2009 NOx/SOx REA, section 4.3.1).

2.4.3.2 Key Uncertainties/Limitations

As mentioned earlier, the 2009 NOx/SOx REA used the SMB model to estimate CLs for soil acidification, and used BCw as an input into the model. The 2009 NOx/SOx REA noted that one limitation of the SMB model is that it is a steady-state model and therefore does not capture the cumulative changes in ecosystem conditions. Additionally, the 2009 NOx/SOx REA also noted that the estimates for BCw rates and forest soil ANC input parameters were the main sources of uncertainty since these parameters are rarely measured and require researchers to use default values. The 2009 NOx/SOx REA also stated that the BCw value is strongly influenced by the classified acidity of the soil parent material, and is poorly measured in non-glaciated soils. (see 2009 NOx/SOx REA section 4.3.9). The 2009 NOx/SOx REA determined that the BCw rates contributed 49% to the total variability in the critical load estimates, and forest soil ANC contributed 46% to the total variability. (see 2009 NOx/SOx REA, section 4.4)

2.4.4 Terrestrial N Enrichment

With respect to terrestrial N enrichment, the 2008 NOx/SOx ISA determined that the evidence was sufficient to infer a causal relationship between N deposition and the alteration of the following:

1. Biogeochemical cycling of N and C;
2. Biogeochemical flux of methane (CH4) and nitrous oxide (N2O); and
3. Species richness, species composition, and biodiversity

At the time of the last review, there was a large body of evidence showing that some of the highest N deposition has occurred in Southern California, where researchers have documented measurable ecological changes related to atmospheric deposition. These changes include increases in nonnative grasses and fire susceptibility for Coastal Sage Scrub (CSS), as well as tree mortality, increased fire intensity, and a change in nutrient cycling in mixed conifer forests (U.S. EPA, 2008).

2.4.4.1 Assessment Approach

The 2009 NOx/SOx REA included a case study for CSS and Mixed Conifer Forest (MCF) communities in the Sierra Nevada Range and San Bernardino Mountains of California. No quantitative assessment was conducted for this case study. Rather, geographic information systems (GIS) analysis supported a qualitative review of past field research to identify ecological benchmarks associated with CSS and mycorrhizal communities, as well as MCF's nutrient-sensitive acidophyte lichen communities, fine-root biomass in Ponderosa pine, and leached NO_3^- in receiving waters. The benchmarks were identified from empirical studies, including CLs, in the southern California region. These benchmarks, ranging from 3.1 to 17 kg N/ha/yr for CSS and MCF, were compared to 2002 CMAQ/NADP data to discern any associations between atmospheric deposition and changing communities. Evidence supported the finding that N alters CSS and MCF communities.

The 2009 NOx/SOx REA also used data from Rocky Mountain National Park to examine the sensitivity and effects of nutrient enrichment on aquatic and terrestrial ecosystems, and found that exposure levels at which negative effects were observed appeared to be generally comparable to levels identified in other sensitive U.S. ecosystems (benchmarks range from 1.5 to 30.5 kg N/ha/yr). In addition, it included a qualitative assessment of ecosystem services, focusing on cultural services, including: habitat for endangered and threatened species, recreational (e.g., hiking, fishing, and hunting), aesthetic (view of the landscape), non-use (existence value), and regulation (e.g., water, regulation of fire intensity).

To assess the impact of fire risk, the 2009 NOx/SOx REA used GIS to overlay N deposition with the locations of MCF and CSS, and CALFIRE data (State of California fire occurrence, prevention and fire-fighting expenditures) to describe the potential benefit of reducing N enrichment to these fire-prone areas. This was accompanied by a discussion of the hedonic benefits to homeowners and aesthetic benefits to general public for reductions in fire damages. (see 2009 NOx/SOx REA, section 5.3.1)

The 2009 NOx/SOx REA provided a qualitative discussion of the services offered by CSS and MCF and a sense of the scale of benefits associated with these services. Specifically, the 2009 NOx/SOx REA stated the following: CSS and MCF are an integral part of the California landscape, and together the ranges of these habitats include the densely populated and valuable coastline and mountain areas of the state. Through recreation and scenic value, these habitats affect the lives of millions of California residents and tourists. Numerous threatened and endangered species at both the state and federal levels reside in CSS and MCF. Both habitats may play an important role in wildfire frequency and intensity, an extremely important problem for California. The potentially high value of the ecosystem services provided by CSS and MCF justify careful attention to the long-term viability of these habitats. (see 2009 NOx/SOx REA, p. 5-60).

2.4.4.2 Key Uncertainties/Limitations

The 2009 NOx/SOx REA noted that the exact relationship between atmospheric N loadings, fire frequency and intensity, and nonnative plants, particularly in the CSS ecosystem, had not been quantified in the scientific literature. The 2009 NOx/SOx REA noted that although various conceptual models linking these factors had been developed, an understanding of cause and effect, seasonal influences, and thresholds remained undeveloped.

Overall, the REA concluded that although the available data used for the targeted effect of terrestrial N enrichment were considered high quality, there was a limited ability to extrapolate these data to a larger regional area.

2.4.5 Mercury Methylation

With respect to mercury methylation, the 2008 NOx/SOx ISA determined that the evidence was sufficient to infer a causal relationship between S deposition and increased methylation of mercury in aquatic environments, where the value of other factors is within adequate range for methylation.

Information available at the time of the last review demonstrated that methylmercury (MeHg) production is mediated primarily by sulfur-reducing bacteria (SRB), and changes in SO_4^{2-} deposition result in changes in both Hg methylation rates and Hg concentration in fish. It was also shown that watersheds with conditions known to be conducive to Hg methylation could be found in the northeastern U.S. and southeastern Canada, though biotic Hg accumulation had been widely observed in other regions that had not been studied as extensively, and where a different set of conditions may exist.

The 2009 NOx/SOx REA acknowledged that a number of factors influence or modify the relationship between SO_4^{2-} and the methylation of Hg. It noted that while there appears to be a relationship between SO_4^{2-} deposition and mercury methylation, the rate of mercury methylation varies according to several spatial and biogeochemical factors whose influence has not been fully quantified (2009 NOx/SOx REA, section 6.2.1).

2.4.5.1 Assessment Approach

Given the factors considered, the 2009 NOx/SOx REA included a qualitative assessment of mercury methylation effects, focusing on Little Rock Lake in Wisconsin. It noted that decreases in SO_4^{2-} deposition were linked to observed decreases in MeHg fish tissue concentrations in the lake.

The 2009 NOx/SOx REA also discussed qualitatively the provisioning and cultural services potentially impacted by mercury methylation. The REA referenced commercial and sport fishing, shell fishing, fishing for subsistence, and the cultural and spiritual significance derived from fishing and consuming local fish or shellfish (particularly for Native Americans and Alaska Native villagers).

2.4.5.2 Key Uncertainties/Limitations

Overall, the REA noted that decreases in SO_4^{2-} deposition will likely result in decreases in MeHg concentrations, but that the rate of methylation varies spatially and with biogeochemical factors so that the correlation of SO_4^{2-} deposition and MeHg could not be quantified for the purpose of interpolating the association across waterbodies or regions (see 2009 NOx/SOx REA, section 6.2.2.2). This limitation hindered the ability to establish large scale dose-response relationships.

3 CONSIDERATION OF NEWLY AVAILABLE INFORMATION

In this chapter, we consider the extent to which our characterization of risk and exposure in the last review remains appropriately informative to the key questions in the current review, as summarized in the IRP. As noted in that document, the ISA, REA (if warranted) and PA developed in this review will provide the basis for addressing the key policy-relevant questions and will inform the Administrator's judgment as to the adequacy of the secondary NAAQS for oxides of nitrogen, oxides of sulfur, and PM. Accordingly, this chapter considers the extent to which any new assessment of risk and exposure is warranted to inform decisions regarding potential adverse effects to public welfare. Based on these considerations, as described below, we plan to develop a new REA to inform the current review.

3.1 KEY CONSIDERATIONS

In any NAAQS review, considerations contributing to a decision to conduct a new risk and exposure assessment include the role played by the risk and exposure information in the EPA's previous decisions on the existing standards and the role of risk and exposure information expected in the current review. Another important consideration is the robustness of the risk and exposure estimates for the existing standards that are available from the last review. In reaching a decision on conducting a new REA, we also consider the extent to which results of a new quantitative risk and exposure assessment are expected to appreciably change our understanding of risk and exposures beyond the insights gained from the assessments conducted for the last review. More specifically, we consider questions such as the following, which are summarized in Figure 3-1:

- Is appropriate scientific and technical information available to support quantitative assessments?
- Is the scientific and/or technical information that could inform updated quantitative assessments substantially different from that available in the previous reviews?
- Would the new information appreciably reduce the uncertainties and limitations identified in previous reviews?
- Would updated quantitative assessments likely inform decision making in the current review by adding substantially to our understanding of pollutant exposures or pollutant-attributable risks, beyond the insights gained from previous reviews and assessments?

Regarding these questions, section 3.2 below considers the newly available air quality information with a focus on current air quality concentrations and deposition and whether those levels across the U.S. have changed significantly since the last review. Additional consideration is also given to new tools, data and information that could inform improvements in the quantification of the relationship between ambient air quality concentrations and deposition, a key uncertainty in the previous review. Sections 3.3 through 3.7 then consider the scientific evidence and data on ecological effects available in the second draft ISA that play critical roles in our characterization of risk and exposure. Particular attention is given to consideration of new information pertaining to those aspects, and the extent to which it might be expected to address key areas of uncertainty identified in the last review. Based on these considerations, section 3.9 then provides preliminary conclusions on the extent to which updated quantitative analyses of ecological risk and exposure are warranted in the current review. This document then presents our proposed plan for performing such analyses in Chapter 4.

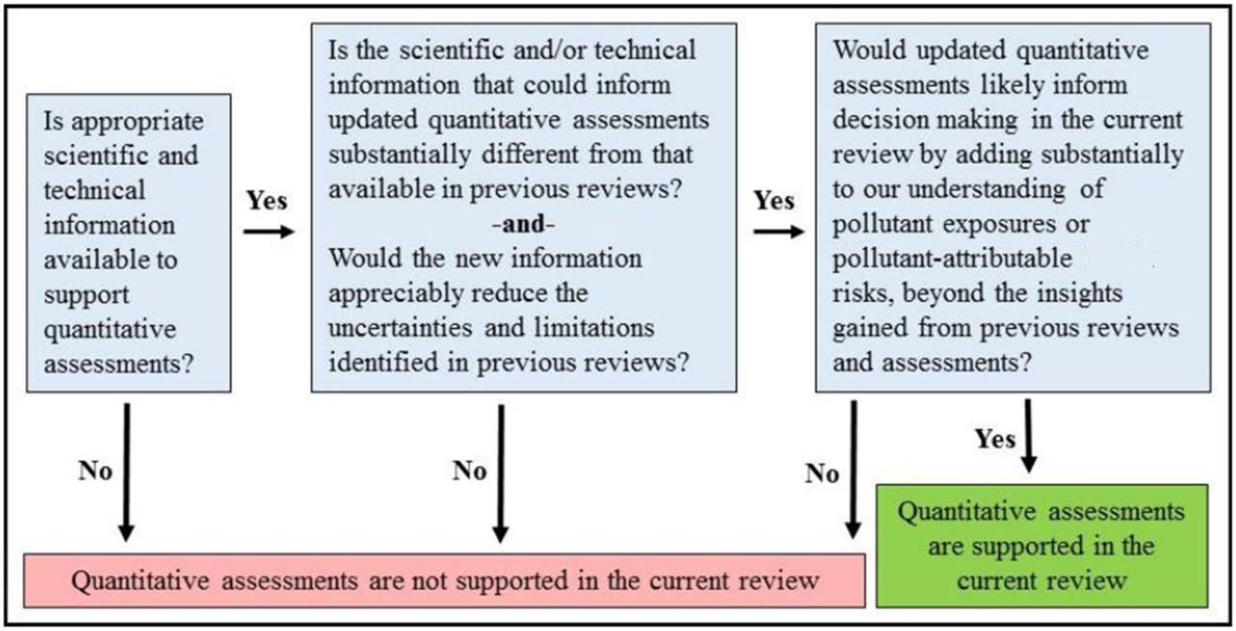

Figure 3-1. Key considerations for updated or new quantitative analyses

3.2 AIR QUALITY INFORMATION

3.2.1 Emissions, Atmospheric Concentrations and Deposition

Sulfur oxides are emitted into the air from specific sources (e.g., fuel combustion processes) and are also formed in the atmosphere from other atmospheric compounds (e.g., as an oxidation product of reduced sulfur compounds, such as sulfides). Sulfur oxides are also transformed in the atmosphere to particulate sulfur compounds, such as SO_4^{2-}. The most prevalent sulfur oxide in the atmosphere is SO_2. Emissions of SO_2 in the U.S. are largely due to coal-fired power plants and diesel fuel combustion (second draft ISA, Appendix 2, section 2.2.1).

Oxidized nitrogen species considered here range from NO and NO_2, collectively referred to as NO_x, to higher order organic and inorganic oxidation products, collectively referred to as NO_z (e.g., pNO_3, HNO_3, HONO, PAN, other organic nitrates). The largest sources of NO_x emissions are related to combustion sources, which includes power plants, industrial facilities, vehicles, and wood burning stoves. Non-controllable sources of NO_x include wildfires, biological soil processes, and lightning (second draft ISA, Appendix 2, section 2.2.1).

Reduced nitrogen species (i.e., $NH_3 + NH_4^+ = NH_X$) are considered in this review. While, NH_3 is not a criteria pollutant, it is considered to the extent that it contributes to atmospheric transformations and loading to ecosystems. NH_3 is a precursor for atmospheric PM, reacting with gas phase nitric acid (HNO_3) to form ammonium nitrate (NH_4NO_3), a major contributor to N deposition. Livestock and fertilized fields are the largest sources of NH_3, but there are combustion related sources as well. Particles containing SO_4^{2-}, NO_3^-, and NH_4^+ are also directly emitted into the atmosphere from sources such as wind-blown dust or sea-salt spray, but in total, direct particulate emissions are a small contribution to total emissions of N and S containing compounds (second draft ISA, Appendix 2, section 2.2.1).

The geographic distributions of NO_x and SO_x emissions reported in the National Emissions Inventory (NEI) reflect the fact that transportation and power generation source sectors dominate NO_x and SO_x emissions, respectively. In the last review, the density of emissions sources of NO_x and SO_x was highest in the eastern U.S., and around population centers and transportation corridors in both the east and west. This remains true for this review, though these areas of the country have also experienced some of the largest reductions in emissions of NO_x and SO_x (second draft ISA, Appendix 2, section 2.2.1). National average SO_2 emissions are estimated to have declined by 81% and NO_x emissions have declined by 53% over the period from 2002 to 2016. Such declines in emissions are likely related to the implementation of national control programs developed under the Clean Air Act Amendments of 1990, including Phase I and II of the Acid Rain Program, the Clean Air Interstate Rule, the Cross-State Air Pollution Rule, and the Mercury Air Toxic Standards, as well as changes in market conditions, e.g., reduction in energy generation by coal (U.S. EIA, 2018).

The largest sources of NH_3 emissions are agricultural: livestock, including confined animal feeding operations, and soils after addition of N containing fertilizers. Motor vehicles can also be a substantial contributor to total NH_3 emissions in urban areas. In contrast to SO_x and NO_x, Xing et al. (2013) estimated that the national emissions of NH_3 increased between 1990 to

2010. Because of this, in many places in the U.S., reduced forms of N are now the largest contributor to N deposition[26] (second draft ISA, Appendix 2, section 2.6.2).

In the atmosphere, NO_x, SO_2, and NH_3 undergo chemical transformations and the resulting compounds are removed from the atmosphere by dry deposition and wet deposition at different rates. For example, NO_2 can oxidize to form nitric acid (HNO_3), which dry deposits rapidly. However, in the presence of NH_3, particulate NH_4NO_3, which deposits via dry deposition slower than both HNO_3 and NH_3. Wet deposition, the scavenging of gases and particles by cloud droplets and precipitation, is the most important removal process for NH_4NO_3. The deposition rates of oxidized sulfur also depend on the chemical form; particulate sulfate does not dry deposit as rapidly as freshly emitted SO_2. Since the chemical form is important to determining the rate of dry and wet deposition, as well as the relationship between air concentrations and deposition, we use process-based models and quality-assured ambient measurements to understand the transformation from emissions to concentrations to deposition. Both measurements, models, and techniques to develop merged model-measurement datasets have been advanced since the previous review (see second draft ISA, Appendix 2, sections 2.5 and 2.6).

Several monitoring networks measure the atmospheric concentrations and wet deposition of NO_x, SO_x, and PM. The most relevant routinely operating networks measuring ambient air concentrations include the Interagency Monitoring of Protected Visual Environments (IMPROVE), the National Core network (NCore) and EPA Chemical Speciation Networks (CSN), Clean Air Status and Trends Network (CASTNet), and the ammonia monitoring network (AMoN). The National Trends Network (NTN) is part of the National Atmospheric Deposition Program and measures wet deposition of NO_3^-, SO_4^{2-}, NH_4^+, and other ions. Several new monitoring efforts are underway since the previous review. Gas phase NH_3 has expanded and as of May 2018, is measured at 98 sites as part of the AMoN network. The NCore network includes measurements of NO, NO_y, SO_2 and PM chemical composition at 63 urban and 17 rural sites. The near-road network includes NO_2 measurements at 80 monitoring sites located near roadways. Of these monitoring networks, the co-located CASTNet, AMoN and NTN sites are of particular interest because they measure both air concentrations and deposition of N and S containing compounds.

However, some limitations remain. Particulate NH_4^+ measurements from CASTNet and IMPROVE networks are thought to have biases that depend on meteorological conditions and require careful consideration when used for analysis. While methods for directly measuring dry

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[26] A portion of the reduced N deposition is from NH_4^+ bound in PM. The NH_4^+ fraction changes according to the relative levels of sulfate and NO_3^- available to form particles.

deposition in routine networks have been deployed in Europe (Skiba et al., 2009), direct measurements of dry deposition in the U.S. are sparse and infrequent. The spatial coverage of the monitoring networks is representative of some, but not all, of the variability in ecosystems and concentrations nationally. To help address the limitations in the available observations of air concentration and deposition, a combination of observations and computational models is often employed (see second draft ISA, section 2.6).

The National Atmospheric Deposition Program's total deposition science committee has been advancing the state of the science to create datasets that estimate annual average deposition. The Total deposition (TDEP) dataset combines measurements and models to estimate annual average deposition for years 2002 – 2016 (Schwede and Lear, 2014). Spatial coverage includes the continental U.S. at 4-km horizontal resolution. Wet deposition is estimated by spatial interpolation of NTN measurements and dry deposition is calculated by the Community Multiscale Air Quality (CMAQ) model. CMAQ is a chemical transport model that simulates the fate and transport of gases and particles by using physically-based, numerical models and explicitly including the processes of advection, dispersion, chemistry, aerosol physics, cloud processes, dry deposition, and wet deposition. The use of modeled estimates is necessary to account for dry deposition, which is not measured on a broad enough scale to inform national applications. Using the TDEP dataset to analyze ecosystem effects could potentially reduce uncertainties associated with the use of deposition values estimated by CMAQ alone, as was done in the last review. In addition, we note the representation of chemical and physical processes in CMAQ has been further improved since the last review, with updates that include gas-phase chemistry relevant to the oxidation of NO_x, dry deposition of particles by gravitational settling, interactions between meteorology and particles, and the influence of temperature, wind, and precipitation on NH_3 emissions (see second draft ISA, Appendix 2, section 2.5).

To further assess how deposition has been estimated to have changed since the last review, Figures 3-2 and 3-3 are included to illustrate TDEP estimates of S deposition across the U.S. for the years of 2002 and 2015, respectively, with Figure 3-4 showing the estimated changes in S deposition between these two years. Similarly, the TDEP estimates of N deposition for 2002 and 2015 are shown in Figures 3-5 and 3-6, with changes between these two years shown in Figure 3-7. These figures mimic the same general patterns, discussed above, for the changes in emissions since the last review. Generally, substantial changes in S deposition has occurred across most of the U.S., with the largest decreases occurring in the eastern U.S. Similarly, N deposition has also generally decreased across the entire U.S., with increases in areas of the country generally dominated by agricultural sources, due to an increase in reduced nitrogen deposition in these areas.

Given that emissions, concentrations and deposition levels of N and S species have changed substantially across the U.S. since the last review and given the availability of new and improved tools and methods for estimating this deposition (e.g., TDEP), we judge that new analyses are supported for this review. Assessment of the most recently available emissions, concentration and deposition datasets are likely to influence important differences in any new or updated quantitative assessments and inform our understanding of changes in pollutant exposures and pollutant-attributable risks since the last review. In addition, application of the improved tools and methods will likely reduce uncertainties and limitations in the characterization of air concentrations and deposition from the last review.

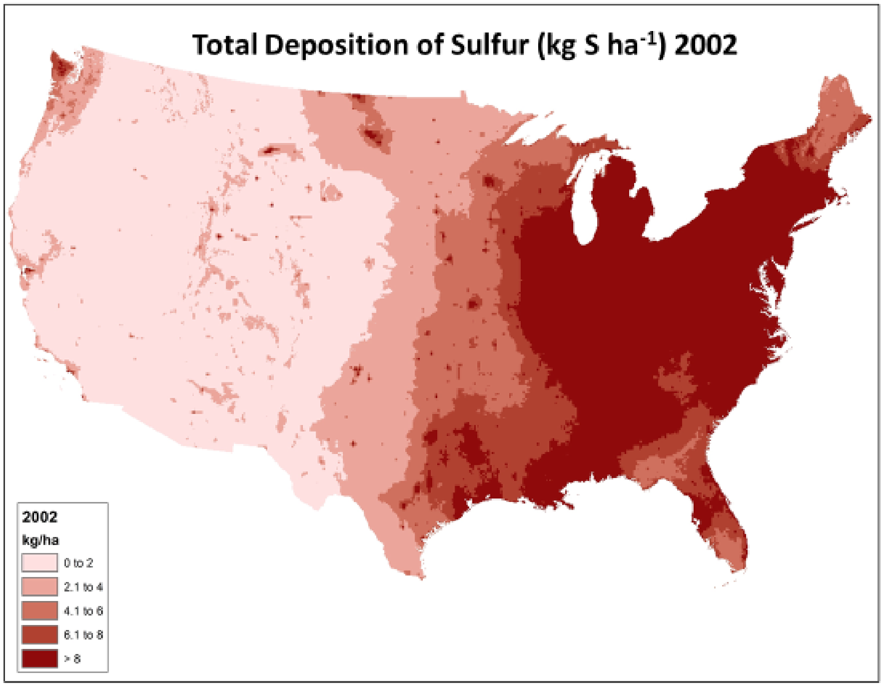

Figure 3-2. **Annual total deposition of sulfur, kg S ha^{-1} in 2002.**

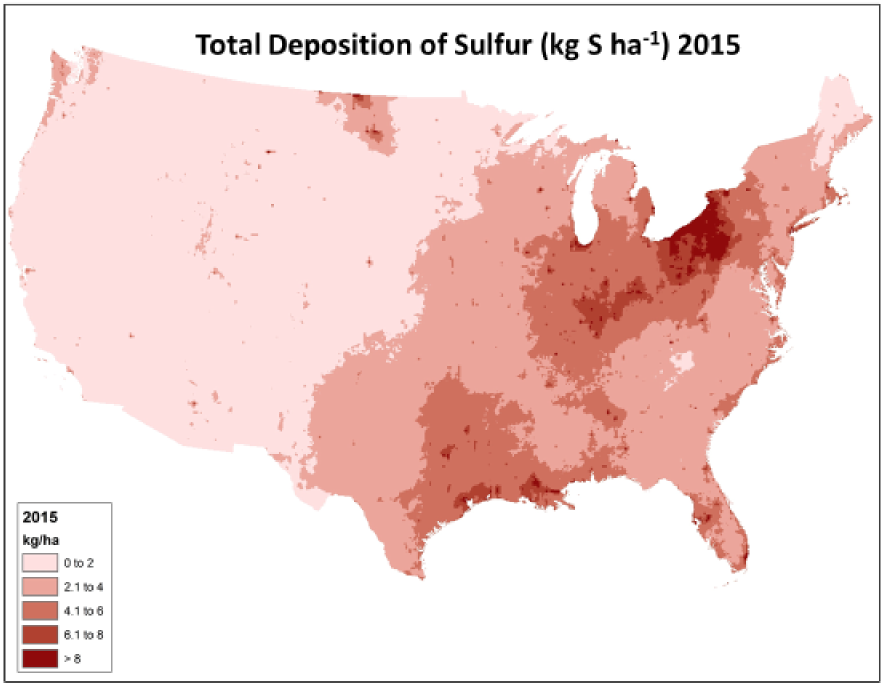

Figure 3-3. Annual total deposition of sulfur, kg S ha^{-1} in 2015.

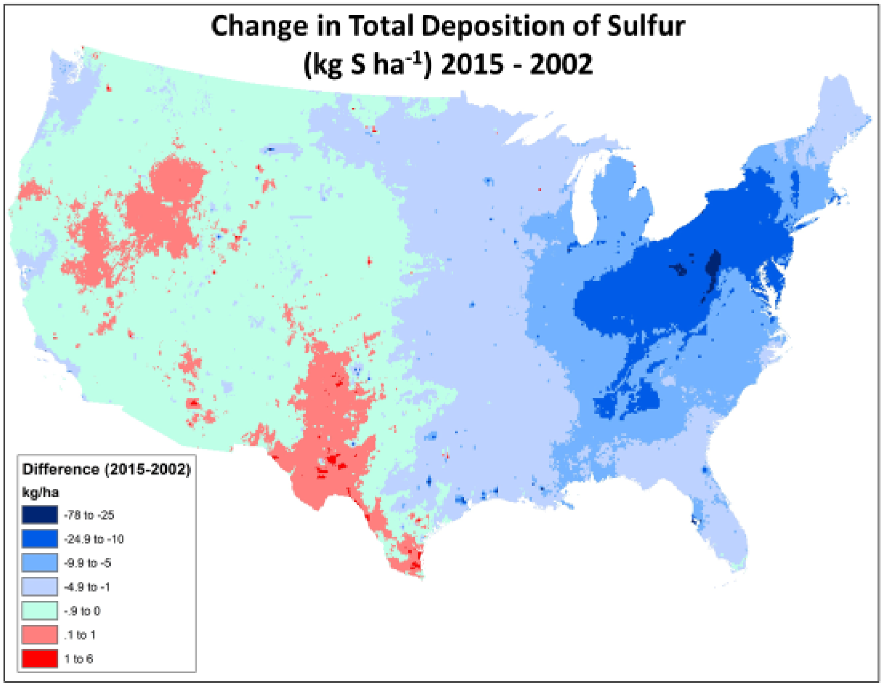

Figure 3-4. Change in annual total sulfur deposition, 2015 – 2002 (kg S ha^{-1})

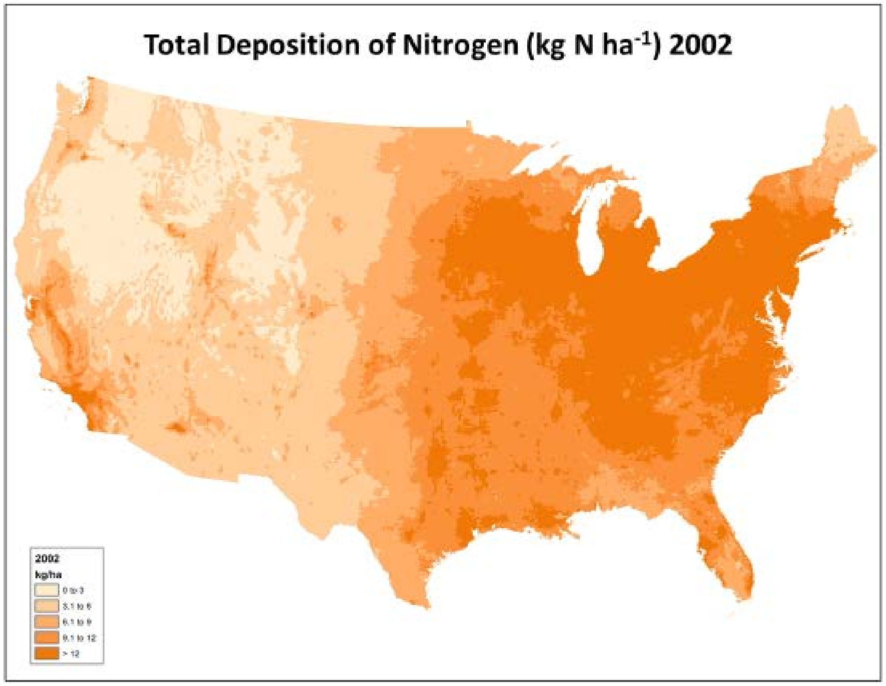

Figure 3-5. Annual total deposition of nitrogen, kg N ha^{-1} in 2002.

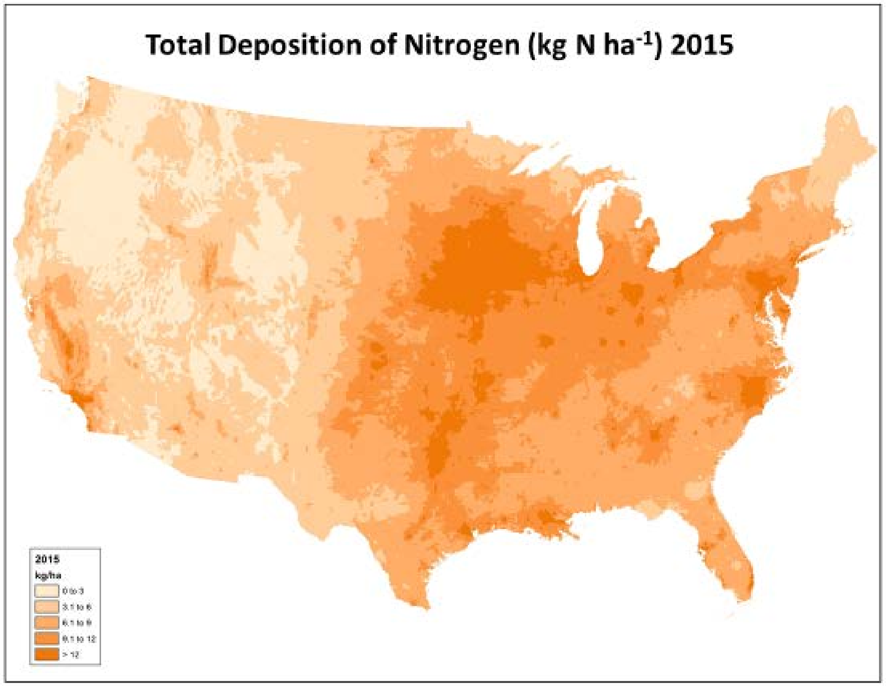

Figure 3-6. Annual total deposition of nitrogen, kg N ha^{-1} in 2015.

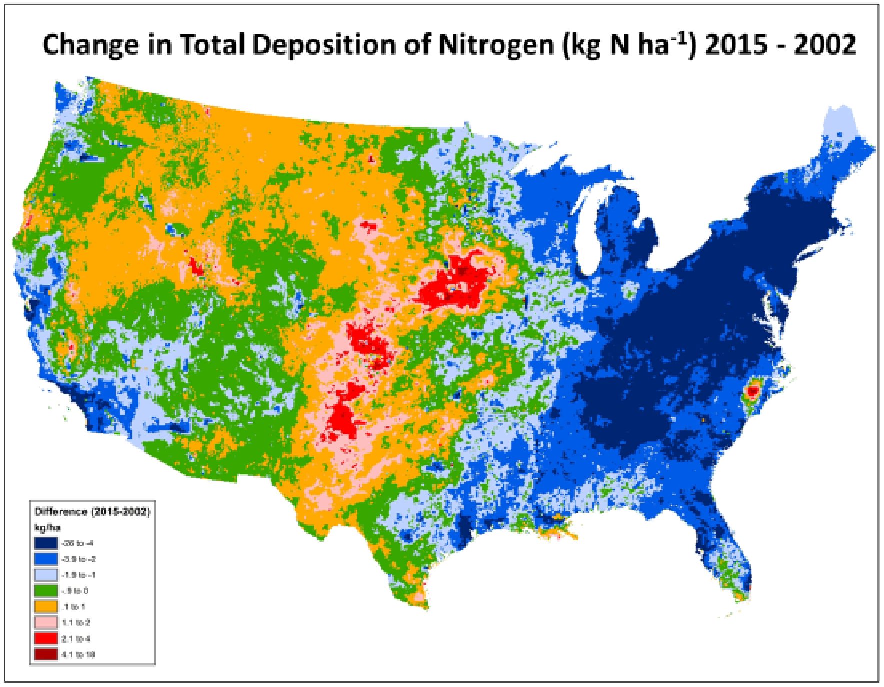

Figure 3-7. Change in annual total nitrogen deposition, 2015 – 2002 (kg N ha^{-1})

3.2.2 Linking Atmospheric Concentration Changes to Changes in Deposition

The new modeling approaches, additional years of measurements, and combined model-measurement TDEP estimates described in the previous section are also relevant for linking atmospheric concentration changes to changes in deposition. In the last review, the "transference ratio" approach was used to estimate the relationship between air quality concentrations and deposition of N and S. The uncertainties relevant to this transference ratio approach are summarized in Chapter 2. Also, since the last review, several studies have examined the transference ratio using measurements and modeling, as described in more detail in the second draft ISA, Appendix section 2.5.2.4. Specific areas of concern are the drivers of the spatial variability of the computed transference ratio, differences in the transference ratio when calculated by different chemical-transport models, and errors associated with using the air concentration of NO_y as a predictor of nitrogen deposition. These recent findings and uncertainties, listed in Chapter 2, will be used to inform the updated approach to relating changes in ambient concentrations to changes in atmospheric deposition, which is described in Chapter 4. Briefly, the updated approach analyzes 20-years of concentration and deposition measurements and CMAQ model results, includes consideration of uncertainty and variability using multiple chemical-transport models, and carefully evaluates how the concentration/deposition relationship may be different near sources versus in remote areas.

3.3 DIRECT EFFECTS

New scientific evidence supports the causality determinations made in the 2008 NOx/SOx ISA regarding the gas-phase effects of NO, NO_2, PAN, SO_2. and HNO_3 on vegetation; there are no new causal determinations.[27]

At the time of the 2008 NOx/SOx ISA, it was known that NO, NO_2, and PAN can have phytotoxic effects on plants by decreasing photosynthesis and inducing visible foliar injury. Since that time, very little new research has been performed on these phytotoxic effects at concentrations currently observed in the U.S. (see second draft ISA, Appendix section 3.6.2). Additionally, according to the second draft ISA, although PAN continues to persist as an important component of photochemical pollutant episodes, there is little evidence in recent years to suggest that PAN poses a significant risk to vegetation in the U.S. (see second draft ISA, Appendix section 3.3)

At the time of the 2008 NOx/SOx ISA, it was known that gaseous SO_2 caused foliar injury as well as decreased photosynthesis, growth, and yield in plants. Additionally, it was known that SO_2 caused mortality in lichens. Due to declines in SO_2 emissions, few additional studies evaluating direct gaseous effects of SO_2 have been conducted in the U.S, and these studies generally focused on recovery (see second draft ISA, Appendix section 3.2). Additionally, per the second draft ISA, there is no clear evidence of acute foliar injury below the level of the current SO_2 standard (see second draft ISA, Appendix section 3.6.1).

The 2008 NOx/SOx ISA reported that experimental exposure of HNO_3 resulted in damage to the leaf cuticle of pine and oak seedlings, which could predispose those plants to other stressors such as drought, pathogens, and other air pollutants. Since the 2008 ISA, one study (Padgett et al., 2009) investigated dry deposition of HNO_3 on the foliage, with findings that supported the earlier research. The 2008 ISA also reported several lines of evidence in lichen studies, including transplant and controlled exposure studies, indicating that HNO_3 concentrations contributed to the decline in lichen species in the Los Angeles basin. Since that time, several new studies have been published that continue to support this evidence (see second draft ISA, Appendix section 3.4).

Limited new evidence regarding direct effects of NO, NO_2, PAN, and SO_2 has become available since the 2008 NOx/SOx ISA. Additionally, there are no data and/or tools available to support a quantitative assessment of direct gaseous effects. Hence, we do not intend to conduct a quantitative assessment for these effects in the REA.

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[27] The causality statements in the second draft ISA relate to gas phase SO_2, NO, NO_2, PAN, and HNO_3 and injury to vegetation.

3.4 NON-NITROGEN AND NON-SULFUR EFFECTS OF PM

Since publication of the 2009 PM ISA, there has been some new evidence to support the understanding of ecological effects associated with PM components, particularly for metals and organics. The causality determination from the last review remains unchanged, finding that there is a likely causal relationship between PM deposition and a variety of effects on individual organisms and ecosystems. While the new studies do provide some additional evidence for community-level responses to PM deposition, the second draft ISA notes that uncertainties remain due to the difficulty in quantifying relationships between ambient concentrations of PM and ecosystem response (see second draft ISA, Chapter 1, section 1.10). Additionally, there are currently no data and/or tools available for evaluating these effects. Hence, we do not intend to conduct a quantitative assessment for these effects in the REA.

3.5 FRESHWATER ACIDIFICATION AND NITROGEN ENRICHMENT

Since the 2008 NOx/SOx ISA, new studies have been published on the effects of N and S deposition on freshwater ecosystems. This evidence includes new CLs for freshwater ecosystems and has led to the development of expanded causality determinations in the second draft ISA from the last review. These CLs provide national coverage and could significantly expand the scope for analyses since the last review, and for N enrichment, to fill a major gap from the last review. The sections that follow provide more details about the new scientific and technical information pertaining to freshwater acidification and N enrichment that are available for consideration in the REA.

3.5.1 Freshwater Acidification

While the causality determination for biogeochemistry changes remains largely unchanged,[28] the causality determination for changes in biota have been expanded in the second draft ISA, having found that the body of evidence is sufficient to infer a causal relationship between acidifying atmospheric deposition and changes in biota, including ***physiological impairment and alteration of species richness, community composition, and biodiversity*** in freshwater ecosystems (rivers, lakes, and streams).

At the time of the 2008 NOx/SOx ISA, it was known that atmospheric N deposition can alter the pools and fluxes of the C and N cycles, causing nitrification and denitrification and NO_3^- leaching to surface waters, increasing acidity. Atmospheric deposition of S directly adds SO_4^{2-} to soil leachate and surface waters, increasing acidity. The processes of acidification and

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[28] See Table 1-1 of the second draft ISA for a side-by-side comparison of causality statements from the last review and this review.

the chemical indicators that describe it were well characterized at the time of the 2008 NOx/SOx ISA, and newer studies have only further described and quantified some of these relationships (second draft ISA, Appendix section 7.1.7). Effects of N and/or S deposition on changes in biota are linked to chemical indicators in surface water, which include water pH, ANC, and the concentrations of SO_4^{2-}, dissolved inorganic aluminum (Al), and base cations. The strongest evidence for a causal relationship between acidifying deposition and aquatic biogeochemistry comes from studies of changes in surface water chemistry (second draft ISA, Chapter 1, section 1.6.1). Consistent evidence from multiple studies spanning several decades shows that in acidified waters, changes in surface water chemistry can cause the loss of acid-sensitive biological species. Biological effects are primarily attributable to low pH and elevated concentrations of inorganic Al (see second draft ISA, Appendix section 8.1). ANC integrates chemical components of acidification, and surface water acidification models calculate ANC as a proxy that relates to pH and inorganic Al concentrations. However, ANC does not directly alter the health of biota (see second draft ISA, Chapter 1, section 1.6.3).

Since the last review, there is new evidence for chemical recovery from acidification in freshwater ecosystems (see second draft ISA, Appendix section 7.1.5.1), but evidence for biological recovery has been more limited. Biological recovery in a freshwater ecosystem can occur only if chemical recovery is sufficient to allow reproduction, growth, and survival of acid-sensitive plants and animals to occur (see second draft ISA, Appendix 8, Section 8.4). Also, chemical recovery of ANC or pH may not necessarily lead to recovery of the ecosystem to its previous condition before acidification, due to changes in relationships among ANC, pH, DOC, and Al; depletion of soil base cation pools; and/or partially reversible (or irreversible) S adsorption on soils. In the 2008 NOx/SOx ISA, studies of biological recovery generally indicated that the time required for full biological recovery is uncertain and that responses in biota lag behind chemical recovery and may take decades from the onset of chemical recovery. New studies documented in the second draft ISA of multiple trophic levels continue to support these findings.

As mentioned earlier in Chapter 2, the 2009 NOx/SOx REA related ANC (as an indicator that relates well to pH) to fish species richness and species-level responses to acidification in lakes and streams in two case study areas, and calculated CLs for those areas. Since that time, new CLs using ANC as a chemical indicator for freshwater acidification have become available in the peer reviewed literature. Some of these new data are based on studies that used SMB models to calculate CLs (see second draft ISA, Appendix section 8.5.4). McDonnell et al. (2014), for example, developed a regional dataset that provides CLs for continuous coverage of streams for the southern Appalachian Mountains region from Georgia to southern Pennsylvania, and from eastern Kentucky and Tennessee to central Virginia and western North Carolina. In addition, Shaw et al. (2014) developed steady-state CLs for lakes in the Sierra Nevada Mountain, where N deposition is the stronger driver of acidification. New CLs have also been determined through the use of biogeochemical watershed models such as MAGIC and the photosynthesis evapotranspiration biogeochemical (Pnet-BGC) model (e.g., second draft ISA, Appendix section 8.5.4; Lawrence et al., 2015; Fakhraei et al., 2014; Fakhraei et al., 2016) for the Adirondacks Mountains, Appalachian Mountains, and for the Great Smoky Mountains National Park. When these CLs, as well others in the National Critical Loads Database of Sulfur and Nitrogen (NCLD v3.0)[29], are considered together, they approximate broad, national coverage since sites are represented across the contiguous United States (CONUS).

In addition to new CLs, new information has become available through the National Aquatic Resource Surveys (NARS) program.[30] Through the National Rivers and Streams Assessment (NRSA), 1.2 million miles of rivers and streams were assessed for their ecological condition and through the National Lakes Assessment (NLA), more than 1,000 lakes, ponds, and reservoirs (representing nearly 50,000 water bodies) were sampled for their water quality and biological and habitat conditions using comparable field and laboratory protocols. Although the surveys were not specifically focused on acid-base chemistry, ANC and pH were among the chosen chemical indicators used to assess biological integrity. However, the NLA and NRSA were designed to be representative of all lakes and streams, so many of the lakes and streams that were sampled are not sensitive to acidification.

The NLA and NRSA datasets are robust and comprehensive. There is potential to use these datasets to evaluate risk of acidification to lakes and streams from exposure to deposition from oxides of nitrogen, oxides of sulfur and particulate matter. However, air quality metrics were not collected as part of the NARS program which limits their use in NAAQS applications. Data from the 2007 and 2012 NLA have been incorporated into a database of lake nutrient chemistry data for the western United States (Williams and Labou, 2017). These data

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[29] Since the last review, the National Atmospheric Deposition Program (NADP) Executive Committee formed the CLs of Atmospheric Deposition Science Committee (CLAD) in April 2010. The CLAD has developed the NCLD, which is a compilation of CLs data and supporting information from many regional- and national-scale projects within the US. The focus of this database is on CL of N and S deposition and the effects on terrestrial and aquatic environments. The database is updated through periodic "calls for data" and corrections. The most recent version of the database, version 3.0, was released in October 2017. We note that studies in the NCLDv.3 are included in the second draft ISA.

[30] This program is designed to assess the status of and changes in quality of the nation's coastal waters, lakes and reservoirs, rivers and streams, and wetlands. These are statistical surveys first conducted in 2007 and the data were not available at the time of the last review. The NARS are made up of four individual surveys that are implemented on a rotating basis; National Lake Assessment (NLA), National River and Stream Assessment (NRSA), National Coastal Condition Assessment (NCCA), and National Wetland Condition Assessment (NWCA). The NLA and NRSA are discussed in this section while the NWCA and NCCA are discussed in Section 3.7.

were used by Williams et al. (2017) along with data from the 1985 EPA Western Lake Survey, a precursor to the NLA used by Nanus et al. (2017) to develop the CLs discussed below. However, no studies are currently available linking the NLA and NRSA (nor the NWCA and NCCA) to atmospheric deposition to develop CLs or exposure-response functions. Further analysis of the NARS data for purposes of this NAAQS review would require regression-based approaches, linking ambient air quality data to the biological or chemical metrics in the data to develop new CLs or exposure-response functions. Such new, deposition-based analyses are an area of interest for future research, but fall outside the scope of this REA.

Given the newly available CLs related to changes in pH and ANC, as well as the important changes in N and S deposition since the last review, we judge that an updated analysis of freshwater acidification would inform our understanding of how the associated risks have potentially changed since the last review. In addition, the availability of new datasets and improvements in assessment tools will likely reduce some of the uncertainties and limitations identified in our analysis of freshwater acidification in the last review.

3.5.2 Freshwater Nitrogen Enrichment

Nitrogen enrichment from atmospheric deposition of N to freshwater ecosystems leads to increased productivity of algae and aquatic plants, altered nutrient ratios, and sometimes decreased oxygen levels (see second draft ISA, Chapter 1, section 1.6). The previous review found the relationship to be causal for changes in biogeochemistry and it remains largely unchanged for this review.[31] However, based on new scientific evidence since the last review, the causality determination for changes in biota, which included species richness, community composition, and biodiversity, has been expanded to include ***altered growth*** as an endpoint.

As discussed in Chapter 2, the 2009 NOx/SOx REA did not include a quantitative assessment of freshwater N enrichment effects due to lack of data. For this review, several of the new studies available include CLs related to biogeochemical changes as well as altered growth, consistent with the expansion of the causality determination in the second draft ISA. Baron et al. (2011) assessed lakes in the Sierra Nevada Mountains, Rocky Mountains, and the northeastern U.S. CL values were estimated by comparing total N deposition (estimated from a combination of the Parameter-elevation Regression on Independent Slopes Model (PRISM) and NADP data for wet deposition and CMAQ modelled dry deposition) to NO_3^- concentrations in the 3 regions. The CL was estimated as the point within the data where NO_3^- concentrations begin to increase, indicating that deposition is exceeding the N uptake of the watershed and leading to N enrichment in the lake. In this study, NO_3^- concentrations were not related to a specific biological

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[31] See Table 1-1 of the second draft ISA for a side-by-side comparison of causality statements from the last review and this review.

endpoint, but N enrichment is connected to a wide range of ecological effects (see second draft ISA, Chapter 1, section 1.6). This analysis resulted in non-lake specific CL estimates of 2, 3 and 6 kg N/ha/yr for the Sierra Nevada's, Rocky Mountains and northeast, respectively.

Two additional studies used regression models to link N deposition with changes in water chemistry that can then provide some insight into potential ecological effects. Williams et al. (2017) assessed lakes in the Georeferenced Lake Nutrient Chemistry (GLNC) database (Williams and Labou, 2017). The Lakes included in the study were located at elevations above 1200 meters and on federally protected lands (National Park Service (NPS) and U.S. Forest Service (USFS). The CL was estimated to be a point at which there was a fundamental change in the lake biogeochemical cycle and used a logistic regression approach to relate total N deposition to the NO_3^- concentration above which there is > 50% chance of the phytoplankton growth in the lake having shifted from N to P nutrient limitation. This resulted in a CL value of 4.1 kg N/ha/yr (with a range of 2.8 – 5.2 kg N/ha/yr), and a lower estimate of 2 kg N/ha/yr to protect against false positive (lakes where deposition does not exceed the CL, but lake chemistry data indicate there has been a shift to P limitation).

Nanus et al. (2017) determined CLs for lakes in the greater Yellowstone area (including 2 National Parks - Yellowstone and Grand Teton) which refined CLs estimates from earlier work by Nanus et al. (2012) for the broader Rocky Mountains area. CLs were estimated using a multi-linear regression approach to predict surface water NO3- concentrations, using basin characteristics and TN deposition. CLs ranged from <1.5 + 1.0 kg N/ha/yr to >4 + 1.0 kg N/ha/yr. These values were based on a NO3- threshold of 1.0 µmol/L based on growth characteristics of indicator diatom species, which are indicative of shifts from oligotrophic to mesotrophic or eutrophic conditions (Saros et al., 2005). CLs for NO3- thresholds of 0.5 µmol/L, 1.6 µmol/L and 2.0 µmol/L were also reported.

The availability of new CLs in this review for freshwater N enrichment potentially fills a data gap from the last review and informs an assessment of how changes in deposition could impact changes in water chemistry. We also note that the availability of the new data through the NARS program could possibly be used in this assessment to identify, and prioritize for further assessment, impacted lakes and streams. At this time, however, we are unsure whether a new quantitative assessment of freshwater N enrichment will be included in the REA since there is uncertainty as to whether it would provide additional insight into potential ecological effects, be broadly applicable in their application, and/or improve our understanding of pollutant-attributable risks since the last review, and as such, substantively inform our ability to assess national standards for freshwater N enrichment.

3.6 TERRESTRIAL ACIDIFICATION AND NITROGEN ENRICHMENT

In terrestrial ecosystems, the acidifying effects from N and S, and the enrichment effects from N can occur simultaneously. However, the strength of one process over the other (i.e., acidification versus N enrichment) can vary depending upon several factors, including soil pH and buffering capacity, the presence and abundance of sensitive species, and the degree of nitrogen limitation on primary production.

Since the 2008 NOx/SOx ISA, more recent research has confirmed and strengthened our understanding of terrestrial acidification (see second draft ISA, Appendix section 5.7). While the causal determinations for changes in biogeochemistry have generally remained unchanged for the second draft ISA, the causal determinations for changes in biota has been expanded for this review to include ***alteration of species richness, community composition, and biodiversity*** as endpoints. Additionally, the second draft ISA includes a new causality determination, finding that there is a causal relationship between atmospheric N and S deposition and the ***alteration of the physiology and growth*** of terrestrial organisms, as well as the ***productivity*** of terrestrial ecosystems.

Similar to acidification, the causality determination for biogeochemical changes from N enrichment has remained largely unchanged since the last review.[32] However, the largest increase in scientific evidence, over any other effect category, is for terrestrial N enrichment ecological effects (see second draft ISA, Chapter 1, section 1.2.3). This new evidence has confirmed and strengthened our understanding of the mechanistic links that inform causal determinations between atmospheric N deposition, biogeochemistry, and biota in terrestrial ecosystems. The second draft ISA includes a new causal determination for N enrichment, having determined that there is a causal relationship between atmospheric N deposition and the ***alteration of the physiology and growth*** of terrestrial organisms and the ***productivity*** of terrestrial ecosystems. The new data also improves our ability to quantify dose response relationships between N deposition and ecological response. New studies include investigations of plant and microbial physiology, long-term ecosystem-scale N addition experiments, regional and continental-scale monitoring studies, and syntheses. Additionally, since the last review, new research techniques have been developed to understand community composition, a larger number of communities have been surveyed, and new regional and continental-scale studies been conducted (see second draft ISA, Chapter 1, section 1.2.3).

The sections that follow provide more details about the new scientific and technical information available for consideration in the REA.

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[32] See Table 1-1 of the second draft ISA for a side-by-side comparison of causality statements from the last review and this review.

3.6.1 Forests

New studies since the 2008 NOx/SOx ISA indicate that terrestrial acidification is an ongoing and widespread issue in areas of elevated acidic deposition. For example, new studies have observed the ongoing depletion of exchangeable base cations in forest soils of the northeastern U.S. (see second draft ISA, Appendix section 4.3.4). There is considerable evidence that N and/or S-driven acidification in forests likely contributes to the mobilization of toxic forms of Al and decreases the availability of nutrients to plants due to leaching of base cations from soil and interference with uptake (see second draft ISA, section 5.2.1). These soil changes can affect tree regeneration (e.g., Sullivan et al., 2013 sugar maple study; Lieb et al., 2011).

Since the 2008 NOx/SOx ISA, there is new evidence of species-specific effects of atmospheric N deposition on tree growth and mortality in the U.S. (e.g., Thomas et al., 2010; Dietze and Moorcroft et al., 2011); but it is uncertain whether changes in growth or mortality are driven by N-enrichment or terrestrial acidification effects. Additionally, Simkin et. al (2016), suggests that species richness of forest understory plant communities generally has unimodal relationships with atmospheric N deposition. This unimodal relationship was found to be pH-dependent, with Simkin et al. (2016) reporting that plant species richness was more likely to decline with increasing atmospheric N deposition if the herb community occurred on more acidic soils.

The 2009 NOx/SOx REA calculated CLs for soil acidification in two forest ecosystems using the SMB model and estimates of base cation weathering from the clay substrate method. Since that document was completed, new CLs have become available from studies that used the SMB model with the clay substrate method for estimating base cation weathering (BCw) rates, and relied on a Bc/Al ratio of either 1 or 10, based on the precedent for use of these numbers by the scientific community.[33] (For a summary of these studies, see second draft ISA, Appendix section 5.5.3).

Most new studies explicitly examining acidification from N and S since the last review have built upon earlier approaches that modeled soil acidification CLs using one of several different models (i.e., SMB, STA, MAGIC, and ForSAFE-VEG)[34],which are summarized in the second draft of the ISA (see second draft ISA, Appendix sections 4.5 and 4.6). In addition, there are several studies (whose CLs are included in the NCLD v3.0) that have evaluated the use of other models besides the clay substrate method for purposes of estimating BCw rates. For

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[33] Bc/Al ratios of 1 and 10 have been used as average numbers to represent different forest types in different parts of the U.S. (e.g., Eastern versus Western, or coniferous versus deciduous forests).

[34] For a summary of these models and related considerations, see second draft ISA, Appendix section 4.5.

example, because of the known limitations of the clay substrate method,[35] Phelan et al. (2014) evaluated the use of the dynamic model, PROFILE, for estimating BCw. Phelan et al. (2014) paired PROFILE with national datasets[36] as a method to estimate BCw rates for forests in the U.S., applying it to 51 forested sites across Pennsylvania.

In the 2008 NOx/SOx ISA, there was consistent evidence that N additions stimulated forest productivity. However, responses included both neutral and negative effects of N additions on tree growth (second draft ISA, Appendix section 6.3.2.1). Since the 2008 NOx/SOx ISA, there is considerable new evidence from deposition gradient studies, forest modeling, and long-term N addition experiments that atmospheric N deposition broadly stimulates tree growth and the productivity of forested ecosystems (see second draft ISA, Appendix section 6.3.2.1); however, the effects can vary by species. For example, studies have shown that conifer species, particularly at high elevations, are more likely to exhibit negative growth responses or mortality in response to added N (second draft ISA, Appendix section 6.2.3.1). One particular study, Thomas et al (2010) used exposure-response functions to relate N deposition to growth and mortality impacts on 23 individual tree species in the Northeast and mid-Atlantic.[37] Leveraging forest inventory data from the early 1980s through the mid-1990s, Thomas et al. (2010) reported declines in growth for three conifer species, consistent with other observational studies (see second draft ISA, section 6.2.3.1). The study also reported higher mortality rates for 8 tree species with increasing atmospheric N deposition while only three species showed a positive relationship. Overall, this analysis was consistent with other multi-variate analyses that explored the key drivers of tree mortality in the region (e.g., Dietze and Moorcroft, 2011).

3.6.2 Lichens and Mycorrhizal Fungi

Since the 2008 NOx/SOx ISA, there is new evidence of decreases in lichen species richness as the result of atmospheric N deposition in the U.S., and there are now ambient observations that atmospheric N deposition in the U.S. is changing mycorrhizal community composition. (see second draft ISA, Appendix section 6.6.2).

As mentioned in Chapter 2 of this document, the 2009 NOx/SOx REA included a qualitative assessment of N enrichment effects through a case study for coastal sage scrub and mixed conifer forests. No quantitative assessment was conducted due to lack of data. Since the

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[35] See section 2.5 for a summary of these limitations.

[36] See Table 2 in Phelan et. al., 2014, for the list of national datasets.

[37] We anticipate that this study will be expanded to include 94 species nationally, relating growth and mortality impacts to N and S deposition (Horn et al., 2018 in review).

last review, an extensive amount of new data with national coverage is available for terrestrial N enrichment CLs for lichens and mycorrhizal fungi.[38]

New lichen CLs and response curves are available through two studies conducted in the Pacific Northwest, for the portions of the North American West Coast Marine Ecoregion in Oregon and Washington (Geiser et al., 2010) and for the portions of the Northwestern Forested Mountains Ecoregion in Northern California, Oregon, Washington, and parts of northern Idaho and Montana (Root et al., 2015). For the lichens dataset, researchers collected information on two responses - lichen community composition and lichen N concentrations - and related those responses to atmospheric concentrations of N (Root et al., 2015) and/or measures of atmospheric deposition (Geiser et al., 2010). The resulting equations constitute exposure-response curves and were used to derive CLs for lichen community composition and lichen N concentrations. These data have been extrapolated for application to the CONUS for the NCLDv3.0.

The mycorrhizal fungi dataset is a collection of empirical CLs for total atmospheric N deposition from various studies included in Pardo et al. (2011). This dataset includes minimum and maximum CLs for community composition.

3.6.3 Herbs and Shrubs

A national CLs dataset is available for the species richness of herbs and shrubs based on the Simkin et al. (2016) study. As mentioned earlier, the Simkin et al. (2016) study analyzed relationships between plant species richness and atmospheric N deposition involving interactions with soil pH (along with precipitation and temperature). Therefore, the dataset relates to both N enrichment and acidification effects. The dataset covers nearly 4,000 herb/shrub species over more than 15,000 plots. This dataset includes a collection of total atmospheric N deposition CLs from Simkin et al. (2016), with CLs calculated separately for “open canopy” (grasslands, shrub lands, and woodlands) and “closed canopy” (forested understory plants) ecosystems.

The CLs in Simkin et al. (2016) were derived using regression analyses relating atmospheric N deposition to species richness. These underlying regression models are equations that constitute an exposure-response relationship (i.e., for both open and closed canopy systems, species richness is related to atmospheric N deposition, soil pH, temperature, and precipitation; see Table 1 in Simkin et al. (2016)). These equations can be used to evaluate potential changes in effects based on varying levels of atmospheric deposition.

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[38] For descriptions of these data, see NADP CLAD report “2017 Summary of Critical Load Maps,” http://nadp.slh.wisc.edu/committees/clad/db/NCLDMapSummary_2016.pdf

3.6.4 Summary

For this review, new or refined national CLs as well as new national exposure response curves are available for various terrestrial ecosystems and receptors, which will expand and enhance our ability to evaluate sensitivity and risk for acidification and N enrichment nationally for multiple species. This new information also represents many of the new endpoints that have contributed to the stronger weight of evidence and expanded causality determinations in the second draft ISA, enabling an evaluation of growth and mortality effects as well as species or community composition changes that was not available at the time of the last review. Additionally, several studies have produced refined CLs and/or refined methods for deriving CLs, which may reduce identified uncertainties from the last review. Given the extent to which new scientific and technical information are expected to fill the data gaps/limitations from the last review and add to our understanding of pollutant-driven risk and exposure, we judge that quantitative assessments evaluating terrestrial acidification and N enrichment effects will provide insight into potential ecological effects and improve our understanding of pollutant-attributable risks since the last review.

3.7 OTHER NITROGEN AND SULFUR EFFECTS

3.7.1 Estuarine Nitrogen Enrichment

The causality determination for atmospheric N deposition and biogeochemical changes in estuarine and near coastal marine ecosystems remains unchanged, with new evidence continuing to support the findings of a causal relationship in the last review. However, since the last review, new paleontological studies, observational studies, and experiments have further characterized the effects of N on phytoplankton growth and community dynamics, macroinvertebrate response, and other indices of biodiversity. Hence, the causality determination pertaining to atmospheric N deposition and alteration of species richness, species composition and biodiversity has been expanded to include ***altered growth, total primary production,*** and ***total algal community biomass*** as endpoints.

The 2009 NOx/SOx REA included a case study for the Potomac River and Potomac Estuary in the Chesapeake Bay and the Neuse River and Neuse River Estuary in Pamlico Sound, using ASSETS EI and the SPARROW model. The 2009 NOx/SOx REA noted uncertainties in the inputs and outputs of the SPARROW model, and noted the importance in choosing case study areas where atmospheric deposition plays a large role in N loading to an estuary. Since the last review, several modeling studies have estimated the amount and proportion of current and future N loading expected to result from atmospheric deposition (see second draft ISA, Table 7-9).

At the time of the last review, CLs were not available. Several new CLs pertaining to salt marsh community structure, microbial activity, biogeochemistry, and loss of eel grass are now available from Greaver et al. (2011) based on a review of three studies (Caffrey et al., 2007; Wigand et al., 2003; and Latimer and Rego, 2010). There are three new studies with threshold values for eel grasses in estuaries now available (see second draft ISA, Appendix section 10.2.5, Table 10.4). These new studies are based on total N loading to the estuary. To assess exceedances of CLs (or threshold levels) related to atmospheric N deposition, watershed-level estimates of total N loading would need to be calculated from modelling or other watershed-level estimates.

While new studies are now available to determine which estuaries are predominantly affected by atmospheric deposition, we note that any modelling to evaluate impacts of N deposition on ecological effects in an estuary would need to be conducted on an estuary-by-estuary basis. Additionally, the CLs are limited in geographic scope, with studies conducted primarily in New England, so would be of limited use in assessing national standards.

New data are also available through the NCCA which provide information on overall estuarine conditions, but as described in section 3.5.1, these conditions are not specifically related to atmospheric deposition, but rather to total nutrient loading. This, and the lack of representativeness of any individual estuarine modelling approach, limits the usefulness of conducting a quantitative assessment. Given these limitations, such information would be of limited usefulness in informing decisions on national-scale standards, therefore we do not intend to conduct quantitative assessments for this effect category.

3.7.2 Wetlands Nitrogen Enrichment

The causality determination for atmospheric N deposition and biogeochemical changes in wetland ecosystems remains unchanged, with new evidence continuing to support the findings in the last review. New research on wetland biogeochemistry since 2008, includes a synthesis of wetland improvements to water quality through denitrification and biological uptake, a meta-analysis of N addition effects on methane (CH_4) and nitrous oxide fluxes, and multiple observations of changes in belowground C cycling in response to added N. These biogeochemical shifts may diminish the wetland ecosystem services of long-term carbon storage and flood protection, as well as reduce the stability and persistence of wetlands on the landscape. Nitrogen loading effects upon productivity are uneven across species, which may affect wetland biodiversity and the wetland ecosystem service of provisioning.

New evidence published from observational studies, experimental N addition studies in the field and in mesocosms, and re-analysis of large data sets supports and extends the conclusions of the 2008 NOx/SOx ISA. Additionally, since the last review, there is newly published evidence of atmospheric N deposition effects on endpoints not covered in the 2008 ISA, including alterations to plant physiology and plant architecture (see second draft ISA, Appendix section 11.5). Given this newly available information, the causality determination for atmospheric N deposition and alteration of species richness, community composition, and biodiversity has been expanded to include alteration of ***growth and productivity*** as well as ***species physiology*** as endpoints.

The 2009 NOx/SOx REA did not include a quantitative assessment for wetlands due to lack of available data. Since the last review, new CLs have become available for ombrotrophic bogs and the purple pitcher plant (*Sarracenia purpurea* L.) (Greaver et al., 2011). The CLs in Greaver et al. (2011) were based on 6 studies (two studies of purple pitcher plant, and four studies related to net primary production and peat accumulation in ombrotrophic bogs). One additional study, Crumley et al. (2016), found threshold levels of deposition for purple pitcher plant consistent with the CLs estimated by Greaver et al. (2011).

To determine the utility of these CLs for purposes of the REA, we considered the geographic coverage, representativeness and uncertainty tied to these data. We have determined that application of these CLs has the highest level of certainty at the individual sites included in the 7 studies. However, only four of the studies (Aldous et al., 2002; Gotelli and Ellison, 2002, 2006; Crumley et al., 2016) included sites within the United States (the remaining studies included only sites in Canada). This limits the geographic scope of potential analyses to 15 study sites located in the northeastern U.S. The geographic scope of the CLs estimates could be expanded by aggregating to Ecoregions, similar to other CLs estimated in Pardo et al. (2011), which would include some of the study sites located in Canada. However, this would only expand the coverage to include Ecoregions in the northeast and north central U.S. while increasing the uncertainty associated with the CLs as values are extrapolated from individual points to larger areas.

Additionally, data is available through the NWCA. The overall scope of the NARS datasets is described in section 3.5. Similar to the other NARS datasets, the NWCA did not include measurements of ambient air quality or atmospheric deposition. While new CLs are available, given the level of uncertainty and limited geographic representation of these studies and the reasons discussed in section 3.5.1 regarding the NARS data, we do not intend to conduct a quantitative assessment for this effect category.

3.7.3 Coastal Acidification

The 2008 NOx/SOx ISA did not address nutrient enhanced coastal acidification. Since the 2008 ISA, N enrichment has been recognized as a possible contributing factor to increasing acidification of marine environments. Specifically, N has been recognized as a possible contributing factor to coastal acidification because the CO_2 produced by organic matter decomposition in eutrophic waters can contribute CO_2 to the water column along with the dissolution of atmospheric anthropogenic CO_2, decreasing the pH (see second draft ISA, Appendix section 10.5). Given the new scientific information available supporting this effect, the second draft ISA found that the relationship between atmospheric N deposition and ***increased nutrient-enhanced coastal acidification*** is likely causal. Additionally, the second draft ISA found that the evidence is suggestive of, but not sufficient to infer, a causal relationship between atmospheric N deposition and changes in biota including ***altered physiology, species richness, community composition, and biodiversity*** due to nutrient enhanced coastal acidification. Despite newly available scientific evidence for this effect category, there are currently no tools or data available to quantitatively assess the risks due to coastal acidification that are associated with atmospheric N and/or S deposition. Because of this, no quantitative analyses are supported for this REA.

3.7.4 S Enrichment in Freshwater and Wetland Ecosystems

3.7.4.1 Mercury Methylation

Recent research since the last review continues to strengthen and inform our scientific understanding of the relationship between atmospheric S deposition and freshwater MeHg production. This understanding has expanded since the last review to include the identification of: 1) additional types of organisms that play a role in the methylation process, 2) additional macro- and micro-environments in which methylating organisms are found, and 3) additional areas within the U.S. containing habitats with conditions suitable for methylation. Building on the body of available science, including that available in 2008, the second draft ISA finds a causal relationship between S deposition and ***the alteration of mercury (Hg) methylation*** in surface waters, sediment, and soils in wetland and freshwater ecosystems (see second draft ISA, Appendix section 12.1).

Specifically, this recent research has demonstrated that in addition to sulfur-reducing bacteria (SRB), certain strains of archaea found in wetland sediments are also active in S reduction. The current review therefore uses the broader term of sulfur-reducing prokaryotes (SRP), when appropriate, to reflect the joint role of both groups of organisms in certain wetland environments (see second draft ISA, Appendix sections 12.1 and 12.3). Further, additional organisms that possess the ability to methylate mercury have also been identified, due in part to the discovery of the genes associated with this ability (see second draft ISA, Appendix section 12.3). New evidence has broadened our understanding of where methylation occurs, both in terms of types of wetland and freshwater ecosystems, as well as the specific areas within these ecosystems where methylation is most likely to occur. For example, it is now known that methylation occurs in agricultural wetlands, rivers and streams and that within aquatic ecosystems, periphyton can play an important role as a host for SRPs. Furthermore, the microbial communities embedded within the periphyton can be quite complex and diverse, are more efficient in the methylation of Hg than are SRPs in sediments, and boost Hg methylation rates within the oxygenated water column of freshwater and wetland ecosystems (see second draft ISA, Appendix section 12.3).

These scientific advances confirm and highlight the case, as described in the second draft ISA (second draft ISA, Appendix section 12.3.3) and mentioned in Chapter 2 of this document, that the relationship between atmospheric S deposition and measured increases in MeHg in associated wetland and freshwater aquatic systems and biota is complex, in part because it is mediated by the activity of SRP which are influenced by multiple interacting physical, chemical and biological variables such as oxygen content, temperature, pH, and labile carbon supply. There are several controlling factors that can influence the rates of mercury methylation (see second draft ISA, Appendix 12, Figure 12-9). Given the complexity, temporal and seasonal variability, and multiple drivers of the MeHg process across the national landscape, no dose-response functions have been established and there is currently a lack of CLs and assessment tools available by which to assess the risks of MeHg enhancement from atmospheric deposition of S in North American ecosystems (see second draft ISA, Appendix 12 sections 12.6 and 12.3.3).

Based on the information presented in the second draft ISA and summarized above, we note that there are remaining uncertainties associated with the linkages connecting atmospheric S deposition to aquatic SO_4^{2-} concentrations to MeHg production in wetland and freshwater aquatic ecosystems. Additionally, there are no known tools or data available for quantitative assessments. Therefore, we do not intend to conduct a quantitative assessment for this effect.

3.7.4.2 Sulfide Phytotoxicity

The 2008 NOx/SOx ISA only included information regarding sulfide phytotoxicity in European systems, and in mesocosm studies that showed sulfide toxicity reduced biomass of wetland plants and aquatic macrophytes under exposure levels higher than those that occur in U.S. regions with high atmospheric S deposition. Newer research, however, shows sulfide toxicity occurring in the field within multiple wetland ecosystems under ambient exposure conditions in North America (see second draft ISA, Appendix sections 12.2.3 and 12.7.3). This new information is sufficient to support a new causal determination in the second draft ISA, finding that the body of evidence is sufficient to infer a 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. Sulfide interferes with nutrient uptake in plant roots and studies show elevated sulfide can result in decreased seed mass, seed viability, seedling emergence rates, decreased seedling height, decreased seedling survival rates, and reductions in total plant cover, all which can lead to shifts in plant community composition (see second draft ISA, Appendix section 12.2.3).

The relationship between atmospheric S deposition and sulfide toxicity is complex and influenced by multiple factors. Since sulfide is the product of microbial SO_4^{2-} reduction, its concentration in water (including sediment pore water) is heavily dependent upon environmental factors that influence microbial activity, particularly the availability of dissolved organic carbon (DOC). In addition, the degree to which aquatic vegetation is exposed to phytotoxic concentrations of sulfide depends on its residence time as a free ion in water. When sulfide binds with iron it precipitates out of solution and is no longer available to plants. Thus, the phytotoxicity of sulfide is regulated in part by the availability of iron in the wetland or freshwater system.

The largest uncertainty that remains regarding sulfide effects, as with methymercury discussed in 3.7.4.1. above, is that associated with the linkages connecting atmospheric S deposition to aquatic sulfate concentrations in wetland and freshwater aquatic ecosystems. We are currently unable to characterize how deposition amounts or changes in deposition affect aquatic concentrations of SO_4^{2-}. Because of this fundamental uncertainty, we do not intend to conduct a quantitative assessment for this effect.

3.8 ECOSYSTEM SERVICES

There are several ways in which ecological effects can be related to public welfare. The most comprehensive model is that of ecosystem services, which can provide information on the linkages between changes in ecological effects and known or anticipated effects to public welfare. Ecosystem services can be generally defined as the benefits that individuals and organizations obtain from ecosystems. The EPA has defined ecological goods and services as the outputs of ecological functions or processes that directly or indirectly contribute to social welfare or have the potential to do so in the future. Conceptually, changes in ecosystem services may be used to aid in characterizing a known or anticipated adverse effect on public welfare. Ecosystem services, those related to Class I areas and endangered species, and analyses of both non-use and use values across larger areas and in the human economy, including commercial uses, can be used separately or together to help inform public welfare decisions.

Appendix 14 of the second draft ISA includes summaries of several new papers published since the 2008 NOx/SOx ISA that connect the effects of N and S deposition to ecosystem services. Some of these studies present valuation of costs and benefits of nitrogen loading across the nation for several ecosystem services such as coastal fish harvests, recreational uses of waterbodies, and lakefront hedonic values. The second draft ISA also includes descriptions of five papers that resulted from a joint workshop between the NPS, EPA, the USFS, and members of academia. The papers present a concise conceptual model linking CLs to final ecosystem services and a framework for assessing the scientific strength of those linkages (Bell et al., 2017; Clark et al., 2017; Irvine et al.; 2017, O'Dea et al., 2017; Rhodes et al., 2017). The second draft ISA also presents an evaluation of studies conducted in Europe and a global-scale analysis of N cycling and impacts on ecosystem services. Additionally, the second draft ISA includes profiles of several threatened or endangered species and their related ecosystem services.

Since the prior review, the NPS has published a list of threatened or endangered species in each of the National Parks.[39] A national study has also been conducted, which identifies 78 threatened and endangered species for which N deposition is a contributing stressor (Hernández et al., 2016). Many of these species are located in National Parks. The second draft ISA includes case studies for several National Parks (including Rocky Mountain, Acadia, Great Smoky Mountains, Sequoia and Kings Canyon National Parks), where new scientific evidence for deposition-related effects is highlighted.

A separate literature search, from 2009 to 2017, and review for ecosystem services was conducted for this REA plan to identify papers related to ecosystem services analysis that were not included in the second draft ISA because they do not always directly relate nitrogen and or sulfur deposition to changes in ecosystem services. The papers can be loosely binned into categories including: freshwater acidification, freshwater nitrogen enrichment, coastal/estuary, coral reefs, wetlands, forests and wilderness, grasslands and deserts, fires, lichens, and biodiversity. A list of these papers is included in the Appendix. These papers provide linkages from deposition to ecological effects and finally to ecosystem services, and provide methods that either describe the potential qualitative risk to the services covered or possible avenues for quantitative evaluation of the changes in services related to N and/or S deposition. Since the last review, information in the recent literature identified by the search increases the number of ecosystem services and ecosystems potentially available for analysis and/or expands and improves previously available methods.

In addition to the results of the literature search new information is available from updated databases such as the Recreational Values Database,[40] government reports such as the

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[39] See https://irma.nps.gov/NPSpecies/

[40] See http://recvaluation.forestry.oregonstate.edu/

Internet Research Information Series [41](IRIS) reports from the USFS, and new analyses done by EPA's National Center for Environmental Economics for the Chesapeake Bay Program.[42] These databases and reports expand and improve upon analyses included in the previous review which could be updated for the current review.

There are a limited number of papers and methods available that provide national-scale estimates of ecosystem services affected by atmospheric deposition. The majority of relevant work has been place-based studies that are useful for case studies although there are some few that could be amenable to a benefit transfer approach to applying the study results to larger geographic areas. Given the limited geographical scope of the available information, as well as the inherent uncertainties associated with quantifying specific ecosystem service effects using the available methods, we do not intend to conduct any specific quantitative analyses of ecosystem services in the REA. Instead, we plan to use the information in the available literature and databases to describe how the public values specific ecosystem services and to link that information qualitatively to the policy relevance of the assessed changes in ecological risks. Availability of this type of information can also help prioritize the selection of case study areas for quantitative assessment (see section 4.4 for more information).

3.9 CONCLUSIONS

The discussion above reflects the EPA staff assessment of the degree to which currently available information, including newly available information since the last review (e.g., as summarized in the second draft ISA), might be expected to appreciably change our understanding of risk and exposures beyond the insights gained from the assessments from the last review. A critical consideration is the extent to which use of newly available information or approaches in new or updated quantitative assessments would provide risk and exposure estimates that are appreciably different or have the potential to reduce uncertainty or limitations in the previous review, and that indicates that a new assessment of risk and exposure is warranted to provide an adequate characterization of ecological risk and exposure, particularly with regard to the current standards.[43]

Based on these considerations, we have concluded that an REA is warranted to quantitatively evaluate acidification and nitrogen enrichment effects within freshwater and

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[41] See https://www.srs.fs.usda.gov/trends/iris/recstats.html

[42] See https://www.epa.gov/environmental-economics/research-environmental-economics-ncee-working-paper-series

[43] In considering this point, the EPA staff additionally recognize that such a characterization of risk and exposure, in addition to the currently available evidence, will be considered in the PA in terms of both evidence-based considerations and risk/exposure-based considerations.

terrestrial ecosystems. Given the lack of information and/or concerns about limitations or uncertainties as described in the sections above, we do not intend to quantitatively evaluate the other effect categories discussed in this chapter. More specifically, we conclude that it is appropriate to focus the REA on the following analyses, for which important new information is available in this review:

- **Air Quality Information.** Since the previous review, advances in scientific methods and the changing levels and spatial distribution of N and S deposition have appreciably changed our understanding of the linkages between ambient air concentrations, atmospheric deposition, and ecosystem exposures. New techniques are now available for combining these measurements and modeling outputs to estimate total deposition with lower uncertainty. The spatial variability and distribution of deposition has changed in recent years, as reduced forms of nitrogen deposition are now the largest source of N deposition in many places. Finally, because concentrations and deposition of oxidized N and sulfur have declined in recent years, there are new opportunities to better assess the linkage between a change in concentration and a change in deposition, while also better quantifying the uncertainties.
- **Freshwater Acidification and N Enrichment.** Since the 2008 NOx/SOx ISA, new studies have been published on the effects of N and S deposition on freshwater ecosystems. This evidence includes new CLs for freshwater ecosystems and has created a greater weight of evidence and led to the development of expanded causality determinations in the second draft ISA from the previous review. These CLs provide national coverage and are expected to significantly expand the scope for analyses since the last review, and for N enrichment, to fill a major gap from the last review. Therefore, we intend to conduct quantitative assessments evaluating freshwater acidification and N enrichment effects using CLs.
- **Terrestrial Acidification and Nitrogen Enrichment.** A substantial body of new information is available for terrestrial acidification and N enrichment for this review. New information pertaining to acidification and N enrichment of terrestrial ecosystems includes studies that evaluate endpoints (herb/shrub and mycorrhizal community composition, lichen species richness, and tree growth and mortality) through CLs and/or response curves that were not available at the time of the last review. Many of these CLs and response curves are national in scope and therefore expected to significantly expand and enhance our ability to evaluate sensitivity and risk for acidification and N enrichment. The availability of this information fills a gap in the last review. Additionally, several new studies are now available that evaluate potential methods for deriving CLs and in particular, for estimating BCw rates in soils. These studies are expected to inform updated CLs assessment in the REA, potentially reducing uncertainties from the prior review. Therefore, we intend to conduct quantitative assessments evaluating terrestrial acidification and N enrichment effects in this review.

In summary, a new REA that utilizes the new information and approaches summarized above to provide a more precise characterization of exposure and risks, and one with reduced uncertainty, would inform the current review. Therefore, we conclude that it is appropriate to develop a risk and exposure assessment, based on the newly available air quality and ecological information, to inform the current review. More information regarding our plans for analyses is included in Chapter 4.

4 PLAN FOR QUANTITATIVE ASSESSMENT

This Chapter describes the proposed quantitative analysis and approach for the REA to characterize ecological risk and exposure associated with NO_x, SO_x, and PM for current air quality conditions, as well as for when air quality is just meeting the current standards, and if appropriate, when meeting potential alternative air quality standards. The REA will focus particularly on the contribution of these pollutants to deposition-driven ecological effects on freshwater and terrestrial ecosystems based on the conclusions in Chapter 3. These ecosystems warrant further assessment to evaluate information newly available since the last review that might be expected to appreciably change our understanding of risk and exposures or have the potential to reduce uncertainty or limitations in the previous review. Given the lack of information and/or concerns about limitations or uncertainties, we do not intend to quantitatively evaluate the other effect categories described in Chapter 3.

4.1 ANALYTICAL FRAMEWORK

The proposed analysis approach for the REA is shown in Figure 4-1 and discussed in more detail below. In general, we intend to use air quality data from monitors and chemical transport models to estimate atmospheric deposition of N and S associated with ambient concentrations of NO_x, SO_x, and PM. This information will also be used to estimate deposition response factors that predict the relationship between ambient concentrations and atmospheric deposition in certain areas of the country and can be used to adjust deposition to represent just meeting the current standards, as well as potential alternative standards, as appropriate. This air quality information can then be used to assess ecological effects at a national scale for current conditions, as well as within selected case study areas for air quality conditions that just meet current and potential alternative standards. Assessment of these effects for freshwater and terrestrial ecosystems can generally be grouped into two main categories: (1) assessment of exceedances using CLs; and (2) assessment of changes in biological and chemical responses using exposure-response curves. In these analyses, CLs and exposure-response curves would be used to relate N and S deposition to changes in biogeochemistry and changes in species-level or community-level ecological or biological responses. The resulting information would then be used in the consideration of the overall ecological risk for the review. Included below is a brief discussion of some of the main components of the analytical framework, including air quality and exposure, CLs, and exposure-response curves, as well as the general application of these components in the REA. More specific information about the assessments for the REA is included in the sections that follow.

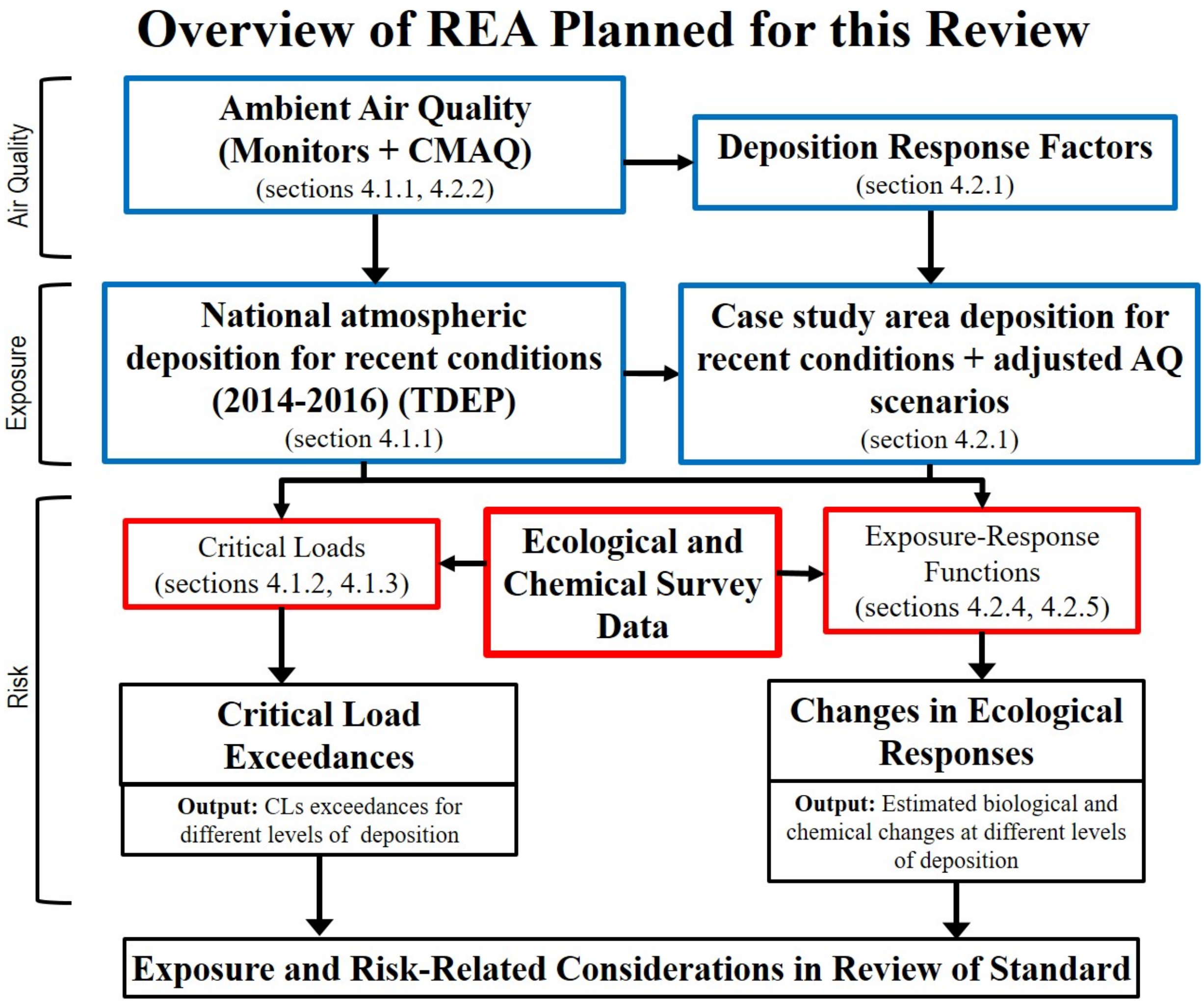

Figure 4-1. Analytical Framework for the REA

4.1.1 Ambient Air Quality and Atmospheric Deposition

The objective for the REA is to characterize risk and exposure associated with just meeting the current standards, as well as any potential alternative standards under consideration. For this review, we plan to consider both the secondary and primary standards as part of the suite of current standards. In the REA, we plan to include a national-scale assessment of ecological risk that will be based on estimates of current air quality concentrations and deposition in the U.S. Applying the same methodology as used to create the TDEP datasets (Schwede and Lear, 2014), recent air quality information would be used to create a national-scale gridded surface of N and S deposition, with separate estimates of the reduced and oxidized nitrogen contributions to total N deposition.[44] This surface will represent average deposition over three years to incorporate emissions and meteorological variations across recent years, with the individual years also available for use. Because much of the U.S. is already meeting the secondary and primary standards for NO_2, SO_2 and PM, this surface can be used to assess ecological risk in most areas for air quality conditions that are at or below the current standards. To further assess ecological effects for air quality conditions under different air quality scenarios, we propose to adjust air quality and create new gridded surfaces of N and S deposition for each of the study areas that reflect deposition levels when just meeting the current standards, and any potential alternative standards, as appropriate. The focus on air quality adjustments in smaller areas of the country will help focus the analyses on specific air quality conditions and reduce uncertainty in the adjustment methodology being applied.

4.1.2 Critical Loads and Exceedances

The second draft ISA uses the CL concept as an organizing principle to relate atmospheric deposition to ecological endpoints that indicate impairment (see second draft ISA, Section 1.2.2.3). “A critical load is formally defined as a quantitative estimate of exposure to one or more pollutants below which significant harmful effects on specified sensitive elements of the environment do not occur according to present knowledge” (Nilsson and Grennfelt, 1988, UNECE, 2004). CL estimates reflect the current state of knowledge and the selected indicators and responses (see second draft ISA, section 1.2.2.3). It is important to recognize that there is no single “definitive” CL for an ecological effect, nor is there a single definition of “harm” across CLs datasets. Given the heterogeneity of ecosystems affected by N and S deposition, published CL values for locations in the U.S. vary depending on both biological and physical factors. In fact, it is not uncommon for there to be multiple CLs available for a given pollutant at a single location due to the nested sequence of disturbances, receptors, and biological indicators considered for a given pollutant. (see second draft ISA, Chapter 1, section 1.2.2.3).

CLs are point estimates of when harmful effects begin to occur. Given the breadth of CL data available through new studies, we intend to use the CLs to provide a national-scale picture of sensitivity[45] and risk[46] (when used in conjunction with atmospheric deposition estimates to calculate exceedances). The CLs can also be useful in identifying risks of concern under different levels of atmospheric deposition and at different spatial scales. Therefore, we intend to use CLs and exceedances as tools for identifying potential case study areas for quantitative

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[44] This information allows for better understanding of the portion of deposition that is controllable under the CAA.

[44] Sensitivity is as the degree to which an ecosystem is affected by atmospheric N and/or S deposition.

[45] Risk is defined as the potential that adverse ecological effects may occur, or are occurring, as a result of exposure to one or more stressors

assessments, including designated areas that are considered important to the public (e.g., Class I areas).

To calculate exceedances of CLs in the REA, we plan to compare CLs to the estimated levels of deposition in the national-scale gridded surface for current air quality. Put simply, this means: Dep – CL = X, if X >0, then it is exceeding. This concept will be applied to the national-scale surface of gridded deposition estimates. While this is a straight-forward comparison, further work is required to understand the potential effect of an estimated exceedance. Additional considerations may also include estimates of the contribution of N and S in the system from air deposition, estimates of the time scale of the effect and potential changes in the effect, associated uncertainties in both the computation of the CL and the analytical application of the CL in the REA, and judgements of the potential adversity of the impact on public welfare.

4.1.3 Changes in Ecological Responses

Exposure-response curves will be a key component of the assessments in the REA. These curves can show predicted ecological responses to specific levels of N or S deposition, as well as demonstrate the responsiveness of individual species and/or communities to changes in atmospheric deposition. These curves are primarily generated by observations of ecological response to experimental additions (e.g., N and S addition studies) or by observation of ecological response along a deposition gradient. We intend to use exposure-response curves to understand impacts under current conditions at a national-scale. In addition, we also plan to use the exposure-response curves to relate different air quality conditions, and associated deposition levels, in case study locations to potential changes in ecological effects.

4.2 AIR QUALITY ANALYSES

The sections that follow include information regarding the air quality and deposition datasets and analytical approaches that we intend to use to evaluate ecological effects in this REA.

4.2.1 Consideration of Current Conditions for National-scale Air Quality Concentrations and Deposition

Since the previous review, N and S deposition has changed and it is important to develop the most up-to-date datasets for the assessment of atmospheric deposition to capture these changes. We propose to rely on measurements of atmospheric concentration and deposition where available, and chemical transport model simulations to provide data for chemical species and locations where measurements are not available.

Accordingly, this review proposes to calculate air concentrations and deposition using the most recent CMAQ chemical transport model (version 5.2.1), with the most up to date meteorological and emission datasets, which will likely be for the calendar years 2014-2016. While there are other photochemical models available, CMAQ has been shown to have relatively low bias in estimating annual average wet deposition, with normalized mean bias reported as 7.9% for SO_4^{2-}, -12.8% for NH_4^+, and -15% for NO_3^- when compared with NADP measurements (Appel et al., 2011). In addition, while historically the CMAQ chemical transport model has reported dry deposition of NH_3 and particulate NH_4^+ separately, as these happen via different physical processes, the model has been recently augmented to be able to track the relative contribution of particulate NH_4^+ and gas-phase NH_3 to wet deposition of reduced nitrogen. This recent augmentation can provide data that will be useful in informing the contribution of particulate NH_4^+ to deposition for this review. For further analyses, we propose to evaluate the CMAQ simulations using the NTN observations of wet deposition as well as the CASTNet, CSN, IMPROVE, and AMoN observations of oxidized nitrogen, particulate chemical composition, and NH_3, respectively. The comparison of CMAQ results with these measurements can then be used to inform the uncertainty analysis.

The best available assessment of atmospheric deposition generally requires combining data from ambient measurements and computer model outputs. To do this, we propose to use the process described in Schwede and Lear (2014) to develop TDEP datasets for 2014, 2015 and 2016 that cover the continental U.S. at 4-km horizontal resolution. These gridded surfaces will also be combined to provide an estimate of average deposition for 2014-2016. The TDEP method estimates wet deposition by spatially interpolating NTN wet deposition measurements and PRISM precipitation observations. These estimates would then be combined with the dry deposition estimates from the 2014-2016 CMAQ simulations, projected from the 12-km CMAQ model resolution onto the 4-km TDEP grid. For further analyses of the potential error and uncertainty in the wet deposition dataset, we intend to use cross validation methods, such as reserving some of the wet deposition observations for evaluation.

The result of this analysis will be a spatially complete data set of 3-yr average deposition across the continental U.S. for 2014-2016, including nitrogen and sulfur deposition, wet and dry deposition, as well as the relative contribution from gas-phase NH_3 and particulate-phase NH_4^+. These data will be used to assess ecosystem effects at a national-scale and under current conditions, where much of the U.S. is meeting the current NO_2, SO_2 and PM standards.

4.2.2 Consideration of Air Quality Scenarios

The goal of the REA is to consider the level of risk under air quality conditions that just meet the current standards, as well as any potential alternative standards under consideration. To do so, we plan to conduct quantitative analyses in study area locations with air quality adjusted to reflect just meeting the current NAAQS, and just meeting other air quality scenarios, as needed. Accordingly, the sections below describe the proposed methodology for adjusting ambient concentrations to reflect specific air quality scenarios and statistically relating those levels of ambient concentrations to deposition levels of N and S. Table 4-1 lists the current NAAQS for PM, NO_2, and SO_2.

Table 4-1. Current National Ambient Air Quality Standards for PM, NO_2, and SO_2

Pollutant	Primary / secondary	Averaging time	Level	Form
NO_2	Primary	1 hour	100 ppb	98th percentile of 1-hour daily maximum concentration, averaged over 3 years
	Primary & secondary	1 year	53 ppb	Annual mean
$PM_{2.5}$	Primary	1 year	**12 µg**/m^3	Annual mean, averaged over 3 years
	Secondary	1 year	**15 µg**/m^3	Annual mean, averaged over 3 years
	Primary & secondary	24 hours	**35 µg**/m^3	98th percentile, averaged over 3 years
PM_{10}	Primary & secondary	24 hours	**150 µg**/m^3	Not to be exceeded more than once per year on average over 3 years
SO_2	Primary	1 hour	75 ppb	98th percentile of 1-hour daily maximum concentration, averaged over 3 years
	Secondary	3 hours	0.5 ppm	Not to be exceeded more than once per year

4.2.2.1 Estimating Ambient Concentrations

The goal of this analysis is to create spatially consistent datasets that represent scenarios where, in well-defined case study areas, concentrations are adjusted to just meet the current standards. A similar methodology would likely be followed if adjustments are needed to reflect just meeting potential alternative standards. These air quality scenarios will be used to estimate N and S deposition in the study area and to quantify ecological effects associated with the atmospheric deposition.

There are a number of unique technical challenges in adjusting air quality to reflect just meeting multiple standards, particularly given that the indicator pollutants for those standards are related differently through emissions, chemistry and transport. The first challenge is that, SO_2, NO_2, and PM, all have different levels of spatial variability. SO_2 and NO_x are directly emitted from sources. As SO_2 and NO_x are transported from the emission sources, they are diluted by atmospheric mixing, are chemically oxidized to other forms, including particles, and removed from the atmosphere by deposition. Locations further from sources have low SO_2 and NOx concentrations and higher concentrations of their oxidation products, like particulate sulfate and particulate nitrate. Because the sulfate and nitrate components of $PM_{2.5}$ are not directly emitted but are instead formed in the atmosphere, these components of $PM_{2.5}$ are often more spatially widespread. A scenario that adjusts SO_2 or NO_x must consider downwind impacts to sulfate and nitrate $PM_{2.5}$, and a scenario that adjusts $PM_{2.5}$ must consider if such changes would require SO_2 or NO_x concentrations that are greater than the standards in upwind areas.

Second, many sensitive ecological areas are located far from large emission sources and are affected by emissions in a large area. A recent study by Lee et al. (2016) examined eight Class I areas in the U.S., and found that 50% of the nitrogen deposition could be attributed to emission sources within 500 km and 90% could be attributed to emission sources as far away as 1500 km. The scenarios should not only consider concentration changes in an isolated emission sources area, but also in the upwind areas whose emissions contribute to deposition.

Finally, a related challenge is that there are additional components of PM that are not contributors to sulfate and nitrate deposition. For example, concentrations in the South Coast Air Basin are greater than the primary $PM_{2.5}$ NAAQS, but not the primary SO_2 NAAQS. Only a small portion of $PM_{2.5}$ in Los Angeles is comprised of particulate sulfate. If SO_2 concentrations were increased, sulfate would also increase, pushing $PM_{2.5}$ concentrations further beyond "just meeting" the standard. Because of the mix of emission sources and contributing components to $PM_{2.5}$, at many locations, there is not a physically realistic way to adjust SO_2, NO_2, and $PM_{2.5}$ concentrations where concentrations are just meeting all the relevant standards at the same time. Instead, the REA will need to identify the controlling standard, and associated pollutant, for the study area. In this analysis, we define the controlling standard as one where, when it is met, any increases in the concentrations of other N and S pollutants would cause the controlling standard to not be met.

Noting these challenges, we propose to first identify the area of influence for a particular study area location based on available information. The study area location is likely to be around 100 km in diameter. As a default, we may assume an area of influence with a radius of 1500 km, based on the findings of Lee et al. (2016). Within this area of influence, we will identify the controlling standard and associated pollutant (i.e. SO_2, NO_x, or PM) by evaluating information from ambient measurements and air quality modeling and considering current levels, as well as historical relationships between emissions and ambient concentrations. Then, using this information, we intend to develop an approach to adjust air quality in the study area location to reflect just meeting the identified controlling standard under consideration. Based on the analytical evidence in the recent SO_2, NO_2 and PM NAAQS reviews (U.S. EPA, 2018; U.S. EPA, 2017c; U.S. EPA, 2010b), we note the successful application of various statistical adjustment approaches in those reviews to estimate realistic changes in air quality concentration to reflect changes in emissions of SO_2, NO_2 and PM. Similar approaches will also be considered for this review, though in some areas we may also need additional information from air quality modeling to better quantify the contribution from an important emissions source. Using the approach developed, we intend to adjust air quality concentrations in the study area to develop an air quality scenario that just meets the current standards at the locations most relevant for the case study area. A similar approach would also be used to adjust air quality in the study area location to reflect just meeting any potential alternative standards under consideration. Using the approaches described in the next section, the changes in air concentration would be related to a change in deposition.

4.2.2.2 Relating Changing Levels of Atmospheric Concentration to Deposition

For our proposed analyses, relating air concentrations to welfare effects requires that a change in ambient concentration be related to a change in atmospheric deposition for input into an equation or model that then relates a deposition of N and/or S to an ecosystem effect. This section discusses the approach under consideration to estimate “deposition response factors,” or factors that can be used to relate changes in ambient concentrations of measured N and S species to changes in atmospheric deposition of N and S in areas across the U.S.

The previous review introduced the transference ratio, TR, where: *Deposition = TR × concentration*. In that review, the TR was estimated by dividing the annual average deposition by concentration in a single CMAQ simulation and averaging over an Ecoregion. There were also noted several uncertainties as described in Chapters 2 and 3, including:

- averaging over an ecoregion introduces uncertainty since the transference ratio is lower near emissions sources and larger far from emission sources, especially as NO_x is oxidized to form compounds that deposit more quickly;
- different chemical transport models report similar estimates of wet deposition but report very different transference ratios; and
- no quantitative assessment of uncertainty was conducted.

We propose to use an updated approach in this review to estimate deposition response factors for each form of N and S deposition, including both dry and wet deposition. For this approach, we note that in most areas of the U.S., concentrations and deposition of both oxidized nitrogen and sulfur have declined since the last review and that measurements at dozens of co-located CASTNet and NTN sites over the last 25 years are available to provide an observational record of how a change in ambient concentration is related to a change in atmospheric wet deposition. In addition, a 20-year CMAQ chemical transport model simulation (Zhang et al., 2018) provides a modeled estimate of the relationship between ambient concentrations and both wet and dry deposition. Taking advantage of these available modeled and measured concentration and deposition data, we propose using statistical models to estimate deposition response factors that relate a change in air concentration to a change in deposition and quantitatively assess the uncertainty in various study locations that will be selected for this analysis. These deposition response factors would then be used in conjunction with the adjusted air quality scenarios (section 4.2.2.1) to estimate the N and S deposition associated with just meeting the current standards, as well as any potential alternative standards under consideration.

In doing this, we propose to identify several pollutants or combinations of pollutants that can be measured and used to relate ambient concentrations to atmospheric deposition. We will refer to these as “deposition response factors.” Criteria for a robust deposition predictor are (*i*) it can be measured in ambient air with known accuracy and (*ii*) it can be used to predict atmospheric deposition with low error. We note that these deposition response factors may be different than the indicators for the current standards (i.e., NO_2, SO_2, $PM_{2.5}$ and PM_{10}) and when this is true, we plan to also assess the relationship between the deposition predictor and the indicator for the current standards (including consideration of form, averaging time and level of the standards).

We have listed our potential deposition response factors in Table 4-1. For some deposition response factors, co-located measurements of ambient concentration and wet deposition from 1990 to the present day exist and can provide a dataset to estimate the change in wet deposition related to a change in air quality concentration. For dry deposition measurements that are not routinely available, the CMAQ modeling information can be used to estimate the relationship between dry deposition and air quality concentrations. Using these datasets, the annual average concentrations and deposition calculated by a 20-year CMAQ chemical transport model simulation will be used to fit a statistical model for each deposition predictor at the grid-level scale (36 km grid cells), as well as at larger spatial scales. Measurements of ambient concentration and wet deposition and their 20-year trends will be used to evaluate the chemical transport model and statistical model results. Each of the steps in this analysis is listed below.

(1) Using a CMAQ simulation for 1990-2010 (Zhang et al., 2018), calculate the annual average deposition (wet, dry, and total) for oxidized N, reduced N, and sulfur at each grid cell location. Also calculate the annual average air concentration of NO_2, NO_y, HNO_3, particulate NO_3^-, NH_3, particulate NH_4^+, SO_2, and particulate SO_4^{2-} at each grid cell location.

(2) For a specific grid cell, use the 20 years of concentration and deposition data to estimate a statistical model that uses annual average atmospheric concentration -- one of the proposed *deposition response factors* -- to predict the annual average dry deposition. For example, the sum of particulate SO_4^{2-} and SO_2 is a proposed deposition predictor for dry deposition of sulfur.

(3) Record the *residual* – the difference between the deposition predicted by the statistical model and the deposition calculated by CMAQ – for each of the predicted annual deposition. Calculate the average residual. This is used as an assessment of the magnitude of error when using this deposition predictor.

(4) Repeat (3) for each grid cell and create a map of the average residual and best fit coefficient. Examine how these vary.

(5) Repeat (3-4) for each deposition predictor listed in Table 4-1. For example, for oxidized N, this could include HNO_3, NOy, or total nitrate (HNO_3 + particulate nitrate). Compare the residual for each different deposition predictor. Compare both the national residual average as well as use the map to make sure there are no areas where the error is exceptionally large.

(6) Repeat (3-5), but instead fit a statistical model to predict wet deposition and total (sum of wet and dry) deposition. Where possible, compare with historical measurements of wet deposition and atmospheric concentration at co-located NADP and CASTNet sites. Examine if the residual lower when estimating wet and dry deposition individually or when added together as total deposition.

(7) Repeat (3-6), but rather than fit a different model for each grid cell, use all the grid cells, excluding those over oceans, to fit a statistical model. Create a map of the average residual for each grid cell. Explore different averaging areas for use in case studies to find a balance that is representative and does not obscure variability in the transference ratio.

(8) Compare the residuals and select a deposition predictor that has low average residual and is nationally relevant. Use the distribution of residuals as an estimate of uncertainty.

Table 4-2. Initial Assessment of Available Deposition Response Factors

Deposition predictor	Description	Available measurements	Reliably related to deposition?
Total nitrate: HNO_3(g) + $PM_{2.5}$ NO_3^-	The sum of gas-phase nitric acid and particulate nitrate	Measured at CASTNet at many locations since 1990	Co-located NTN and CASTNet monitors show oxidized N wet deposition can be reliably predicted with total NO_3^- air concentration measurements (Sickles and Shadwick, 2013)
Total sulfate: SO_2(g) + $PM_{2.5}$ SO_4^{2-}	The sum of gas-phase SO_2 and particulate SO_4^{2-}	Measured at CASTNet at many locations since 1990.	Co-located NTN and CASTNet monitors show S wet deposition can be reliably predicted with total sulfate air concentration measurements
NOy	Sum of oxidized N in the gas phase and particulate nitrate	Measured at NCore sites, although most are in urban areas.	Most NOy measurements are not co-located with deposition measurements, which makes it difficult to evaluate. Sickles and Shadwick (2013) demonstrated that this sometimes has (and sometimes does not have) low error for predicting deposition.
$PM_{2.5}$ NH_4^+	Particulate ammonium	Measured at CASTNet and CSN but measurements are biased and known to be lower than atmospheric concentrations.	Co-located NTN and CASTNet monitors show reduced N deposition cannot be reliably predicted using PM2.5 NH_4^+. This could be explained partially by bias in CASTNet NH_4^+ measurements. Another consideration is most of the US is thought to have excess NH_3 relative to sulfate and nitrate, which suggests that NH_4^+ concentrations are more controlled by sulfate and nitrate levels, rather than NH_3 levels.
NH_x	The sum of gas-phase NH_3 and particulate NH_4^+	Co-located CASTNet, NTN, and AMoN sites measure gas phase NH_3 and particulate NH_4^+. New approaches to measure NH_x more robustly are under evaluation.	Co-located CASTNet, NTN, and AMoN sites suggest that NH_4^+ wet deposition can be reliably predicted using NH_x measurements. However, NH_3 is not part of this review and therefore the scope of this review would need to be expanded before this deposition predictor could be considered.
NO_2, SO_2, $PM_{2.5}$, PM_{10}	Existing primary NAAQS are based on these compounds	Nation-wide measurement networks, but mostly located near urban areas or large emission sources. Few are co-located with deposition measurements.	The analysis described above will examine how reliably these compounds can be used to predict deposition.

Based on preliminary analyses, total nitrate (defined as the sum of nitric acid and particulate nitrate) could be the most promising deposition predictor for oxidized nitrogen deposition. Analysis of co-located CASTNet and NTN measurements have found total nitrate to be well correlated with oxidized nitrogen wet deposition (Figure 4-2) in the Eastern U.S. where year-to-year variability in precipitation is relatively low. Analyses also suggest that the sum of SO_2 and particulate sulfur is well correlated with sulfur wet deposition. However, preliminary results suggest that in many locations, particulate NH_4^+ is not a good predictor of reduced nitrogen deposition, as recent measured trends show declining particulate NH_4^+ concentrations with increasing or stable reduced-nitrogen deposition.

This approach addresses several of the uncertainties inherent in the approach used to relate concentrations and deposition in the previous review. By using a statistical approach and the 20-year time series, we can quantify and compare the level of uncertainty for different deposition response factors. Rather than average over ecoregions, we will assess the spatial variability in the transference ratio and select an averaging area best suited to the ecological effect of interest. As described in Section 4.5, we will conduct comparisons with two chemical-transport models. More analysis (discussed in Section 4.5) is planned to identify sources of variability and quantitatively assess the uncertainty in using each of these deposition response factors.

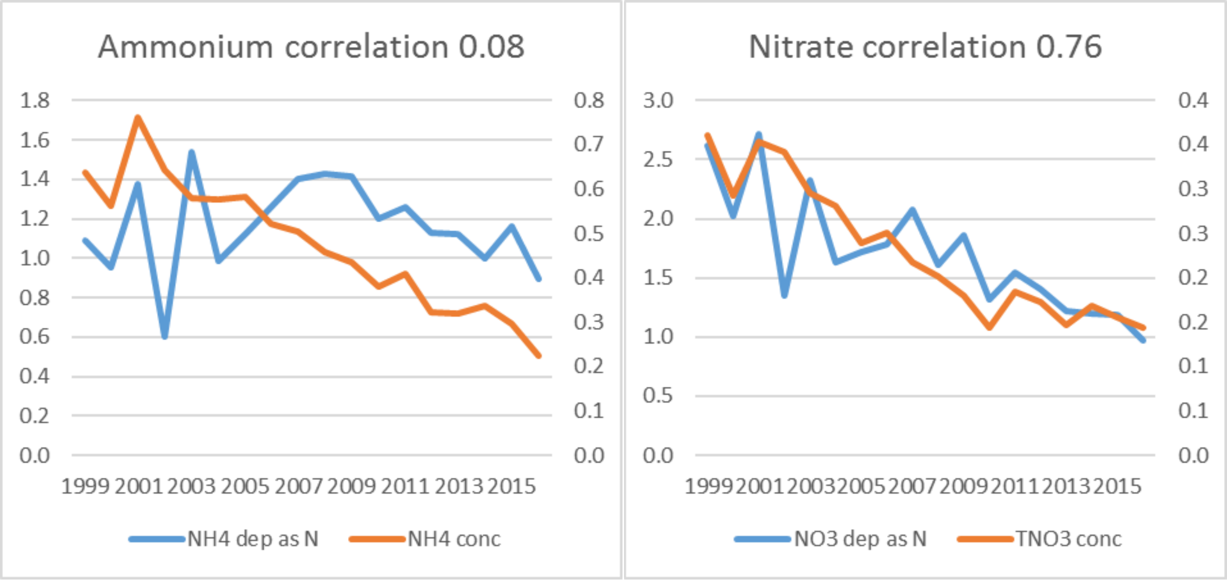

Figure 4-2. **NADP National Trends Network wet deposition (blue, left axis, kg/ha N) and CASTNet air concentration measurements (orange, right axis, µg/m^3 N) for Acadia National Park from 1999-2016 annual average. CASTNet measurement of particulate NH_4^+ is not well correlated with reduced N wet deposition (*r* = 0.08, left figure), while total nitrate (gas-phase nitric acid plus particulate nitrate) air concentration is well correlated (*r* = 0.76) with oxidized N wet deposition (right figure).**

4.3 ECOLOGICAL EFFECTS ASSESSMENT

4.3.1 Aquatic Acidification and Nitrogen Enrichment

4.3.1.1 Freshwater Acidification

The connection between SO_2 and NO_x emissions, atmospheric deposition of N and/or S, and the acidification of surface waters is well documented with several decades of evidence for the eastern U.S. (see second draft ISA, Appendix section 7.1.1.2 and Driscoll et al., 2016). As mentioned in Chapter 2, the 2009 NOx/SOx REA evaluated freshwater acidification in the Adirondacks and Shenandoah, using ANC as the chemical indicator to derive CLs. In the REA for this review, we intend to use a similar approach to evaluate freshwater acidification on a national scale, and to better understand the linkages between chemical changes (changes in ANC) in freshwater ecosystems attributable to acidifying deposition of N and/or S and changes in biological effects on receptors, such as fish. To accomplish this, we intend to use a combination of water quality measurements, geology, and surface water steady-state CLs data to define the spatial distribution of acid-sensitive ecosystems across the U.S.

Biogeochemical dynamic models have also been used to assess impacts on water quality (e.g., pH and ANC) compared with pre-acidification (i.e., pre-industrial) water quality conditions (see second draft ISA, Appendix section 7.1.5.1, and Sullivan et al., 2011). Atmospheric N and S deposition in recent decades have shown marked decreases due to significant power sector, industrial, and mobile emissions reductions, allowing for the chemical and biological recovery of water bodies (see Section 1.11.1 of the second draft ISA for more information on recovery). Therefore, the evaluation of current ecological conditions will need to be done in the context that aquatic ecosystems are on a recovery trajectory in response to decreases in atmospheric N and/or S deposition. We intend to evaluate current conditions of aquatic ecosystems and their response to atmospheric N and/or S deposition using steady-state and target CLs and model output from biogeochemical models. We intend to use CLs for aquatic acidification from the NCLD v3.0 as well as any relevant new peer-reviewed publications (e.g., Blett et al., 2014; Sullivan et al., 2015; Fakhraei et al., 2014; Fakhraei et al., 2016; Shaw et al., 2014; Lawrence et al., 2016), where the data are not yet included in the NCLD v3.0. In developing the REA, consideration will also be given to any new CLs in scientific literature, but a comparison of all CLs will be completed to more fully understand the differences.

ANC is an important chemical indicator, defined as the total amount of strong base ions minus the total amount of strong acid anions:

$$ANC = (Ca^{2+} + Mg^{2+} + K^{+} + Na^{+} + NH_4^{+}) – (SO_4^{2-} + NO_3^{-} + Cl^{-})$$

While ANC is not directly linked to the physiological impacts of aquatic biota, it is commonly used because it integrates overall acid status. Unlike pH, it is not affected by dissolved carbon dioxide (CO_2), and water quality models are generally better at modeling ANC than other indicators (see second draft ISA, Appendix, section 8.1). In addition, ANC has been extensively related to the health of biota and other surface water constituents like pH and Al (see second draft ISA, Appendix, sections 8.3.6.3). In the case where DOC plays an important role in acid-base balance of the surface water, the measure of base cation surplus (BCS) that adjusts for the organic acid status of surface waters (Lawrence, 2007) will be considered. BCS is a metric like ANC, but accounts for the charge balance natural organic acidity contributes to the ionic concentration of the surface water (see second draft ISA, Appendix section 8.1).

Given the connection between atmospheric N and S deposition to biogeochemical changes that may induce harmful biological effects, we intend to use ecological literature included in the second draft ISA, Appendix section 8.3 and survey data from various sources to explore revising the aquatic status ANC categories of potential effects that were used in the 2009 NOx/SOx REA (e.g., Table 4.2-1 from the 2009 NOx/SOx REA). We will also work to refine the relationship between ANC and the biological responses. Compiled information for major taxonomic groups and other biogeochemical and ecological information (e.g. pH, habitat condition) will be used to inform the aquatic status ANC categories of potential effects described in the second draft ISA (see second draft ISA, Appendix section 8.3). Where data are available, we intend to develop exposure-response relationships (total species-richness vs. pH or ANC, biomass vs. pH or ANC, etc.) by building on the research of Layer et al. (2013), Baldigo et al. (2018), and other studies listed in the second draft ISA, Appendix section 8.3. Additionally, we intend to evaluate the CLs with respect to range of ANC thresholds of 20 to 100, which represent the range of ecosystem protection for biota as described by the aquatic status ANC categories. This falls within the range that is most commonly used in the NCLD v3.0 and among CL assessments in the U.S. (see second draft ISA, Appendix Table 8-8).

For the case study areas, we intend to use the MAGIC model to evaluate potential ecological and biological changes due to changes in atmospheric deposition. Knowing the pre-acidification water quality conditions provides a benchmark to measure how near chemical recovery is to the pre-acidification level. The time it takes to reach chemical recovery will also be determined using the MAGIC model. A host of environmental factors and the severity of atmospheric deposition-driven acidification influence how fast aquatic ecosystems respond to reduction in atmospheric N and/or S deposition. In sensitive ecosystems that are strongly impacted chemical recovery varies significantly, but can take decades. The selection of case study areas will aim to include a subset of aquatic ecosystems from various sensitive regions with long-term water quality records with supporting environmental data to be modeled using the biogeochemical models.

4.3.1.2 Freshwater Nitrogen Enrichment

We intend to further consider the extent of the analyses for the REA that use new CLs data from Baron et al. (2011), Williams et al. (2017), and Nanus et al. (2017), as described in Chapter 3, to evaluate aquatic N enrichment effects in freshwater lakes and streams in the REA. These CLs use NO_3 concentrations as an indicator of N enrichment, associated with varying biological and chemical criteria as discussed in section 3.5.2 and the second draft ISA Appendix 8, section 8.5.4.1[47]. The potential analyses are described below but, as we develop the REA, we plan to further consider which, if any, of these analyses will be included in the REA with the judgement being based on whether the results can substantively inform our ability to assess national standards for freshwater N enrichment.

The Baron et al. (2011) and Williams et al. (2017) studies provide CLs with broad geographic scope, which apply to all lakes within a given study region. We could potentially apply these CLs in the specific regions covered under each of the studies as follows: apply the CLs from Baron et al. (2011) to lakes included in the study in the Sierra Nevada Mountains, Rocky Mountains, and the northeastern U.S. to assess risk of elevated NO_3 in surface waters[48]; apply the CLs from the Williams et al. (2017) study to western lakes that fall within the sampled criteria (elevation > 1200m), with a higher level of confidence attributed to lakes that were included in GLNC to assess risk of shifts from N to P-limitation of phytoplankton growth. The CLs from the Nanus et al. (2017) study provide CLs that are specific to each lake within the greater Yellowstone area. We could apply these CLs to the greater Yellowstone area as defined in the study to potentially assess the risk of changes to diatom growth and community composition. We could also explore the application of these CLs to the larger Rocky Mountain area assessed by Nanus et al. (2012) and potentially to other areas of the western U.S. In addition, the regression parameters in Nanus et al. (2017) constitute exposure-response functions that potentially relate the NO_3 levels in individual lakes to atmospheric N deposition as well as other watershed level factors, including slope and amount of barren ground. These functions could be used to calculate expected NO_3 levels based on different atmospheric deposition patterns. The resulting NO_3 levels could be compared the categories of potential effects for NO_3 described above. The combination of these response functions and categories of potential effects

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[47] We note that elevated NO_3 concentrations in lakes and streams are also indicative of possible N saturation in adjacent terrestrial ecosystems leading to leaching of NO_3 into the waterbodies (see second draft ISA Appendix 4, section 4.3.2 and Appendix 7, section 7.1.2).

[48] Lake data in Baron et al. (2011) were taken from Linthurst et al. (1986) for eastern lakes and Eilers et al. (1987) for western lakes

could potentially be used to assess the risk posed by different levels of atmospheric N deposition to freshwater lakes. Depending on the study being applied, risk may be assessed at individual lakes and/or aggregated at larger areas.

4.3.2 Terrestrial Effects

The connections between emissions, atmospheric deposition of N and/or S, and the alteration of plant community composition and species richness in terrestrial ecosystems is well documented (see second draft ISA, Appendix section 5). For terrestrial ecosystems, the main findings of the current ISA are that N and S deposition cause alteration of soil biogeochemistry and effects on multiple levels of biological organization ranging from physiological processes to shifts in biodiversity and ecological function (ISA ES, p. lxxii).

Thus, this section describes potential quantitative analyses for soil biogeochemistry and groups of organisms. Some data are available at the level of biogeochemical effects while other datasets reflect growth and mortality effects on individual species of plants or effects to community composition and biodiversity. The sections below provide details on the studies that we intend to use, as well as any specific modifications and decisions that will be made or considered, when conducting the quantitative assessments.

It is important to note that in terrestrial ecosystems, the acidifying effects from N and S, and the enrichment effects from N can occur simultaneously within the geographic boundaries of an ecosystem. However, the dominance of one process over the other (i.e., acidification versus N enrichment) can vary depending upon several factors, including soil pH and buffering capacity, the presence and abundance of sensitive species, and the influence of nitrogen limitation on primary production. Because acidification and N enrichment effects are intertwined, it is often difficult in large scale gradient studies to isolate mechanisms (i.e., acidification versus N enrichment), as both are co-occurring across the landscape. When possible, the mechanism of effect will be noted in the description of the datasets.

4.3.2.1 Biogeochemical Effects in Soils

As mentioned in Chapter 2, the 2009 NOx/SOx REA used a case study approach for sugar maple and red spruce, deriving CLs using Bc/Al as the chemical indicator for acidification of forested soil that related to tree health. In the REA for this review, we intend to use a similar approach, but now will expand the CLs nationally. We intend to use the Bc/Al ratio as a soil chemical indicator, given that it is commonly used, relates well to the Ca/Al ratio, and serves as an important input into the SMB model for calculating soil CLs (see second draft ISA, Appendix section 5.2.1).

There is precedent for using Bc/Al levels between 1 and 10, because these values are documented to cause deleterious effects to trees, as the chemical limit in CLs calculated using the SMB model. For example, McNulty et al. (2007) chose Bc/Al levels of 1 (for coniferous forests) and 10 (for deciduous forests). Additionally, Duarte et. al (2013) and Phelan et. al (2014) conducted studies in the Northeastern U.S., each using a Bc/Al level of 10. For the REA, we intend to calculate national CLs using the SMB model and Bc/Al levels of 1 and 10 to represent different forest types in different parts of the U.S.. We will also explore in the REA whether there are levels between 1 and 10 that should be applied, considering any new scientific evidence for relating Bc/Al to categories of ecological effects. This approach is similar to the approach proposed for the use of ANC as a chemical indicator for freshwater acidification and NO_3 as a chemical indicator of freshwater N enrichment.

BCw is one of the most influential yet difficult to estimate parameters for deriving the forest soil acidification CLs (see second draft ISA, Appendix section 4.5.1). There are two primary methods available for estimating BCw: the clay-substrate method and the PROFILE model. The clay-substrate method has been applied nationally, but as noted in the 2009 NOx/SOx REA, has known issues especially when estimating BCw in unglaciated soils. Given the results of Phelan et al. (2014) in the Pennsylvania case study, we intend to explore the use of PROFILE modelling to obtain more accurate national estimates of BCw rates for the REA, considering uncertainties identified in Whitfield et al. (2018). Additionally, researchers have recommended applying PROFILE in different areas of the U.S. to improve our understanding of how the model performs and to compare the BCw rates estimated by PROFILE to those estimated using the clay correlation-substrate model. (see second draft ISA, Appendix section 5.5). We will use the results from Phelan et al. (2014) to explicitly quantify the relationship between PROFILE estimated BCw and clay-substrate-method quantified BCw.

4.3.2.2 Species-Level Effects

4.3.2.2.1 Trees

We intend to conduct quantitative analyses to understand growth and mortality effects on individual tree species across the contiguous U.S under current air quality conditions using the exposure-response curves described in Horn et al. (in review).[49] These curves are national in scope and represent statistical equations that relate growth and mortality responses to N deposition, S deposition, and other factors including temperature, precipitation, tree size, stand basal area.

There are 156 species assessed in Horn et al. (in review). Additionally, the dataset includes benchmarks for the points on the curve where a species experiences various levels of reductions in growth and survival − 1%, 2%, 5%, 10%. However, the equations themselves can

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[49] In developing the REA, if Horn et al. (in review) is not published, we plan to revisit the proposed plan for the REA and consider analyses that focus on other published literature (e.g., Thomas et al., 2010).

be used to calculate any percentage desired. Trees were sampled at United States Forest Service (USFS) FIA plots, with at least 2,000 trees sampled for each species constituting a dataset of over 50,000 plots and more than 1.2 million trees. There were 94 species that met the N>2,000 sample size cutoff for both growth and survival (highlighted in the publication) and an additional 14 species that met the cutoff for survival only[50]. TDEP data years used for the deposition estimates overlap with the period of re-measurement for each plot. Samples at each plot were collected every 5 years on average. Data years for the dataset span from 2000-2016, with each plot linked with the average deposition experienced between the first and last measurement period.

There are two methods under consideration for spatially associating the response curves from Horn et al. (in review) to the specific locations of the tree species being evaluated. One approach considers analytics based on the USFS FIA plot data, and the other based on the USFS continuous surface maps (Wilson et al., 2013). Both methods have their advantages and disadvantages, which are summarized more below.

- USFS FIA plot data. Our confidence is high in this data because it is based on empirical field measurements. However, although the density of plots is high when viewed at the continental scale, at smaller scales the density may be low enough that inferences across areas are limited. Thus, because there is one sample taken every 6,000 acres across the landscape according to the FIA plot layout, there are several areas that are important to public welfare, such as Class I Wilderness Areas and National Parks, that are composed of only a few FIA plots.
- Wilson et al. (2013) USFS maps: These maps are based on the FIA data that provides continuous surface maps of predicted abundances for 322 species in 250 m grid cells across the contiguous U.S. (including all 94 tree species included in Horn et al. (in review)). These abundances are based on statistical relationships with various factors (i.e., phenology, temperature, precipitation, topography, and ecoregion). Abundance in Wilson et al. (2013) is based on basal area, which is a common metric in forestry that represents the area of tree trunks within an area of land (ft^2 ha^{-1}). The accuracy of these predictions varies by species and the scale of inference, with higher accuracy for more abundant species and larger areas. Wilson et al. (2013) and Riemann et al. (2010) provide several metrics to assess the accuracy of projections for all 322 tree species. All tree species in Horn et al. (in review) are included in Wilson et al. (2013).

For the analyses, we will determine the most appropriate methodology for applying response curves across the landscape in the REA by considering the benefits and limitations of both datasets. These data will be compared to the N and sulfur deposition values from the national-scale TDEP surface for current air quality conditions. This information will help to

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[50] Note the study focused on the 94 species that met the criteria for both, though all results are provided in the Appendix of the paper.

further identify sensitive areas of the U.S. that are impacted by recent deposition levels and may be good candidates for the case study approach.

4.3.2.3 Community-Level Effects

4.3.2.3.1 Mycorrhizal Fungi

The mycorrhizal fungi dataset is a collection of empirical CLs for total atmospheric N deposition from various studies included in Pardo et al. (2011) as well as newer studies reviewed by the ISA (e.g. Allen et al., 2016). This dataset includes minimum and maximum CLs for changes in community composition. CLs for mycorrhizal fungi will be used to assess the potential for current levels of deposition to affect mycorrhizal community composition in forests.

4.3.2.3.2 Lichen Community

The underlying analyses in Geiser et al. (2010) and Root et al. (2015) include response-functions relating N deposition to lichen community composition. The functions described are currently based on data from the Pacific Northwest, so extrapolation to other areas of the country would have higher levels of uncertainty. While the NCLD v.3.0 includes other lichens CLs from Pardo et al. (2010) based on individual site-specific studies, we intend to use only the CLs from Root et al. (2015) and Geiser et al. (2010) for assessing risk because they use more consistent methodologies. Furthermore, we note that new work is underway to extend the work of Root et al. (2015) and Geiser et al. (2010) using similar methods on a national scale (personal communications with Linda Geiser, 2018). If this work becomes available, it may be used to broaden the geographic scope of the lichen data considered in the REA.

Because of the spatially restricted nature of the lichen study from Geiser et al. (2010) and Root et al. (2015), we will likely restrict the application of these exposure-response curves to the Western U.S. (specifically, to the two Ecoregions in the west where data were collected: North American West Coast Marine Ecoregion in Oregon and Washington and the Northwestern Forested Mountains Ecoregion in Northern California, Oregon, Washington, and parts of northern Idaho and Montana). We will consider extrapolation to other portions of these Ecoregions, but not nationally. Using the relationships in Geiser et al. (2010) and Root et al. (2015), we will calculate sensitivity and risk to lichen communities using the CLs and the exposure-response curve.

4.3.2.3.3 Herbs/Shrubs

Given their national coverage, we intend to use the CLs from Simkin et al. (2016) to evaluate changes in species richness for herbs/shrubs communities. We note that while the NCLD v.3.0 also includes a collection of herbs/shrubs empirical CLs from Pardo et al. (2011), we have chosen to use those derived from Simkin et al. (2016) because the study is national in coverage, has explicitly quantified uncertainty, and used a common methodology in contrast with those from Pardo et al. (2011). The statistical equations in Simkin et al. (2016) constitute exposure-response curves that relate the species richness of herbs/shrubs at a site to N deposition as well as other factors, including soil pH, temperature, and precipitation. We will use these response curves to assess the risk posed by N deposition to herb/shrub community species richness. Risk will be assessed at individual sites or aggregated at larger areas. We will also explore the use of these curves to examine loadings that would minimize reductions in species richness at individual points/sites and across the country. However, avoiding reductions in total numbers of species does not avoid changes in composition and losses of individual species (e.g., a gain for 2 species and loss for 2 with no change in total species richness). Thus, if additional information becomes available at the species level, we will incorporate it into the analysis in the REA.

4.3.2.3.4 Forests and Grasslands

While we intend to conduct quantitative analyses to understand growth and mortality effects on individual tree species across the contiguous U.S under current air quality conditions using the exposure-response curves described in Horn et al. (in review), we also plan to apply the exposure-response curves to consider changes in the growth and survival of multiple species in the selected study areas for various air quality scenarios that include just meeting the current standards, as well as any potential alternative standards, as appropriate. In doing so, we intend to evaluate potential effects on tree species within a forested or grassland area and, as such, potential changes in the species composition for that area. We note that in our evaluation that, in some areas, we may find that some species are especially abundant and responsive to N and S deposition, and drive much of the changes in forest or grassland growth and survival rates. On the other hand, another area may be dominated by unresponsive species, and only locally rare species are estimated to be impacted. Therefore, the analyses will also provide information regarding the extent to which there are differences in effects for various species within each study area and between the set of study area locations.

4.4 CASE STUDY AREA SELECTION

A case study approach is being proposed in the REA to provide the ability to include more refined analyses than can be done at the national scale and that can reflect various combinations of changing air quality and deposition, as needed and as appropriate. For the case study component of this REA, we intend to characterize ecological risk and exposure in 5-10 study area locations for when air quality is just meeting the current standards, and if appropriate, when meeting potential alternative air quality standards. While we do not intend for the study area analyses to provide a comprehensive national assessment, they are intended to provide assessment for a small varied set of study areas that will be informative to EPA's consideration of potential risks and exposures that may be associated with various air quality scenarios.

In selecting the case study locations, we intend to consider specific criteria, with the goal of including case studies in the REA that are particularly informative to the review. Based on currently available data, tools and information and considering the proposed approaches to the analyses, as discussed above, we propose the following set of criteria for consideration in the selection of the study area locations:

- **Influence of current air quality on deposition-related effects.** As recognized above, historic air quality (and associated deposition) has had appreciable impacts on ecosystems in many areas. In such areas, the continued role of this historic deposition in the area's deposition-related impacts poses challenges to us in assessing the effects of current deposition or deposition associated with the current standards. Therefore, it is particularly important for study area selection to understand, as well as possible, the influence of current air quality (compared to historic air quality) on deposition-related effects. In areas with relatively greater influence of current air quality, we would expect the deposition-related risk and exposures to be responsive to changes in deposition. The impacts for various air quality scenarios in such areas would be expected to be particularly informative to consideration of the adequacy of the current standards. Thus, case study locations in which the deposition-related effects are not dominated by the influence of historic air quality would be good case study candidates. Accordingly, features of an area to be considered will include geology, historical deposition levels, recovery potential, and has the area experienced historically high deposition levels such that, for example, a terrestrial system is N saturated.
- **Sensitivity.** Related to the prior criterion is one that considers the extent of ecosystem sensitivity. To insure consideration of the depositional impacts on at-risk ecosystems, it will be important to consider including areas for which ecosystems or receptors are sensitive to N and/or S deposition, at risk due to such deposition, and specifically sensitive to changes in deposition.
- **Importance to public welfare.** To assess impacts on public welfare (in order to inform consideration of the adequacy of public welfare protection afforded by the current standards), good candidate areas will be areas for which there is a connection of potential impacts to ecosystems and effects on public welfare. For example, pertinent considerations may include: is the area in a National Park or Class I area where there is a clear connection to public welfare? Does the area include endangered or threatened species? Are there culturally or economically significant species present?
- **Diversity and occurrence.** A good set of candidate areas would encompass a set of diverse conditions occurring across the country. Given this, we propose to consider selecting a set of areas comprising a broad array of NOx/SOx/PM-related chemical environments, while also ensuring inclusion of areas that reflect current atmospheric deposition patterns (e.g. areas dominated by oxidized as opposed to reduced forms of N). Similarly, we propose to consider areas where the resident ecological receptors also occur more broadly (e.g. tree species in the area are widespread and abundant in locations outside of the study area) as well as areas that encompass several different ecosystems.

- **Data availability and adequacy.** It is important for the case study locations to have adequate data available, thus reducing uncertainty in the risk and exposure analyses.
    - Air quality data: Given that our objective relates to the relationship between air quality concentrations and deposition, a good candidate study area would be one in which: air concentration and wet deposition measurements for multiple N and S species are available for recent years and the monitors are either co-located or located close to each other; CMAQ model performance in predicting concentrations and deposition of N and S species is reasonable; and the statistical model fit to predict wet deposition and total deposition has reasonably low uncertainty. An additional related consideration is the magnitude of air quality adjustment that would be required for the area's air quality to just meet the current standards, with the understanding that smaller air quality adjustments will likely be less uncertain.
    - Freshwater and terrestrial ecosystem data: Given the reliance on CLs and exposure-response function data in our proposed analyses, a good candidate area would have: CLs and exposure-response function data available; CLs data available for multiple endpoints or effects (sections 4.1.2 and 4.1.3); and exposure-response function data available for multiple receptors (sections 4.2.4 and 4.2.5) and for multiple tree species (section 4.2.5.2). Additionally, in a good candidate area, CLs data would have sufficient individual site measurements and tree species with exposure-response functions to allow robust quantitative analyses.
- **Availability of additional information.** An additional criterion for consideration is the availability of other pertinent data. For example, the second draft ISA included six case studies, chosen because they were areas for which there was substantial amount of published work on ecological response to N and/or S deposition available (second draft ISA, Appendix 16) 51. A consideration may be whether the qualitative scientific and air quality information in one or more of the case studies in the second draft ISA be used when considering the criteria above. Other information may also be available that makes an area a good candidate study location. For example, we may want to consider whether there is any other qualitative information that is useful (e.g., ecosystem services information or survey data).

4.5 CONSIDERATION OF VARIABILITY AND UNCERTAINTY

To characterize ecological risks, exposure and risk assessors commonly use an iterative process of gathering data, developing models, and estimating exposures and risks, given the goals of the assessment, scale of the assessment performed, and limitations of the input data available. However, significant uncertainty often remains and emphasis is then placed on characterizing the nature of that uncertainty and its impact on exposure and risk estimates.

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[51] The locations included were the northeastern U.S., Adirondack State Park, southeastern Appalachia, Tampa Bay, Rocky Mountain National Park, and southern California. The case studies identified current acidification and nutrient status, as well as empirical and modelled CLs.

Below we summarize the most important uncertainties potentially affecting the risk and exposure estimates and how we can address some of them using newly available information. Conclusions drawn from these characterizations based on new information and any new evaluations performed in the current REA will be synthesized following the approach outlined by the World Health Organization (WHO, 2008). This synthesis will be used to identify, evaluate, and prioritize the most important uncertainties relevant to the estimated exposure and risk outcomes. We intend to use both a qualitative approach along with quantitative approaches where possible in the REA as discussed below.

The approach to be used here varies from that described by WHO (2008) in that a greater focus will be placed on evaluating the direction and the magnitude[52] of the uncertainty; that is, qualitatively rating how the source of uncertainty, in the presence of alternative information, may affect the estimated exposures and ecological risk results. Following the identification of key uncertainties, staff will subjectively scale the overall impact of the uncertainty by considering the relationship between the source of uncertainty and the exposure concentrations (e.g., low, moderate, or high potential impact). Also to the extent possible, staff will also include an assessment of the direction of influence, indicating how the source of uncertainty could affect estimated exposures or risk estimates (e.g., the uncertainty could lead to over- or underestimates). Further, and consistent with the WHO (2008) guidance, staff will discuss the uncertainty in the knowledge base (e.g., the accuracy of the data used, acknowledgement of data gaps) and decisions made where possible (e.g., selection of model forms). The output of the uncertainty characterization will be a summary describing, for each identified source of uncertainty, the magnitude of the impact and the direction of influence the uncertainty may have on the exposure and risk characterization results. The discussion below illustrates some of the main considerations for uncertainty and variability in air quality, CLs datasets broadly, and specific uncertainty considerations for specific effect categories.

4.5.1 Air Quality

The review intends to examine uncertainty and variability in (*i*) our approach to assess current conditions and (*ii*) our approach for estimating the deposition response factors as the change in deposition related to a change in air concentration. To assess uncertainty in our estimate of current conditions, specifically, the merged model and measurement datasets of air concentrations and deposition from 2014 to 2016, we intend to examine the difference between modeled values and measurements. The spatially-interpolated wet deposition dataset will be assessed using cross validation, such as “leave-p-out” methods where a subset of the

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[52] This is synonymous with the “level of uncertainty” discussed in WHO (2008), section 5.1.2.2.

observations is reserved to assess the uncertainty in the interpolation. The CMAQ-calculated dry deposition could also be evaluated with measurements where possible, but given the scarcity of dry deposition measurements, we propose examining three sensitivity studies. First, we could conduct Comprehensive Air Quality Model with eXtensions (CAMx) simulations with similar model configurations and the same emissions and meteorological inputs as the 2016 CMAQ simulations. Earlier work and CASAC committee have noted differences between CMAQ and CAMx deposition estimates from previous model versions, and a large fraction of the air quality modeling community (including Federal Land Managers) use CAMx in their work. Second, we could conduct simulations using CMAQ augmented to include the Zhang dry deposition scheme used in CAMx. This simulation is to understand uncertainty arising specifically from alternative model configurations for dry deposition. Third, we could conduct simulations using CMAQv5.2.1 with mosaic land cover dry deposition to quantify the variability in deposition for different land cover types. Forests, grasslands, and lakes each support ecosystems with differing sensitivity to nitrogen and sulfur. Each of the land cover types are also physically different surfaces, which influences the micrometeorological conditions that govern dry deposition. For example, the dry deposition velocity of nitric acid over rougher forest areas can be a factor of 4 larger than over water areas. The purpose of this simulation would be to examine the effect of considering deposition specific to each land cover type.

We propose to also pursue multiple approaches to assess uncertainty in our estimate of the deposition response factors – the change in deposition related to a change in air concentration. First, because we are estimating the deposition response factors by fitting a statistical model, we can assess the range of errors expected by analyzing the residuals, or the difference between the statistical model predictions and the data used to fit the statistical model. Second, since the statistical model is fit using CMAQ model results, we can also fit a statistical model to co-located measurements of air concentration and wet deposition. We have greater certainty when our estimate of the deposition response factors is similar when using either CMAQ model data or co-located measurements to fit the statistical model. Next, we can examine different approaches to spatial averaging of the air concentration and deposition data before fitting the statistical model, in order to develop an approach with lower uncertainty that can still capture the spatial variability needed to assess ecosystem impacts. Finally, we can examine our findings collectively from these uncertainty assessments and develop a framework to integrate them into a quantitative assessment of the total uncertainty in our estimate of the deposition response factors.

4.5.2 Critical Loads Data

Potential uncertainty/limitations for the CLs datasets depend upon several factors: 1) whether the dataset includes CLs from one study (i.e., that used a consistent methodology) versus multiple studies (i.e., where different methods may have been applied); 2) whether CLs were extrapolated from study areas to broader geographical areas; 3) whether there is statistical uncertainty in inputs or calculations within modeled CLs.

CLs are also built using different air quality surfaces and projections and this inherently introduces a degree of uncertainty in the CL estimates. This uncertainty manifests in several ways. If the air quality estimates have quantified uncertainties, but do not have a directional bias, then the CL should be considered an average estimate, with some range of uncertainty. However, if uncertainty in the air quality estimates is known to be biased, then the CL may be considered either a high or low estimate based on the directionality of the bias. For example, a CL based on air quality estimates that are known to be low should be considered the low end of the CL estimates, with a range of uncertainty that is greater than the estimate. An important component of uncertainty related to air quality with respect to CLs will be in the comparison of deposition surfaces developed in the REA to CLs developed with different air quality and how the uncertainties inherent in both the CL and the deposition surface relate to each other.

The following table (Table 4-2) summarizes the major sources of uncertainty for the CLs included in this plan based on the considerations outlined above.

Table 4-3. Major Sources of Uncertainty for the CLs Considered for the REA.

Critical Load	Consistent Methodology?	Extrapolation?	Quantified Statistical Uncertainty	Air Quality
Mycorrhizal fungi	No (multiple studies)	Yes	No	Multiple AQ inputs
Lichens	Yes within Root et al. (2015) and Geiser et al. (2010), some differences between.	Yes	No (in the NCLD) or Yes (source publications)	IMPROVE + CMAQ + NADP (Geiser et al. 2010), IMPROVE (Root et al. 2015)
Herbaceous species richness	Yes	No (point based) or Yes (ecoregion-based).	Yes	CMAQ + NADP
Forest soil acidification	Yes	No	No (in the NCLD) or Yes (source publications)	Not an input
Freshwater acidification	Yes	No	No (in the NCLD)	Not an Input
Freshwater N enrichment	Yes within Baron et al. (2011), Williams et al. (2017) and Nanus et al. (2017), different between.	Yes within Baron et al. (2011) and Williams et al. (2017). No within Nanus et al. (2017)	Yes within Williams et al. (2017) and Nanus et al. (2017). No within Baron et al. (2011)	PRISM+NADP+CMAQ (Baron et al. 2011), CMAQ (Williams et al. 2017), NADP+RMS+PRISM (wet) + TDEP (dry) (Nanus et al. 2017)

Given these considerations, we expect uncertainty for the mycorrhizal fungi dataset to be high as compared to the other datasets given that the data was built by combining the results from multiple studies and approaches, it includes extrapolation, and there is no explicit quantification of uncertainty included in the NCLD. Additionally, we expect uncertainty for the lichens dataset to be moderate given that data from two studies in Washington and Oregon were extrapolated nationally, and at least in the NCLD v.3 there is no inclusion of uncertainty. Within the areas/ecoregions of study, the uncertainty in the lichen CLs will be lower. For herbs/shrubs, we intend to use CLs from the Simkin et al (2016) study, which included an explicit quantification of uncertainty that was found to be low. In addition, researchers reported that the CLs from Simkin et al. (2016) were consistent with CLs derived from other studies. Therefore, there is high confidence in the data.

The CLs for freshwater N enrichment have differing uncertainties. We intend to apply the CLS for Baron et al. (2011) and Williams et al. (2017) across the study areas, which includes higher uncertainty due to extrapolation. In both datasets, we intend to also apply the CLs only to lakes included in the calculation of the CLs, which will reduce the uncertainty. Additionally, the Williams et al. (2017) study included quantification of uncertainty. We intend to apply the CLs from Nanus et al. (2017) across the study area. Similar to the Simkin et al. (2016) study, Nanus et al. (2017) included quantification of uncertainty as well as being consistent with CLs derived from other studies.

The CLs for freshwater and terrestrial acidification were modelled and empirical values which did not include air quality as a model input. Both datasets have lower levels of uncertainty based on the criteria discussed above, however, other uncertainties apply to these CLs which are discussed below.

An important component of uncertainty analysis is sensitivity analysis. Sensitivity analysis has been used extensively to look at critical load model parameters uncertainty ranges (Skeffington, 2006; Heywood et al., 2007; Fakhraei et al., 2016; Fakhraei et al., 2017). For example, Skeffington (2006) and Heywood et al. (2007) used a Monte Carlo probabilistic method to assess the degree of confidence in the CL exceedance and the coefficient of variation of the CL. They concluded that CL uncertainties are typically surprisingly small because of a “compensation of errors,” despite the input parameters often being poorly known. While natural variation and uncertainty in model parameters are important considerations, existing Monte Carlo sensitivity methods/tools are widely available, with many case study examples that can to be used in the REA to provide CL confidence, uncertainty, and CL exceedance probability to assess the risk to ecosystems from atmospheric N and/or S deposition. We propose using a probabilistic Monte Carlo approach to describe uncertainty and to describe the likelihood of the CL to be exceeded.

4.5.2.1 Freshwater and Terrestrial Acidification CLs

4.5.2.1.1 Freshwater

Freshwater ecosystems are complex and dynamic environmental systems and the impacts of ambient air pollution and the resulting deposition is influenced by a host of environmental processes that interact with the spatial variability of soils, geology, and biota. Given this complexity, CL models and even more complex biogeochemical models are an inevitable simplification of the natural system, which leads to inherent uncertainty in these assessments (Morgan and Henrion, 1990). The most noted simplification is the steady-state assumption for such dynamic ecosystems. Steady-state CLs are often derived from mathematical mass-balance models under assumed or modeled equilibrium conditions based on present water quality measurements. While steady-state CL models vary in complexity about the processes represented, a fundamental aspect of this approach is the steady-state assumption which rarely occurs in natural ecosystems since the ambient environment and atmosphere are continuously changing. Future weather modification may compound this limitation given that recovery of aquatic ecosystems from acidification will be affected by physical, chemical, and biological modification as annual mean temperature and precipitation levels lead to changes in runoff and bedrock and soil weathering (Greaver, 2016). Nevertheless, the steady-state assumption still provides an approximation that allows a quantitative risk assessment of atmospheric N and/S deposition impacts on aquatic ecosystems.

The models and their parameters used to determine aquatic CLs have been extensively examined over the past 30 years (e.g., Skeffington, 2006; Rapp and Bishop, 2009). In all cases, the strength of the aquatic CL estimate and the exceedance calculation relies on the ability of the method to estimate the watershed-average base cation supply (i.e., input of base cations from weathering of bedrock, soil and deposition), runoff volume, base cation and acidifying deposition, and NO_3^- leaching. Runoff volume and base cation deposition make smaller contributions to uncertainty as compared to base cation supply. It is the base cation supply that buffers the inputs of the acid anions from atmospheric deposition and is the factor that has the most influence on the CL calculation and the largest uncertainty (Skeffington, 2006; Li and McNulty, 2007; Fakrhaei et al., 2017).

Compounding the difficulty in estimating the base cation supply is the lack of methods to directly measure bedrock and soil weathering rates. In most cases, estimates of watershed-average base cation supply is determined indirectly by simple mass-balance differencing using present watershed water chemistry measurements or by more complex biogeochemical watershed models that still use water chemistry measurements as the bases for the determination. The CL is based on the present-day water chemistry and a model that drives the pre-industrial condition.

A host of different methods have been used to determine base cation supply, from more complex biogeochemical models (e.g., MAGIC, PnET-BGC, PROFILE) to simple models, such as the F-factor as part of the SSWC model (Hernriksen et al., 1990). While the importance of base cation supply to the estimate of CLs is well recognized, limited comparisons across models have been completed with the focus being on comparing dynamic biogeochemical models (Tominage et al., 2010). However, the use of dynamic biogeochemical models is more difficult because they require substantial input data, whereas simple models like the F-factor along with the SSWC model are often more widely used given the fewer inputs needed. Although the SSWC/F-factor critical load model has been widely published around the world (e.g., Henriksen and Posch et al., 2001; Dupont et al., 2005; Shaw et al., 2014), many model limitations have been noted. Rapp and Bishop (2009) extensively examined the F-factor approach and found key limitations related to the use of present-day water chemistry data to parameterize the F-factor. They found that water chemistry data from the period where the watershed is recovering from acidification can lead to uncertain estimates of the base cation supply. In addition, the F-factor was sensitive to short-term variation in the water chemistry data that reduced the confidence in the CL. However, the use of long-term averaged water chemistry data help to minimize this impact. Another limitation is the influence of increasing DOC concentrations in lakes and streams observed in recent water quality data (Monteith et al., 2007) and its impact on the accuracy of the F-factor value on the CL estimate, because DOC as a strong influence on the acid/base status of the water. The models used to derive the CLs in the NCLD v.3.0 vary from simple (e.g., SSWC or FAB) to more complex models (e.g., biogeochemical models); hence, we intend to complete a comprehensive comparison of CL models to account for their differences and biases.

4.5.2.1.2 Terrestrial

Given that we intend to update the soils acidification dataset and explore the use of PROFILE for purposes of estimating BCw rates, the level of uncertainty for this dataset it not yet known. However, we provide a summary below of the uncertainties that will be considered further in the REA. There are two main sources of uncertainty in the forest soil acidification CLs: the Bc/Al ratio as a chemical indicator and the BCw rates as an input into the SMB model. As mentioned earlier, the Bc/Al ratio is commonly used as an indicator because it relates well to the Ca/Al ratio. It should be noted, however, that while soil Bc/Al and Ca/Al ratios are recognized as good indicators of soil acidity risk in most U.S. forest ecosystems, these metrics are not static and the responses of sensitive receptors to such changes are variable depending on many outside factors. For example, the ratios themselves change depending on spatial scale and seasonal fluctuations in soil conditions. In addition, the methods for both sampling and laboratory analyses of base cations in soils can be inconsistent between studies as reported in Appendix 5 of the second draft ISA. For example, Bc/Al or Ca/Al ratios reported in studies are often based on seedling studies in controlled environments, and these responses are often less consistent for seedlings or trees growing outside such controlled environments.

As mentioned in Chapter 2, the 2009 NOx/SOx REA included the use of the clay substrate method for calculating BCw rates. The estimated BCw rates were identified in that review as a major source of uncertainty. Given the results of Phelan et al. (2014) in the Pennsylvania case study, we intend to explore the use of PROFILE modelling to obtain more accurate national estimates of BCw rates for the REA, considering uncertainties identified in Whitfield et al. (2018).

4.5.3 Exposure-Response Functions

The studies that have developed exposure-response functions (Horn et al. in review, Nanus et al. 2017, Geiser et al. 2010, Root et al. 2015, Simkin et al. 2016) all include some analysis of uncertainty and variability. For example, the functions were developed using air quality estimates averaged across several years. This potentially misses inter-annual variability in atmospheric deposition and introduces some uncertainty into the exposure -response function. Also, the functions in Horn et al (in review) were developed across the entire range of a given tree species. This potentially misses within species variability, which we intend to assess for a few important species. Finally, like CLs, an important component of uncertainty related to air quality with respect to exposure-response functions will be in the comparison of deposition surfaces developed in the REA to exposure-response functions developed with different air quality and how the uncertainties inherent in both the exposure-response function and the deposition surface relate to each other. Given these uncertainties and limitations for the exposure-response datasets, we will further consider their potential impact on the risk and exposure results in the REA.

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6 APPENDIX

Table 6-1. Results of Literature Search for Ecosystem Services Articles 2009 - 2016

Author	Short Title	Location Notes
Amiraslany et al. (2012)	Economic value of fire conditions and the effects of wildfire on hiking	NM
Beier et al (2017)	Loss of ecosystem services due to chronic pollution	Adirondacks
Caputo et al. (2017)	Impacts of acidification and potential recovery recreational fisheries	Adirondacks
Caputo et al. (2016)	Effects of soil acidification on long-term ecological and economic outcomes	Adirondacks
Christman et al. (2014)	Willingness to Pay to Reduce Wild Fire Risk in Wildland-Urban Interface	NV
Englin et al (2008)	Wildfire and the Economic Value of Wilderness Recreation.	CA
Holmes et al. (2013)	The effects of personal experience on preferences for wildfire protection	FL
Huang et al. (2013)	Toward full economic valuation of forest fuels-reduction treatments.	AZ
Kaval et al. (2008)	Using GIS to test homeowner willingness to pay to reduce wildfire	CO
Loomis et al. (2008)	Contingent valuation of fuel hazard reduction treatments.	CA, FL
Loomis et al. (2009)	Willingness to pay function for fuel treatments to reduce wildfire acreage	CA, FL
Loomis et al. (2010)	Forest Service Use of Nonmarket Valuation in Fire Economics	US Wide
Loomis et al. (2015)	Are WTP Estimates for Wildfire Risk Reductions Transferrable	CA, FL
Mueller et al. (2009)	Hedonic Analysis of the Short and Long-Term Effects of Repeated Wildfires	Southern CA
Sanchez et al. (2016)	Valuing hypothetical wildfire impacts on recreation demand.	Southern CA
Stetler et al. (2010)	The effects of wildfire and environmental amenities on property values	MT
Azevedo et al. (2015)	Willingness to pay for Clear Lake cleanup.	Clear Lake, IA
Banzhaf et al. (2016)	Policy Analysis: Valuation of Ecosystem Services in the S. Appalachian Mnts	Appalachians
Bin et al. (2013)	Impact of Measures of Water Quality on Coastal Waterfront Property Values	South FL
Carter et al. (2012)	The economic value of catching and keeping or releasing saltwater sport fish	Southeast US
Cho et al. (2011)	Negative externalities on property values resulting from water impairment	NC
Egan et al. (2009)	Valuing Water Quality as a Function of Water Quality Measures	IA
Foster (2008)	Valuing preferences for water quality improvement	FL
Freeman et al (2008)	Statistical analysis of drinking water treatment plant costs	Worldwide
Guignet et al. (2014)	The Implicit Price of Aquatic Grasses	Chesapeake Bay
Hindlesy et al. (2011)	Addressing onsite sampling in recreation site choice models	Southeastern US
Huang et al. (2010)	Quantifying the economic effects of hypoxia on a fishery for brown shrimp	NC

Author	Short Title	Location Notes
Huang et al. (2011)	Management of an annual fishery in the presence of ecological stress	Southeast
Jackson et al. (2012)	Economic value of stream degradation across the central Appalachians	MD, PA,VA,WV
Jacobsen et al. (2009)	Income Effects on Global Willingness to Pay for Biodiversity Conservation	Worldwide
Ji et al. (2016)	Water-based Recreation/Water Quality Indices Revealed Preference Approach	IA
Keeler et al. (2013)	Advances in measuring the value of water quality to people	MN, IA
Keeler et al. (2015)	Recreational demand for clean water	MN, IA
Liao et al. (2016)	The Effects of Ambient Water Quality and watermilfoil on Property Values	ID
Liu et al. (2014)	Estimating the impact of water quality on surrounding property values	OH
Martín-López (2008)	Economic Valuation of Biodiversity Conservation: the Meaning of Numbers	Worldwide
Melstrom et al. (2013)	Valuing recreational fishing in the Great Lakes	Great Lakes
Meyer (2012)	Intertemporal valuation of river restoration	MN
Moore et al. (2015)	A stated preference study of the Chesapeake Bay and watershed lakes	Chesapeake Bay
Morgan et al. (2010)	Water Quality and Residential Property Values: A Natural Experiment	VA
Nelson et al. (2015)	Estimate the total economic value of improving water quality	UT
Netusil et al. (2014)	Valuing water quality in urban watersheds	OR and WA
Ramachandran et al. (2015)	Validating Spatial Hedonic Modeling with a Behavioral Approach	Cape Cod, MA
Richardson et al. (2009)	The total economic value of threatened, endangered and rare species	Worldwide
Roberts et al. (2008)	Preferences for environmental quality under uncertainty	Tulsa, OK
Tuttle et al. (2011)	Hedonic Studies: Valuing Environmental Amenities in Northern New York.	Northern NY
Tuttle et al. (2015)	A hedonic analysis of lake water quality in the Adirondacks	Adirondacks
Van Houtven et al. (2014)	Expert elicitation and stated preference methods to value ecosystem service	VA
Viscusi et al. (2008)	The economic value of water quality	US Wide
Walsh et al. (2010)	Spatial Extent of Water Quality Benefits in Urban Housing Markets	FL
Walsh et al. (2011)	The spatial extent of water quality benefits in urban housing markets	FL
Walsh et al. (2015b)	Modeling the Property Price Impact of Water Quality	Chesapeake Bay
Welle et al. (2008)	Property owners' willingness to pay for restoring impaired waters	MN
Welle et al. (2011)	Property owners' willingness to pay for water quality improvements	MN
Phaneuf et al. (2013)	Measuring nutrient reduction benefits for policy analysis	Southeastern US

FULL CITATIONS FROM Table 6-1

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