

# **Integrated Science Assessment for Oxides of Nitrogen, Oxides of Sulfur, and Particulate Matter— Ecological Criteria**

**(Final)**

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## **Disclaimer**

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# EXECUTIVE SUMMARY

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## ES.1 Purpose and Scope of the Integrated Science Assessment

This *Integrated Science Assessment (ISA) for Oxides of Nitrogen, Oxides of Sulfur, and Particulate Matter—Ecological Criteria* is a comprehensive evaluation and synthesis of the most policy-relevant science aimed at characterizing the ecological effects caused by these criteria pollutants.<sup>1</sup> These criteria pollutants are reviewed here together because they all contribute to nitrogen (N) and sulfur (S) deposition, which causes substantial ecological effects. In this document, the term “oxides of nitrogen” refers to total oxidized N ( $\text{NO}_Y$ ), including nitric oxide (NO) and nitrogen dioxide ( $\text{NO}_2$ ) and all other gaseous and particulate oxidized N containing compounds formed from NO and  $\text{NO}_2$ .<sup>2</sup> Total sulfur oxides ( $\text{SO}_X$ ) includes gaseous chemical species (e.g., sulfur dioxide [ $\text{SO}_2$ ], sulfur monoxide [SO], disulfur monoxide [ $\text{S}_2\text{O}$ ], and sulfur trioxide [ $\text{SO}_3$ ]) as well as particulate species, such as ammonium sulfate [ $(\text{NH}_4)_2\text{SO}_4$ ] ([U.S. EPA, 2011](#)). Particulate species include  $\text{SO}_X$  species like sulfites ( $\text{SO}_3^{2-}$ ) and sulfates ( $\text{SO}_4^{2-}$ ), but among these two species usually only  $\text{SO}_4^{2-}$  make a major contribution to particulate mass. Throughout this document  $\text{SO}_X$  is defined as the sum of  $\text{SO}_2$  and particulate sulfate ( $\text{SO}_4^{2-}$ ), which together represent virtually all of the  $\text{SO}_X$  mass in the atmosphere.<sup>3</sup> Particulate matter (PM) is composed of some or all of the following components: nitrate ( $\text{NO}_3^-$ ),  $\text{SO}_4^{2-}$ , ammonium ( $\text{NH}_4^+$ ), metals, minerals (dust), and organic and elemental carbon.

This ISA serves as the scientific foundation for the review of the ecological effects associated with the secondary (welfare-based) National Ambient Air Quality Standards (NAAQS) for  $\text{NO}_Y$ ,  $\text{SO}_X$ , and PM. The health effects of these criteria pollutants are considered in separate assessments for  $\text{NO}_Y$  ([U.S. EPA, 2016b](#)),  $\text{SO}_X$  ([U.S. EPA, 2016a](#)), and PM ([U.S. EPA, 2019](#)).<sup>4</sup> The Clean Air Act definition of welfare effects includes, but is not limited to, effects on soils, water, wildlife, vegetation, visibility, weather, and

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<sup>1</sup> The general process for developing an ISA, including the framework for evaluating weight of evidence and drawing scientific conclusions and causal judgments, is described in a companion document, *Preamble to the Integrated Science Assessments* ([U.S. EPA, 2015](#)), [www.epa.gov/isa](http://www.epa.gov/isa).

<sup>2</sup> This ISA reserves the abbreviation  $\text{NO}_X$  strictly as the sum of NO and  $\text{NO}_2$ —consistent with its use in the atmospheric science community—and uses the term “oxides of nitrogen” to refer to the broader list of oxidized nitrogen species. Oxides of nitrogen refers to  $\text{NO}_Y$  as the total oxidized nitrogen in both gaseous and particulate forms. The major gaseous and particulate constituents of  $\text{NO}_Y$  include nitric oxide (NO), nitrogen dioxide ( $\text{NO}_2$ ), nitric acid ( $\text{HNO}_3$ ), peroxyacetyl nitrate (PAN), nitrous acid (HONO), organic nitrates, and particulate nitrate ( $\text{NO}_3^-$ ). This ISA uses the definitions adopted by the atmospheric sciences community.

<sup>3</sup> The same definition of  $\text{SO}_X$  used in the 2011  $\text{NO}_X\text{SO}_X$  Policy Assessment ([U.S. EPA, 2011](#)).

<sup>4</sup> In this ISA, the blue electronic links can be used to navigate to cited materials as well as appendices, sections, tables, figures, and studies from this ISA.

climate, as well as effects on man-made materials, economic values, and personal comfort and well-being.

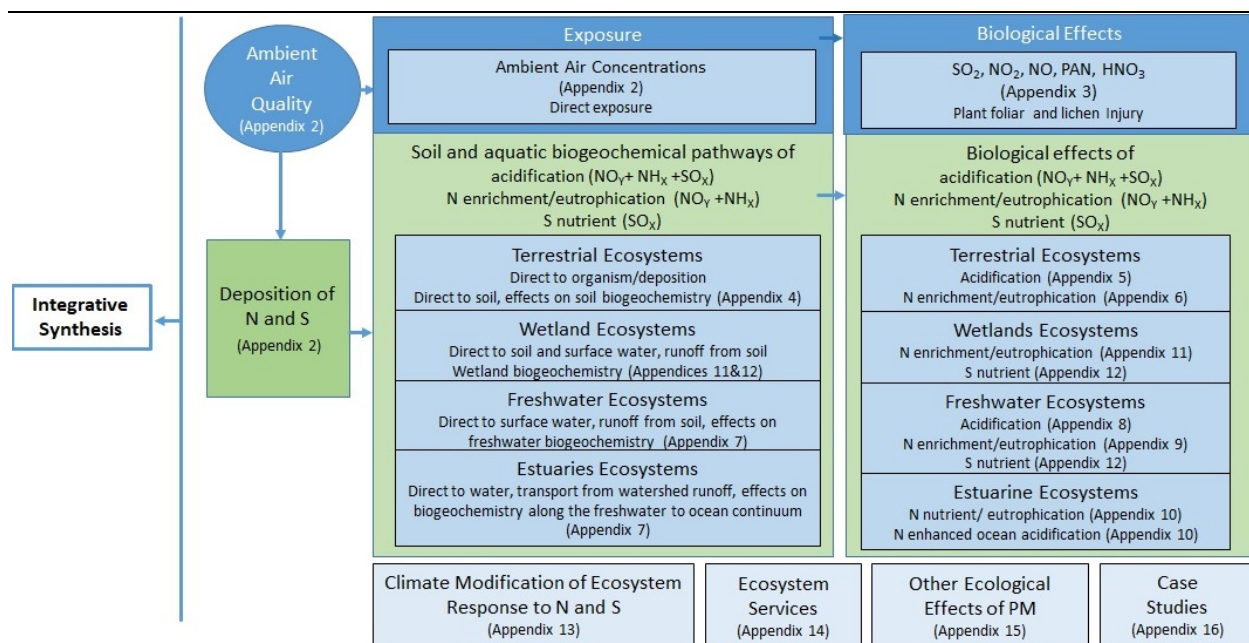
The current secondary NAAQS for NO<sub>Y</sub> and SO<sub>X</sub> were set to protect against direct damage to vegetation by NO<sub>2</sub> or SO<sub>2</sub>. The secondary NAAQS for NO<sub>2</sub> is identical to the primary standard set in 1971: an annual average not to exceed 0.053 ppm N dioxide. The secondary NAAQS for SO<sub>2</sub>, set in 1973, is a 3-hour average of 0.5 ppm SO<sub>2</sub>, not to be exceeded more than once per year. The current secondary standards for PM are intended to address PM-related visibility and nonvisibility welfare effects. These standards are a 3-year annual mean PM<sub>2.5</sub> concentration of 15 µg/m<sup>3</sup>, with the 24-hour average PM<sub>2.5</sub> and PM<sub>10</sub> set at concentrations of 35 µg/m<sup>3</sup> and 150 µg/m<sup>3</sup>, respectively.

This ISA updates the 2008 *ISA for Oxides of Nitrogen and Oxides of Sulfur—Ecological Criteria* [hereafter referred to as 2008 ISA ([U.S. EPA, 2008](#))], as well as the ecological portion of the 2009 *ISA for Particulate Matter* ([U.S. EPA, 2009](#)) with studies and reports published from January 2008 through May 2017. There are some studies included that were published more recently than the May 2017 literature cutoff date; these studies were added based on recommendations from the Clean Air Scientific Advisory Committee (CASAC). The U.S. EPA conducted in-depth searches to identify peer-reviewed literature on relevant topics. Subject-matter experts and the public were also able to recommend studies and reports during a kick-off workshop held by the U.S. EPA in March 2014 for NO<sub>Y</sub> and SO<sub>X</sub> and in June 2016 for PM. CASAC recommended the inclusion of additional studies during the review of the first draft. To fully describe the state of available science, the U.S. EPA also carried over the most relevant studies from previous assessments to include in this ISA.

This ISA determines whether NO<sub>Y</sub>, SO<sub>X</sub>, and PM concentrations in the air or deposition from the air cause ecological effects. The ecological effects of deposition are grouped into three main categories: (1) acidification (caused by gaseous NO<sub>Y</sub>, SO<sub>X</sub>, and particulate NH<sub>4</sub><sup>+</sup>, NO<sub>3</sub><sup>-</sup>, SO<sub>4</sub><sup>2-</sup>), (2) N enrichment/N driven eutrophication (caused by gaseous NO<sub>Y</sub> and particulate NH<sub>4</sub><sup>+</sup> and NO<sub>3</sub><sup>-</sup>), and (3) S enrichment (caused by SO<sub>X</sub> and particulate forms of SO<sub>4</sub><sup>2-</sup>). Ecological effects are further subdivided into terrestrial, wetland, freshwater, and estuarine/near-coastal ecosystems. These ecosystems and effects are linked by the connectivity of terrestrial and aquatic habitats through biogeochemical pathways of N and S.

A schematic of the document organization is given by [Figure ES-1](#). The Integrated Synthesis (IS) brings together key information on specific subject matter found in the appendices. Appendix 1 is an introduction to the purpose and organization of the material covered in Appendix 2–Appendix 16. Appendix 2 characterizes the sources and atmospheric processes involving NO<sub>Y</sub>, SO<sub>X</sub>, and PM, as well as trends in ambient

concentrations and deposition. Appendix 3 describes direct effects of gas-phase  $\text{NO}_y$  and  $\text{SO}_x$  on plants and lichens. Appendix 4 describes N and S deposition effects on terrestrial biogeochemistry, and Appendix 5 and Appendix 6 describe the biological effects of terrestrial acidification and terrestrial N enrichment, respectively. Appendix 7 describes N and S deposition effects on aquatic biogeochemistry. Appendix 8 through Appendix 10 characterize the biological effects of freshwater acidification, freshwater N enrichment, and marine eutrophication, respectively. Appendix 11 describes the effects of N deposition on wetlands. Appendix 12 describes the wetland and freshwater effects of S enrichment. Appendix 13 discusses the climate modification of ecosystem response to N and S deposition, and Appendix 14 presents information on N and S deposition effects on ecosystem services. Information on the ecological effects of forms of PM beyond those related to N or S deposition is presented in Appendix 15 (the nonecological welfare effects associated with PM, such as visibility, climate, and material effects, are considered as part of a separate review of PM [81 FR 87933, December 6, 2016]). Appendix 16 includes six locations in the U.S. selected as case study areas that are candidates for additional analysis of risk and exposure. These candidate sites were selected because they have abundant data on ecological effects.



HNO<sub>3</sub> = nitric acid; N = nitrogen; NH<sub>x</sub> = reduced nitrogen; NO = nitric oxide; NO<sub>2</sub> = nitrogen dioxide; NO<sub>y</sub> = nitrogen oxides; PAN = peroxyacetyl nitrate; PM = particulate matter; S = sulfur; SO<sub>2</sub> = sulfur dioxide; SO<sub>x</sub> = sulfur oxides.

**Figure ES-1 Roadmap of the Integrated Science Assessment (ISA) linking atmospheric concentrations and deposition, soil and aquatic biogeochemistry, and biological effects.**

## ES.2 Emissions, Ambient Air Concentrations, and Deposition

The atmospheric chemistry from emission to deposition discussed in this ISA<sup>1</sup> is for the criteria pollutants NO<sub>y</sub>, SO<sub>x</sub>, and PM. NO<sub>y</sub> and SO<sub>x</sub> cause ecological effects in the gas phase and/or after N and S deposition to surfaces. Particulate matter (PM) effects discussed in this document focus on N and S containing species, which together usually make up a large fraction of the PM<sub>2.5</sub> mass in most areas of the U.S. NH<sub>x</sub> (NH<sub>x</sub> = NH<sub>3</sub> + NH<sub>4</sub><sup>+</sup>) includes both gas-phase NH<sub>3</sub> and the PM component NH<sub>4</sub><sup>+</sup>. NH<sub>3</sub> is estimated to account for 19–63% of total observed inorganic N deposition, depending on region (Appendix 2.1). Therefore, NH<sub>3</sub> is discussed in this ISA along with NO<sub>y</sub> and relevant PM components to better understand and compare their contributions to both wet and dry N deposition.

<sup>1</sup> The term concentration is used throughout the ISA to denote either a mass per unit volume or a volume per unit volume (mixing ratio). The use of concentration to denote abundance expressed as mixing ratio is firmly entrenched in the literature; therefore, it is retained here.

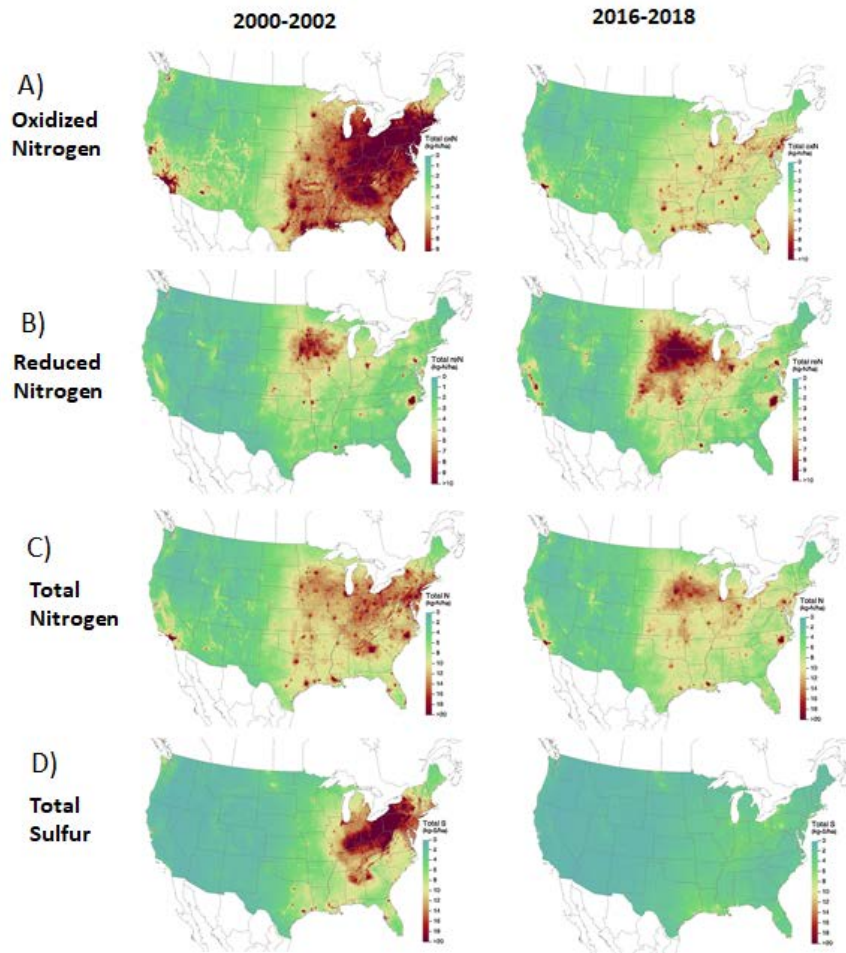
Both gaseous and particulate forms of  $\text{NO}_Y$ ,  $\text{SO}_X$ , and  $\text{NH}_X$  contribute to atmospheric wet and dry deposition. The major components of PM in the U.S. are  $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$ ,  $\text{NH}_4^+$ , organic matter, elemental C, crustal material, and sea salt. Of these,  $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$ , and  $\text{NH}_4^+$  usually have a strong influence on acid deposition, and  $\text{NO}_3^-$  and  $\text{NH}_4^+$ , and in some cases organic N (organic nitrates and reduced organic N), contribute substantially to N deposition and eutrophication.

The sources and precursors to gaseous and particulate forms of  $\text{NO}_Y$ ,  $\text{SO}_X$ , and  $\text{NH}_X$  vary. The main contributors to acidifying precipitation are formed from precursor emissions of the gases  $\text{SO}_2$ ,  $\text{NO}_X$ , and  $\text{NH}_3$  (Appendix 2.2). Electricity-generating units (EGUs) are the source of about half of national gaseous emissions of  $\text{SO}_2$ , mainly from coal-fired power plants. Notably,  $\text{SO}_2$  emissions from EGUs have been decreasing.  $\text{NO}_X$  emissions have a wider distribution of sources, with substantial contributions from highway and off-highway vehicles, lightning, and EGUs. Fertilizer application and animal waste are the main national-scale sources of  $\text{NH}_3$ , with animal waste contributing the most. Primary  $\text{PM}_{2.5}$  and  $\text{PM}_{10}$  emissions are dominated by dust and combustion products of fires, but much of the  $\text{PM}_{2.5}$  mass in the U.S. is produced by reaction of gas-phase precursors to form secondary  $\text{PM}_{2.5}$ . In this process, particulate  $\text{NH}_4^+$ ,  $\text{NO}_3^-$ , and  $\text{SO}_4^{2-}$  are primarily derived from the gaseous precursors  $\text{NH}_3$ ,  $\text{NO}_X$ , and  $\text{SO}_2$  (Appendix 2.3). Formation of particulate N and S is described in the 2019 *ISA for Particulate Matter* ([U.S. EPA, 2019](#)). An understanding of the sources, chemistry, and atmospheric processes for these gas-phase and PM species is necessary to understand acidifying and N deposition.

Since the passage of the Clean Air Act Amendments in 1990, the emissions of  $\text{NO}_X$  and  $\text{SO}_2$  have declined dramatically. Total emissions of  $\text{SO}_2$  decreased by 89% from 1990 to 2017, resulting in a decrease in  $\text{SO}_2$  concentrations of 89% in the eastern U.S. and 45% in the western U.S. Emissions of  $\text{NO}_X$  in the U.S. declined by 61% between 1990 and 2017, while nationwide annual average 98th percentile  $\text{NO}_2$  concentrations decreased by 53% from 1990 to 2017. These reductions have in turn led to decreases in  $\text{PM}_{2.5}$  concentrations because of declines in the amount of  $\text{SO}_4^{2-}$  and  $\text{NO}_3^-$  produced, and a decrease in the fraction of  $\text{PM}_{2.5}$  accounted for by  $\text{SO}_4^{2-}$ . Between 1989 and 2017, average particulate  $\text{SO}_4^{2-}$  concentration decreased by 75% in the eastern U.S. and 35% in the western U.S., and average particulate  $\text{NO}_3^-$  concentration decreased by 51% in the eastern U.S. and 37% in the western U.S.

Averaged across the contiguous U.S., deposition of total N (oxidized + reduced N, in kg N/ha/yr) has changed only slightly since 2000 (Appendix 2.6.2). [Figure ES-2](#) shows that between 2000 and 2018 large decreases in oxidized nitrogen ([Figure ES-2A](#)) have combined with large increases in reduced nitrogen deposition ([Figure ES-2B](#)) to produce a small decrease in total nitrogen deposition ([Figure ES-2C](#)). There is large spatial

variability in N deposition over the contiguous U.S. ([Figure ES-2C](#)). According to National Atmospheric Deposition Program Total Deposition Committee's (TDEP's) estimates for 2016–2018 (Appendix 2.6.2), much of the eastern U.S. is estimated to receive at least 10 kg N/ha/yr dry + wet deposition, with some areas receiving more than 15 kg N/ha/yr. [Figure ES-2](#) A through C shows that between 2000 and 2018, large decreases in oxidized nitrogen deposition occurred.



Ha = hectare; kg = kilogram; N = nitrogen; OxN = oxidized nitrogen; ReN = reduced nitrogen; S = sulfur.

Source: We acknowledge the Total Deposition (TDep) Science Committee of the National Atmospheric Deposition Program (NADP) for their role in making the TDep data and maps available.

**Figure ES-2** Wet plus dry deposition of (A) oxidized nitrogen, (B) reduced nitrogen, (C) total nitrogen, and (D) total sulfur over the 3-year periods 2000–2002 and 2016–2018.



For S, wet deposition tends to dominate over dry deposition in large areas of the contiguous U.S. However, in some regions, mostly in the west, dry deposition of mainly SO<sub>2</sub> is greater than wet deposition. Anthropogenic emissions of S and subsequent deposition have declined markedly since the 1990s, with the most pronounced declines in the eastern U.S., as shown in [Figure ES-2D](#). Currently, the highest values of total (wet + dry) SO<sub>x</sub> deposition in the U.S. are in parts of the Ohio Valley region where they range between 15 and 20 kg S/ha/yr.

Both N and S deposition contribute to acidification of ecosystems. The acidity of rainwater has decreased, as indicated by the increase of rainwater pH across the U.S. since 1990, coincident with decreases in the wet deposition of nitrate and sulfate. However, widespread areas are still affected by acidifying precipitation, mainly in the eastern U.S. (see Appendix 2.6.1). Total acidifying deposition (wet + dry N + S, expressed as H<sup>+</sup> equivalents) fluxes for 2016 to 2018 ranged from a few tenths of H<sup>+</sup> keq/ha/yr over much of the western U.S. to over 1.5 H<sup>+</sup> keq/ha/yr in parts of the Midwest and the Mid-Atlantic regions, and in other isolated hotspots surrounding areas of concentrated industrial or agricultural activity (Figure IS-6). Estimated deposition fluxes greater than 1.5 keq/ha/yr covered a much smaller portion of the U.S. in 2016–2018 than in 2000–2002.

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### **ES.3 Ecological Effects**

In this ISA, information on ecological effects from controlled exposure, field addition, ambient deposition, and toxicological studies, among others, are integrated to form conclusions about the causal nature of relationships between NO<sub>y</sub>, SO<sub>x</sub>, and PM and ecological effects. Studies on the ecological effects are considered in relation to a range of ambient concentration and deposition loads that are within two orders of magnitude from current conditions [Preamble ([U.S. EPA, 2015](#)), Section 5c]. A consistent and transparent framework [Preamble ([U.S. EPA, 2015](#)), Table II] is applied to classify the ecological effect evidence according to a five-level hierarchy:

1. Causal relationship
2. Likely to be a causal relationship
3. Suggestive of, but not sufficient to infer, a causal relationship
4. Inadequate to infer a causal relationship
5. Not likely to be a causal relationship

The conclusions presented in [Table ES-1](#) are based on recent findings integrated with information from the 2008 ISA ([U.S. EPA, 2008](#)). The conclusions of [Table ES-1](#) are based on careful consideration of errors and uncertainty in the supporting studies. We

also consider the coherence of findings integrated across studies of underlying geochemical and biological mechanisms. There are 18 causality statements in this ISA (Table ES-1). Fourteen are causal relationships repeated from the 2008 ISA or modified from the 2008 ISA to include specific endpoints. For these causality statements, new research strengthens the evidence base and is consistent with the 2008 ISA. There is one likely causal relationship repeated from the 2009 ISA for Particulate Matter. Three causal relationships are new endpoint categories not evaluated in the 2008 ISA. Although NO<sub>y</sub> and SO<sub>x</sub> can cause phytotoxic injury, current monitored concentrations of gas-phase NO<sub>y</sub> and SO<sub>x</sub> are not high enough to injure vegetation. For all other identified causal relationships identified in this ISA, the evidence indicates a causal association from current levels of S and/or N deposition.

**Table ES-1 Causal determinations for relationships between criteria pollutants and ecological effects from the 2008 NO<sub>x</sub>/SO<sub>x</sub> Integrated Science Assessment (ISA) or the 2009 ISA for Particulate Matter (PM), for other effects of PM, and the current draft ISA.**

Effect Category	Causal Determination	
	2008 NO <sub>x</sub> /SO <sub>x</sub> ISA	Current Draft ISA
<b>Gas-phase direct phytotoxic effects</b>		
Gas-phase SO <sub>2</sub> and injury to vegetation Section IS.3 and Appendix 3.6.1	Causal relationship	Causal relationship
Gas-phase NO, NO <sub>2</sub> , and PAN and injury to vegetation Section IS.3 and Appendix 3.6.2	Causal relationship	Causal relationship
Gas-phase HNO <sub>3</sub> and injury to vegetation <sup>a</sup> Section IS.3 and Appendix 3.6.3	Causal relationship	Causal relationship
<b>N and acidifying deposition to terrestrial ecosystems</b>		
N and S deposition and alteration of soil biogeochemistry in terrestrial ecosystems <sup>b</sup> Section IS.5.1 and Appendix 4.1	Causal relationship	Causal relationship
N deposition and the alteration of the physiology and growth of terrestrial organisms and the productivity of terrestrial ecosystems <sup>c</sup> Section IS.5.2 and Appendix 6.6.1	Not included	Causal relationship
N deposition and the alteration of species richness, community composition, and biodiversity in terrestrial ecosystems <sup>c</sup> Section IS.5.2 and Appendix 6.6.2	Causal relationship	Causal relationship

**Table ES-1 (Continued): Causal determinations for relationships between criteria pollutants and ecological effects from the 2008 NO<sub>x</sub>/SO<sub>x</sub> Integrated Science Assessment (ISA) or the 2009 ISA for Particulate Matter (PM), for other effects of PM, and the current draft ISA.**

Effect Category	Causal Determination	
	2008 NO <sub>x</sub> /SO <sub>x</sub> ISA	Current Draft ISA
Acidifying N and S deposition and the alteration of the physiology and growth of terrestrial organisms and the productivity of terrestrial ecosystems <sup>d</sup> Section IS.5.3 and Appendix 5.7.1	Not included	Causal relationship
Acidifying N and S deposition and the alteration of species richness, community composition, and biodiversity in terrestrial ecosystems <sup>d</sup> Section IS.5.3 and Appendix 5.7.2	Causal relationship	Causal relationship
<b>N and acidifying deposition to freshwater ecosystems</b>		
N and S deposition and alteration of freshwater biogeochemistry <sup>e</sup> Section IS.6.1 and Appendix 7.1.7	Causal relationship	Causal relationship
Acidifying N and S deposition and changes in biota, including physiological impairment and alteration of species richness, community composition, and biodiversity in freshwater ecosystems <sup>f</sup> Section IS.6.3 and Appendix 8.6	Causal relationship	Causal relationship
N deposition and changes in biota, including altered growth and productivity, species richness, community composition, and biodiversity due to N enrichment in freshwater ecosystems <sup>g</sup> Section IS.6.2 and Appendix 9.6	Causal relationship	Causal relationship
<b>N deposition to estuarine ecosystems</b>		
N deposition and alteration of biogeochemistry in estuarine and near-coastal marine systems Section IS.7.1 and Appendix 7.2.10	Causal relationship	Causal relationship
N deposition and changes in biota, including altered growth, total primary production, total algal community biomass, species richness, community composition, and biodiversity due to N enrichment in estuarine environments <sup>h</sup> Section IS.7.2 and Appendix 10.7	Causal relationship	Causal relationship
<b>N deposition to wetland ecosystems</b>		
N deposition and the alteration of biogeochemical cycling in wetlands Section IS.8.1 and Appendix 11.10	Causal relationship	Causal relationship

**Table ES-1 (Continued): Causal determinations for relationships between criteria pollutants and ecological effects from the 2008 NO<sub>x</sub>/SO<sub>x</sub> Integrated Science Assessment (ISA) or the 2009 ISA for Particulate Matter (PM), for other effects of PM, and the current draft ISA.**

Effect Category	Causal Determination	
	2008 NO <sub>x</sub> /SO <sub>x</sub> ISA	Current Draft ISA
N deposition and the alteration of growth and productivity, species physiology, species richness, community composition, and biodiversity in wetlands Section IS.8.2 and Appendix 11.10	Causal relationship	Causal relationship
<b>S deposition to wetland and freshwater ecosystems</b>		
S deposition and the alteration of mercury methylation in surface water, sediment, and soils in wetland and freshwater ecosystems <sup>i</sup> Section IS.9.1 and Appendix 12.7	Causal relationship	Causal relationship
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 Section IS.9.2 and Appendix 12.7	Not included	Causal relationship
	2009 PM ISA	Current Draft ISA
<b>Other ecological effects of PM</b>		
PM and a variety of effects on individual organisms and ecosystems Section IS.10 and Appendix 15.8	Likely to be a causal relationship	Likely to be a causal relationship

C = carbon; Hg = mercury; HNO<sub>3</sub> = nitric acid; ISA = Integrated Science Assessment; N = nitrogen; NO = nitric oxide; NO<sub>2</sub> = nitrogen dioxide; PAN = peroxyacetyl nitrate; S = sulfur; SO<sub>2</sub> = sulfur dioxide.

<sup>a</sup>The 2008 ISA causality statements for gas-phase HNO<sub>3</sub> was phrased as “changes in vegetation.”

<sup>b</sup>The 2008 ISA included two causality statements for terrestrial biogeochemistry phrased as “relationship between acidifying deposition and changes in biogeochemistry” and “relationship between N deposition and the alteration of biogeochemical cycling of N.”

<sup>c</sup>The 2008 ISA causality statement for biological effects of N enrichment in terrestrial ecosystems was phrased as “relationship between N deposition and the alteration of species richness, species composition, and biodiversity.”

<sup>d</sup>The 2008 ISA causality statement for biological effects of acidifying deposition in terrestrial ecosystems was phrased as “relationship between acidifying deposition and changes in terrestrial biota.”

<sup>e</sup>The 2008 ISA included three causality statements for freshwater biogeochemistry phrased as “relationship between acidifying deposition and changes in biogeochemistry related to aquatic ecosystems,” “relationship between N deposition and the alteration of biogeochemical cycling of N,” and “relationship between N deposition and the alteration of biogeochemical cycling of C.”

<sup>f</sup>The 2008 ISA causality statement for biological effects of acidifying deposition in freshwater ecosystems was phrased as “relationship between acidifying deposition and changes in aquatic biota.”

<sup>g</sup>The 2008 ISA causality statement for biological effects of N deposition in freshwater ecosystems was phrased as “relationship between N deposition and the alteration of species richness, species composition, and biodiversity in freshwater aquatic ecosystems.”

<sup>h</sup>The 2008 ISA causality statement for biological effects of N deposition to estuaries was phrased as “relationship between N deposition and the alteration of species richness, species composition, and biodiversity in estuarine ecosystems.”

<sup>i</sup>The 2008 ISA causality statement for biological effects of S deposition effects on ecosystems was phrased as “relationship between S deposition and increased methylation of Hg, in aquatic environments where the value of other factors is within adequate range for methylation.”

[Figure ES-3](#) presents a visualization of the causality statements integrated into a single diagram. There is not a one-to-one correspondence between the number of causality statements, of which there are 18, and the cells indicated to have causal relationships in the diagram because some causal statements include effects across more than one level of biological organization. The main findings are that gaseous  $\text{NO}_y$  and  $\text{SO}_x$  cause phytotoxic effects, while N and S deposition cause alteration in (1) biogeochemical components of soil and water chemistry and (2) multiple levels of biological organization ranging from physiological processes to shifts in biodiversity and ecological function.

NOx SOx PM Integrated Science Assessment for Ecological Effects*														
Indicator			Gases ‡	Nitrogen Deposition				Sulfur Deposition				Nitrogen and Sulfur Deposition		
Class of Pollutant Effect			Direct Phytotoxic	N-enrichment/Eutrophication				Sulfide Toxicity		Mercury Methylation		Acidification		
Ecosystem			Terrestrial	Terrestrial	Wetland	Fresh Water	Estuary	Wetland	Fresh Water	Wetland	Fresh Water	Terrestrial	Fresh Water	
Scale of Ecological Response	Ecosystem	Productivity	Not evaluated in causal framework	Causal	Causal	Causal	Causal	Causal	Causal	Not evaluated in causal framework	Not evaluated in causal framework	Causal	Not evaluated in causal framework	
		Community	Not evaluated in causal framework	Causal	Causal	Causal	Causal	Causal	Causal	Causal	Causal	Causal	Causal	
	Population	Individual	Not evaluated in causal framework	Causal	Causal	Causal	Causal	Causal	Causal	Causal	Causal	Causal	Not evaluated in causal framework	
		Individual	Causal	Causal	Causal	Not evaluated in causal framework	Not evaluated in causal framework	Causal	Causal	Not evaluated in causal framework	Not evaluated in causal framework	Causal	Causal	
	Geochemistry	Soil or sediment chemistry	Not evaluated in causal framework	Causal	Causal	Not evaluated in causal framework	Not evaluated in causal framework	Not evaluated in causal framework	Not evaluated in causal framework	Not evaluated in causal framework	Causal	Causal	Causal	Not evaluated in causal framework
		Surface water chemistry	Not evaluated in causal framework	Not evaluated in causal framework	Causal	Causal	Causal	Causal	Not evaluated in causal framework	Not evaluated in causal framework	Causal	Causal	Not evaluated in causal framework	Causal

**Causality framework**

Causal
Likely causal
Suggestive
Inadequate
Not likely
Not evaluated in causal framework

\* A causal relationship is likely to exist between deposition of PM and a variety of effects on individual organisms and ecosystems, based on information from the previous review and limited new findings in this review

‡ Includes: NO, NO<sub>2</sub>, HNO<sub>3</sub>, SO<sub>2</sub>, and PAN

**Figure ES-3 Causal relationships between the criteria pollutants and ecological effects.**

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**ES.4 Direct Phytotoxic Effects of Gas-Phase Oxides of Nitrogen (NO<sub>Y</sub>) and Oxides of Sulfur (SO<sub>X</sub>)**

The current NO<sub>2</sub> and SO<sub>2</sub> secondary NAAQS are set to protect against direct damage to vegetation by exposure to gas-phase oxides of NO<sub>Y</sub> and SO<sub>X</sub>. NH<sub>3</sub> can also have direct phytotoxic effects, but reduced N gases such as NH<sub>3</sub> are not criteria air pollutants. Research continues to support causal relationships between SO<sub>2</sub>, NO<sub>2</sub>, NO, peroxyacetyl nitrate (PAN), HNO<sub>3</sub>, and injury to vegetation (e.g., visible foliar injury, damage to photosynthesis, decline of growth and abundance; (Table IS-1, Section IS.4, Appendix 3), but research that tests plant response to the lower exposure levels representative of current atmospheric NO<sub>Y</sub> and SO<sub>X</sub> concentrations is limited. Consequently, few studies are available to help determine whether current monitored concentrations of gas-phase NO<sub>Y</sub> and SO<sub>X</sub> are high enough to injure vegetation. It is also known that these can be gases taken up by plants and alter the N cycle in some ecosystems.

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**ES.5 Ecological Effects of Nitrogen and Sulfur Deposition**

It is clear from the body of evidence that NO<sub>Y</sub>, SO<sub>X</sub>, and PM contribute to total N and S deposition. In turn, N and S deposition cause alteration of the biogeochemistry and the physiology of organisms, resulting in harmful declines in biodiversity in terrestrial, freshwater, wetland, and estuarine ecosystems in the U.S. Decreases in biodiversity mean that some species become relatively less abundant and may be locally extirpated. In addition to the loss of unique living species, the decline in total biodiversity can be harmful because biodiversity is an important determinant of the stability of ecosystems and their ability to provide socially valuable ecosystem services (see more on biodiversity in Section IS.2.2.4).

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**ES.5.1 Acidification of Terrestrial and Freshwater Ecosystems**

Several decades of research have documented that N and S deposition cause freshwater and terrestrial ecosystem acidification in the U.S. New evidence strengthens the causal relationships for ecosystem acidification determined in the 2008 ISA (Table IS-1).

Many of the terrestrial and freshwater ecosystems most sensitive to acidification in the U.S. are found in the Northeast and Southeast. In the West, freshwater and terrestrial ecosystems acidified from deposition are now limited in extent and occur mostly in high-elevation sites. Watershed sensitivity to acid inputs depends on characteristics such as underlying geology (Appendix 4 and Appendix 7) and the sensitivity of species in the local biological community (Appendix 5 and Appendix 8). Regional heterogeneity of deposition levels that cause ecological effects are in part due to historic exposure and

climate. In the East, especially the southern Appalachian Mountains, and the Northeast, the effects of acidifying deposition have been studied for several decades.

Acidified aquatic habitats have a lower number of species (species richness) of fishes, macroinvertebrates, and phytoplankton. The effects of acidifying deposition on aquatic ecosystems also include physiological impairment or mortality of sensitive species and shifts in biodiversity of both flora and fauna. Organisms at all trophic levels are affected by acidification, with clear linkages to chemical indicators for effects on algae, zooplankton, benthic invertebrates, and fish. Acid-neutralizing capacity (ANC) is a measure of the buffering capacity of natural waters against acidification. Even though ANC does not directly alter the health of biota, it is a key metric of acidification that relates to pH and aluminum levels. Biological effects are primarily attributable to low pH and high inorganic aluminum concentration. Characterization of ANC and its levels of concern have not changed appreciably with the newly available information since the 2008 ISA. Few or no fish species are found in lakes and streams that have very low ANC (near zero) and low pH (near 5.0), and the number of fish species generally increases with higher ANC and pH (Appendix 8.3). The fish lost to acidification include culturally and recreationally important species.

Acidified terrestrial habitats are characterized by the detrimental physiological effects seen on vegetation, including inhibited growth and decreased plant health. Acidifying deposition can decrease membrane stability and freezing tolerance in young red spruce needles. For many species, calcium (Ca) depletion from the soil and aluminum mobilization cause decreased root uptake of Ca and disrupt fine root physiological functions. Reduced availability of (base) cations in the soil can also make trees more vulnerable to other stresses, such as damage from insects and other pathogens. Within the eastern U.S., the physiological effects of acidifying deposition have been well documented for the several culturally and commercially important tree species with known ecosystem services, particularly sugar maple (*Acer saccharum*) and red spruce (*Picea rubens*). Consistent and coherent evidence available before and since the 2008 ISA suggests acidifying deposition among these species can decrease foliar cold tolerance, increase rates of crown dieback, decrease tree growth, suppress seedling regeneration, and increase mortality rates. (Section IS.5.3; Appendix 5). Since the 2008 ISA, studies from the northeastern U.S. have shown that Ca addition can alleviate many of these effects, demonstrating that acidification effects can be ameliorated in the short term by soil amendments, suggesting the potential for recovery. However, Ca additions have been studied in only a few areas. Acidifying deposition has also been linked to changes in forest understory plant community composition in the northeastern U.S., grass and forb biodiversity in eight ecoregions across the U.S., and decreased grassland plant species richness in Europe.



Examples of improvement in acidification have been documented in some aquatic ecosystems in the regions most affected. Along with those improvements in acidification, chemical recovery has been observed in the Northeast, as seen by trends in water quality indicators ( $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$ , pH, ANC, inorganic monomeric Al, MeHg) towards inferred preindustrial values or, in the case of inorganic Al and MeHg, below water quality threshold values protective of biota and human health. Chemical recovery has not been observed in studies of the southern Appalachians. In a few examples in the Northeast, chemical recovery co-occurs with the movement of biological indicators toward recovery. However, biological recovery has been highly variable among ecosystems and taxonomic groups. Biological recovery lags behind, sometimes by decades, chemical recovery. In addition, the biological recovery trajectory may exhibit hysteresis, in which a system does not follow the same path from acidification to recovery. Most biological communities studied to date where signs of reversal are found have not returned to preacidification conditions and are unlikely to do so, given the extirpation of some species, fundamental alterations in function and structure, decade-long depletion of base cations, and changes in other interacting influences such as climate and land use.

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## **ES.5.2 Nitrogen Enrichment/Eutrophication of Terrestrial, Wetland, and Aquatic Ecosystems**

Terrestrial, wetland, freshwater, and estuarine ecosystems in the U.S. are affected by N enrichment/eutrophication caused by N deposition. N enrichment/eutrophication refers to N nutrient-driven changes in growth, physiology, and biodiversity. These effects have been consistently documented across the U.S. for hundreds of species. New evidence strengthens the causal relationships for ecosystem N enrichment/eutrophication determined in the 2008 ISA (Table IS-1).

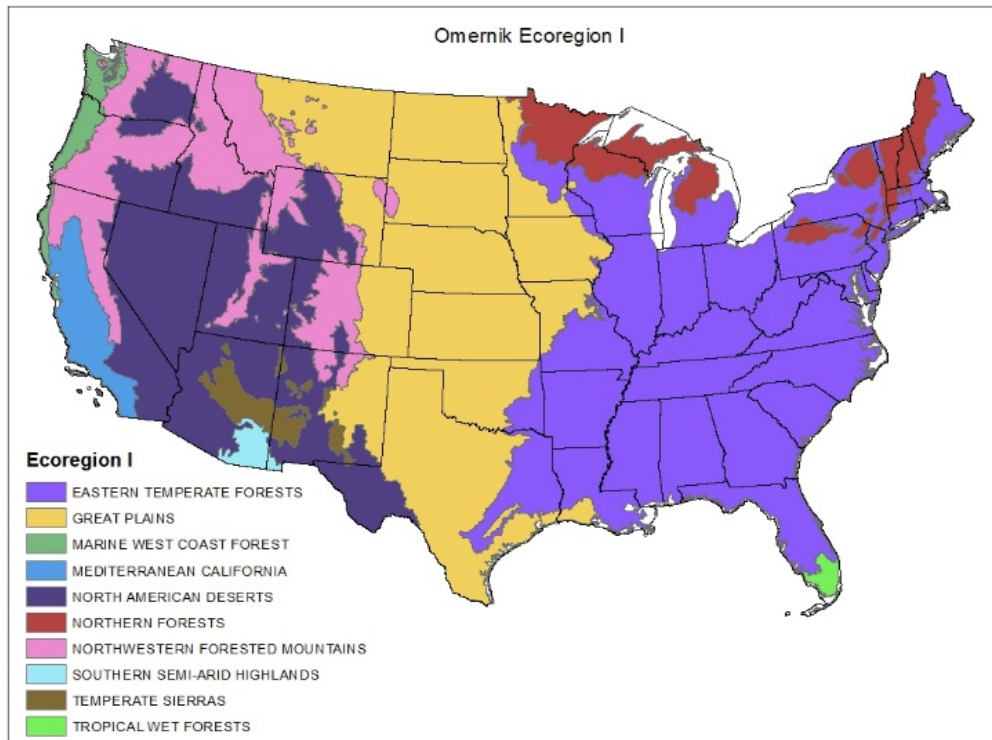
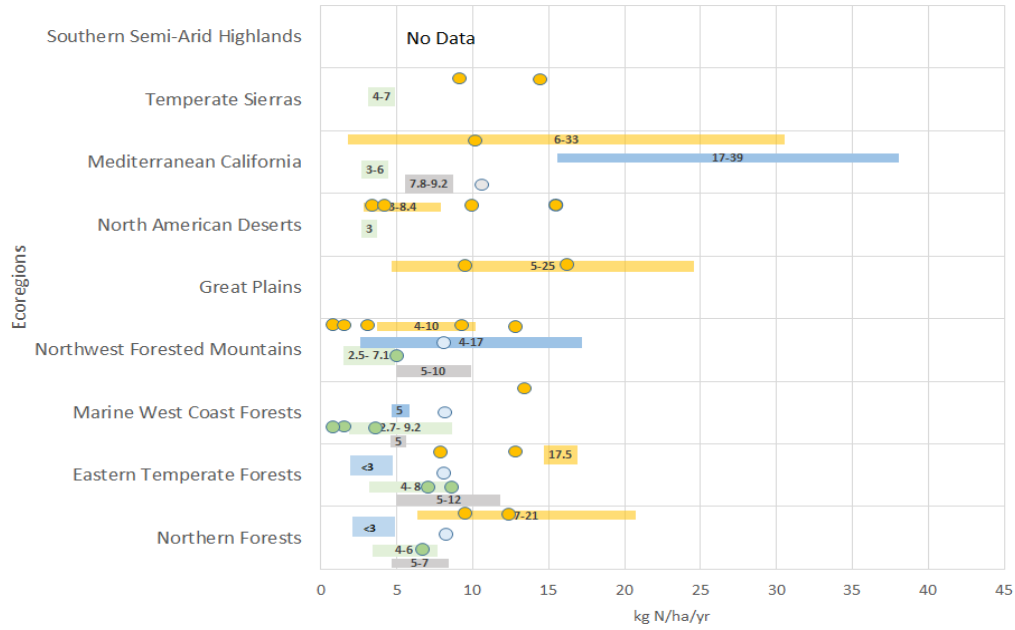
The 2008 ISA documented that the N enrichment effect in sensitive terrestrial and wetland ecosystems starts with the accumulation of N in the soil. This increases the availability of N, a nutrient that increases the growth of some species of soil microbes and vascular plants at the expense of other species, which may decrease biodiversity. Since the 2008 ISA, the largest increase in ecological evidence is for terrestrial N driven enrichment/eutrophication effects (Section IS.5.1, Section IS.5.2; Appendix 4, and Appendix 6).

This new research confirms the causal relationship between N deposition and ecological effects documented in the 2008 ISA and improves our understanding of the mechanistic links that inform causal determinations between N deposition, biogeochemistry, and biota in terrestrial ecosystems (Table IS-1). A new causal determination has been added to reflect more specific categories of effects to include physiology, growth, and ecosystem productivity. Further, there is now stronger empirical evidence from across most regions

of the U.S. to quantify critical loads (CLs) for N deposition. The figure below provides estimates of CLs across broad ecoregions and shows the ranges for different functional groups within these systems ([Figure ES-4](#)). Under the CLs, significant harmful effects from N deposition do not occur according to present knowledge, while at or above CLs, N deposition can cause a myriad of ecological effects, including decreased tree growth and increased mortality, and declines in grasses/forbs, lichens, and mycorrhizal fungi.

Since the 2008 ISA, studies have strengthened evidence of species-specific effects of N deposition on tree growth and mortality in the U.S. Although overall tree growth has generally been enhanced by N deposition over the last several decades, there is wide variation among species in growth and mortality responses. Moreover, within some individual species, N deposition can increase growth and/or survival at low levels, while decrease growth and/or survival at higher levels. Species with varying responses have also been shown to co-occur in places in the U.S., suggesting overstory tree community composition shifts with N deposition.

Since the 2008 ISA, studies have also strengthened the findings of N effects on decreasing lichen and mycorrhizal fungi biodiversity and provided additional CL estimates. In terrestrial ecosystems, new evidence provides support that epiphytic lichens (an algal- and/or cyanobacteria-fungal symbiont) and mycorrhizae (a plant-fungal symbiosis at the tips of plant roots) are the organisms most sensitive to atmospheric N deposition and acidifying deposition. Although lichens typically are only a small portion of terrestrial biomass, these changes in lichen communities are meaningful because lichens provide food and habitat for insects, birds, and mammals; contribute to nutrient and hydrologic cycling; have many traditional human uses; and have considerable potential for pharmaceutical use. Changes in the community composition of mycorrhizal fungi and declines in mycorrhizal abundance have been observed in the U.S. These fungi are important for supplying nutrients and water to plants, influencing soil C sequestration, and producing fruiting bodies (mushrooms) used by humans and wildlife.



CL = critical load; ha = hectare; kg = kilogram; N = nitrogen; yr = year.

The rectangles indicate the range of CLs designated by [Pardo et al. \(2011\)](#); the circles indicate new papers that have specified CLs; data from Table 6-28.

**Figure ES-4 Summary of critical loads for nitrogen in the U.S. for shrubs and herbaceous plants (yellow), trees (blue), lichens (green), and mycorrhizae (gray). Values expressed by major U.S. ecoregions.**

For wetland ecosystems, the 2008 ISA documented that wetlands receiving a larger fraction of their total water budget in the form of precipitation are more sensitive to the effects of N deposition. For example, bogs and fens (55–100% of hydrological input from rainfall) are more sensitive to N deposition than coastal wetlands (10–20% as rainfall). Since the 2008 ISA, CLs for U.S. coastal and freshwater wetlands have been established. The CL for freshwater wetlands is based on C cycling, as well as biodiversity represented by the morphology and population dynamics of the purple pitcher plant (*Sarracenia purpurea*). The CL for coastal wetlands is based on several different ecological endpoints, including plant community composition, microbial activity, and biogeochemistry.

The 2008 ISA documented that the process of N eutrophication is similar in freshwater and estuarine ecosystems and typically begins with a nutrient-stimulated algal bloom that is followed by anoxic conditions. The lack of oxygen in the water due to the respiration and decomposition of the algae affects higher trophic species. The contribution of N deposition to total N loading varies among freshwater lakes and stream ecosystems. Atmospheric deposition is the main source of new N inputs to most headwater stream, high-elevation lake, and low-order stream watersheds far from the influence of other N sources like agricultural runoff and wastewater effluent. N deposition was known at the time of the 2008 ISA to alter biogeochemical processes, nutrient ratios, and concentrations in recipient freshwater ecosystems. New CLs published since the 2008 ISA support previous observations of increased productivity of phytoplankton and algae, species changes, and reductions in diversity in atmospherically N enriched lakes and streams. The productivity of many freshwater ecosystems is N limited. Thus, even small amounts of N can shift nutrient ratios and affect the trophic status of lakes and streams. As reported in the 2008 ISA and newer studies, a shift from N limitation to either colimitation by N and P or limitation by P has been observed in some alpine lakes in the U.S. and other countries, with these shifts correlated with elevated N deposition.

Estuaries support a large biodiversity of flora and fauna and play a role in nutrient cycling. At the time of the 2008 ISA, N was recognized as the major cause of harm to the majority of estuaries in the U.S. Elevated N inputs to coastal areas can alter key processes that influence N and C cycling in near-coastal environments. Data evaluating sources of N to estuaries, from the 2008 ISA and newer studies reviewed in this ISA, indicate that N from atmospheric sources ranges from <10% to approximately 70% of total estuary N inputs; the atmospheric input for most estuaries is between 15 to 40% of total N inputs. N from atmospheric and other sources contributes to increased primary productivity, leading to eutrophication. In some coastal areas eutrophication from N loading may affect carbonate chemistry under certain circumstances, potentially contributing to acidifying conditions along with atmospheric anthropogenic CO<sub>2</sub> inputs and other factors. Since

2008, 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 in streams, rivers, lakes and estuaries. For this ISA, new information is consistent with the 2008 ISA, and the causal determinations for N enrichment in aquatic systems have been updated to reflect more specific categories of effects, including measures of productivity and altered growth of biota ([Table ES-1](#)).

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### **ES.5.3 Sulfur (S) Enrichment of Wetland and Freshwater Ecosystems**

SO<sub>x</sub> deposition increases SO<sub>4</sub><sup>2-</sup> concentration in surface waters. New evidence supports links between aqueous S concentrations in freshwater ecosystems and both mercury (Hg) methylation and sulfide toxicity ([Table ES-1](#)); however, quantitatively linking these outcomes to atmospheric deposition remains a challenge.

Increasing SO<sub>4</sub><sup>2-</sup> concentration in surface waters can stimulate the microbial transformation of inorganic Hg into methylmercury (MeHg; Appendix 12). MeHg is the most persistent and toxic form of Hg affecting animals in the natural environment. Indicators of S deposition effects upon Hg methylation include increases in MeHg concentrations or fraction of total Hg in water, sediments, and peat, as well as increases in MeHg concentrations in periphyton, submerged aquatic plants, invertebrates, and fish. New evidence confirms the relationship between aqueous concentrations of SO<sub>4</sub><sup>2-</sup> and MeHg and broadens our understanding of where methylation occurs from the wetlands and lakes reported in the 2008 ISA to include rivers, reservoirs, streams, and saturated forest soils. Hg methylation occurs at anoxic-oxic boundaries in peat moss and periphyton, as well as in wetland, lake, estuarine, and marine sediments. There are published quantitative relationships between surface water SO<sub>4</sub><sup>2-</sup> concentrations and MeHg concentrations, MeHg and total Hg in water, and Hg load in larval mosquitoes and fish. There is also evidence that decreasing S deposition loads over time (observational studies of SO<sub>x</sub> deposition, experimental studies of simulated SO<sub>x</sub> wet deposition) result in lower concentrations of MeHg in water, invertebrates, and fish.

There is new evidence since the 2008 ISA to infer a causal relationship between S deposition and sulfide phytotoxicity, which alters growth and productivity, species physiology, species richness, community composition, and biodiversity in wetland and freshwater ecosystems (Appendix 12). This new causal statement reflects new research on sulfide phytotoxicity in North American wetlands, as the 2008 ISA described sulfide phytotoxicity only in European ecosystems. Current levels of S deposition cause sulfide toxicity in wetland and aquatic plants. Indicators of sulfide phytotoxicity caused by S deposition include increases in water or sediment sulfide concentrations. Sulfide

negatively effects growth, competitive ability, and persistence in several wetland species, including the economically important species of wild rice and the keystone sawgrass species in the Everglades marshes. To date, no published studies have established regional sensitivities to sulfide phytotoxicity, although studies have observed its effects in New York, Minnesota, and Florida freshwater marshes. There are no S deposition-based critical loads for Hg methylation or sulfide phytotoxicity, although researchers have proposed water quality values to protect biota against these effects in several ecosystems (Appendix 12).

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#### **ES.5.4 Ecological Effects of Particulate Matter Other Than Those Associated with Nitrogen and Sulfur Deposition**

There is a likely causal relationship between PM and ecological effects on biota other than those associated with N and S deposition ([Table ES-1](#); Appendix 15). Since publication of the 2009 PM ISA, new literature has built upon the existing knowledge of ecological effects associated with PM components, especially metals and organics. In some instances, new techniques have enabled further characterization of the mechanisms of PM on soil processes, vegetation, and effects on fauna. New studies provide additional evidence for community-level responses to PM deposition, especially in soil microbial communities. However, uncertainties remain due to the difficulty in quantifying relationships between ambient concentrations of PM and ecosystem response.

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#### **ES.6 Ecosystem Services**

“Ecosystem services” refers to the concept that ecosystems provide benefits to people, directly or indirectly ([Costanza et al., 2017](#)) and produce socially valuable goods and services deserving of protection, restoration, and enhancement.

The ecosystem services literature has expanded since the 2008 ISA to include studies that better characterize ecosystem service valuation and quantification related to acidification and N enrichment/eutrophication.

Several new studies have paired biogeochemical modeling and benefit transfer equations informed by willingness-to-pay surveys to estimate the monetary damage done to ecosystems and the services they provide in the Adirondacks and Shenandoah regions due to ecosystem acidification (Appendix 14). Despite this progress, for many regions and specific services, poorly quantified relationships between deposition, ecological effects, and services are the greatest challenge in developing specific data on the economic benefits of emission reductions.

In the 2008 ISA, there were no publications that had specifically evaluated the effects of N deposition on ecosystem services associated with N driven enrichment/eutrophication. Since the 2008 ISA, several comprehensive studies have been published on the ecosystem services related to excessive N in U.S. water bodies. These include an evaluation of services affected by multiple N inputs (including N deposition) to the Chesapeake Bay, a synthesis of the cost-benefits on N loading across the nation, an estimation of the social cost of nitrogen when applied as fertilizer, and an analysis of how N lost from its intended area of application (e.g., agricultural fields) affects ecosystem services of adjacent ecosystems. Most notably, new work identifies over 1,000 links between N deposition and human beneficiaries.

Considering the full body of literature on ecosystem services related to N and S, the following conclusions are offered: (1) there is evidence that N and S emissions/deposition have a range of effects on U.S. ecosystem services and their social value; (2) some economic studies demonstrate such effects in broad terms, but it remains methodologically difficult to derive economic costs and benefits associated with specific regulatory decisions/standards; and (3) numerous, but still inadequately quantified, relationships are now documented between N and S air pollution and changes in final ecosystem goods and services.

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## **ES.7 Integrating across Ecosystems**

Overall, new evidence since the 2008 ISA increases the weight of evidence for ecological effects, confirming concepts previously identified and improving quantification of dose-response (or deposition-ecological indicator) relationships, particularly for N and S deposition. The ecological effects are described by the causality determinations in [Figure ES-5](#), which reorganizes the information in [Figure ES-3](#) to show a visualization of the effects of NO<sub>y</sub>, SO<sub>x</sub>, and PM by ecosystem type (e.g., terrestrial, wetland, freshwater, and estuarine). With this organization, the multiple effects occurring in each ecosystem due to various pollution combinations of NO<sub>y</sub>, SO<sub>x</sub>, and PM are emphasized. Between two and four different classes of pollutant effects may occur in each ecosystem type in the U.S. For more information on key messages, see the expanded discussion in the Integrated Synthesis; detailed information on specific ecosystem types and specific classes of pollutant effects included in the ISA may be found in the appendices.

Ecosystem			Terrestrial			Wetland			Fresh Water			Estuary	
Class of Pollutant Effect			Direct Phytotoxic	Acidification	N-enrichment/Eutrophication	N-enrichment/Eutrophication	Sulfide Toxicity	Mercury Methylation	Acidification	N-enrichment/Eutrophication	Sulfide Toxicity	Mercury Methylation	N-enrichment/Eutrophication
Indicator			Gases ‡	N+S dep	N dep	N dep	S dep	S dep	N+S dep	N dep	S dep	S dep	N dep
Scale of Ecological Response	Ecosystem	Productivity	Not evaluated in causal framework	Causal	Causal	Causal	Causal	Not evaluated in causal framework	Not evaluated in causal framework	Causal	Causal	Not evaluated in causal framework	Causal
	Community	Biodiversity	Not evaluated in causal framework	Causal	Causal	Causal	Causal	Not evaluated in causal framework	Causal	Causal	Causal	Not evaluated in causal framework	Causal
	Population	Individual	Growth rate	Not evaluated in causal framework	Causal	Causal	Causal	Causal	Not evaluated in causal framework	Causal	Causal	Not evaluated in causal framework	Causal
	Individual	Physiological alteration, stress or injury	Causal	Causal	Causal	Causal	Causal	Not evaluated in causal framework	Causal	Not evaluated in causal framework	Causal	Not evaluated in causal framework	Not evaluated in causal framework
	Geochemistry	Soil or sediment chemistry	Not evaluated in causal framework	Causal	Causal	Causal	Causal	Not evaluated in causal framework	Causal	Not evaluated in causal framework	Not evaluated in causal framework	Causal	Not evaluated in causal framework
		Surface water chemistry	Not evaluated in causal framework	Not evaluated in causal framework	Not evaluated in causal framework	Causal	Not evaluated in causal framework	Causal	Causal	Causal	Not evaluated in causal framework	Causal	Causal

**Causality framework**

Causal	Likely causal	Suggestive	Inadequate	Not likely	Not evaluated in causal framework
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‡ Includes: NO, NO<sub>2</sub>, HNO<sub>3</sub>, SO<sub>2</sub>, and PAN

**Figure ES-5 Causal relationships between the criteria pollutants and ecological effects organized under ecosystem type.**



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## ES.8 References

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