DATE

EPA-CASAC-20-XXX

The Honorable Andrew R. Wheeler
Administrator
U.S. Environmental Protection Agency
1200 Pennsylvania Avenue, N.W.
Washington, D.C. 20460

Subject: CASAC Review of the EPA's *Integrated Science Assessment for Oxides of Nitrogen, Oxides of Sulfur, and Particulate Matter – Ecological Criteria (Second External Review Draft – June 2018)*

Dear Administrator Wheeler:

The Clean Air Scientific Advisory Committee (CASAC) Secondary NAAQS Review Panel for Oxides of Nitrogen and Sulfur met on September 5-6, 2018, and <<*Insert teleconference date*>> to peer review the EPA's *Integrated Science Assessment for Oxides of Nitrogen, Oxides of Sulfur, and Particulate Matter – Ecological Criteria (Second External Review Draft)*, hereafter referred to as the Second Draft ISA. The Chartered CASAC approved the Panel's report on <<*Insert teleconference date*>>. The CASAC's consensus responses to the agency's charge questions and the individual review comments from the Panel are enclosed.

Overall, the CASAC finds that the Second Draft ISA is a well-organized and clearly written document. The CASAC commends the EPA for the notable improvements in the document that were directly responsive to the CASAC's comments (EPA-CASAC-17-004) on the First Draft ISA. The CASAC provides recommendations that are highlighted below and detailed in the consensus responses to further strengthen and improve the Second Draft ISA. With these recommended changes, the document will serve as a scientifically sound foundation for the review of the secondary National Ambient Air Quality Standards (NAAQS) for oxides of nitrogen, oxides of sulfur, and particulate matter, and no further review from the CASAC is needed.

The *Executive Summary* is a concise review of the science underlying the current NAAQS review. The CASAC suggests that several topics receive more emphasis in the Executive Summary. These topics include: the importance of ammonia (reduced nitrogen), concepts of chemical and biological recovery, and a brief definition of ecosystem services. It is not always clearly stated in the Executive Summary or in the Integrated Synthesis that all of the causal associations except the direct effects of gaseous oxides of sulfur (SO_x) and total oxides of nitrogen (NO_y) on vegetation are occurring at current levels of nitrogen (N) and sulfur (S) deposition. The CASAC suggests that this be clarified.

Chapter 1.2, a discussion of connections, concepts and changes (since the 2008 ISA), is directly responsive to previous CASAC comments. It would be helpful to include in the chapter additional discussion of the concepts of ecosystem services and chemical and biological recovery.

Chapter 1.3, a discussion of emissions and atmospheric chemistry, is an accurate condensation of material presented in Appendix 2 and has a generally appropriate level of detail. As further discussed in the consensus responses, the CASAC recommends some clarifications and additions to both Chapter 1.3 and Appendix 2 on the topics of particulate matter ($PM_{2.5}$) speciation and the treatment of ammonia (NH_3) emissions and measurements.

In *Chapter 1.4*, the current understanding of gas-phase direct phytotoxic effects has been well summarized. However, there is no clear demarcation in the chapter of effects that are due to direct gaseous uptake by plant canopies versus deposition to external canopy or ground-level surfaces transported into the soil. The CASAC suggests that discussions of direct effects of SO_x and oxides of nitrogen (NO_x) not be limited to phytotoxic effects, but consider a wider range of effects, such as phytotoxicity and visual injury, nonvisual physiological harmful effects, and possible fertilizer effects. The CASAC also notes that there is no summary of the effects of NH_3 on vegetation in either Chapter 1.4 or supporting Appendix 3 and recommends that the potential role of NH_3 be discussed.

Chapter 1.5 provides a clear summary of the effects of nitrogen and acidifying deposition on terrestrial ecosystems. The CASAC notes that a recent publication by Horn et al. (2018) on individual forest tree species mortality and growth responses across the U.S is a major contribution to the understanding of NO_x and SO_x effects on forests that should be included in this chapter. Revisions to further strengthen the chapter are discussed in the consensus responses.

Chapter 1.6 effectively summarizes the known effects of atmospheric deposition of sulfur and nitrogen on acidification and nutrient enrichment in freshwaters. The CASAC notes that the term "dissolved organic carbon," or DOC, used in the ISA refers to the measurement of just part of the dissolved organic matter (DOM) that includes carbon and other constituents such as organic nitrogen, phosphorus, and sulfur. The term DOM should be used in this chapter and throughout the ISA when referring more broadly to the role of organic matter.

The treatment of estuarine and coastal atmospheric nitrogen enrichment impacts is greatly improved in *Chapter 1.7* and related appendices. It is important to emphasize that reduced forms of nitrogen are an increasingly important fraction of atmospheric deposition which may be linked to an observed increase in harmful algal blooms and declines in water quality, including hypoxia. The CASAC finds that there is a need to deemphasize acidification in association with the atmospheric deposition of nitrogen to estuarine and near-coastal waters. The proposed connections between nitrogen enrichment and acidification are largely speculative and not supported by long-term monitoring of pH and related environmental variables in estuarine ecosystems.

Chapter 1.8 is a clearly-written, succinct summary of current knowledge of the effects of atmospheric nitrogen deposition on a variety of wetland end points including biodiversity, endangered species, nitrogen leaching, and links to other chemical constituents including greenhouse gases. The causal statements are well supported by the evidence provided in the chapter and related appendices.

In *Chapter 1.9* and related appendices, the discussion of freshwater and wetland ecosystem sulfur enrichment has been much improved. The CASAC notes that the EPA has added substantial new, policy-relevant, peer-reviewed literature and the causal statements are well supported by the available literature.

Chapter 1.10 and Appendix 15 effectively review a vast amount of literature on the ecological effects of particulate matter other than nitrogen and sulfur deposition (including direct and indirect effects, effects on solar radiation, and effects of trace metals and organics). Chapter 1.10 could be improved by including a brief description of the role that "crustal material" plays as a significant source of cations in PM.

Chapter 1.11, a discussion of recovery of ecosystems from nitrogen and sulfur deposition, is responsive to previous CASAC comments concerning reorganization of the ISA. The CASAC suggests that the EPA consider whether the definition of ecological recovery is too narrow. It is suggested that, rather than defining recovery as restoration of pre-industrial conditions, the EPA consider a definition more attuned to interdependencies between humans and ecosystems. In this regard, a system that has recovered could be defined as one that generates ecological processes and functionality similar to those found in the latter half of the 19^{th} century.

Chapter 1.12, climate modification of ecosystem response to nitrogen and sulfur addition, is brief with detailed material included in appendices. This revision reflects the CASAC's previous recommendations. However, as indicated in the consensus responses, the CASAC suggests that the chapter contain additional information about specific research findings. The CASAC also finds that there is a need for clarification of the scope of what is considered to be "climate" (meteorological factors over a five-year horizon, in contrast to long-term climate change, or associated changes to CO_2 concentrations and impacts on biogeochemistry).

The introduction and overview of key scientific uncertainty in *Chapter 1.13* is interesting. However, the CASAC suggests that the framework for defining and viewing uncertainty be more thoroughly incorporated throughout the document. An introduction to the adopted uncertainty framework could be included within the concepts reviewed in Chapter 1.2. It is further suggested that Chapter 1.13 contain a guidance paragraph on how to use data on system uncertainty.

The CASAC finds that *Chapter 1.14* on ecosystem services is somewhat disconnected from the previous chapters. The chapter could be improved by including an expanded introduction of how ecosystem services research provides an understanding of why nitrogen and sulfur matters for public welfare. This introduction could be included in Chapter 1.14 or within the definition of key concepts in Chapter 1.2. Other suggestions to improve Chapter 1.14 and Appendix 14 are included in the consensus responses.

The CASAC appreciates the opportunity to provide advice on the ISA and looks forward to the EPA's response.

Sincerely,

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Clean Air Scientific
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CASAC Secondary NAAQS
Review Panel for Oxides of
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Dr. Ivan J. Fernandez
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Enclosure

NOTICE

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Consensus Responses to Charge Questions on the EPA's *EPA's Integrated Science Assessment for Oxides of Nitrogen, Oxides of Sulfur, and Particulate Matter – Ecological Criteria (Second External Review Draft – June 2018)*

The Second External Review Draft Integrated Science Assessment for Oxides of Nitrogen, Oxides of Sulfur, and Particulate Matter – Ecological Criteria (hereafter referred to as the Second Draft ISA) was prepared by the EPA's National Center for Environmental Assessment as part of the ongoing review of the secondary (welfare-based) National Ambient Air Quality Standards (NAAQS) for Oxides of Nitrogen, Oxides of Sulfur and Particulate Matter (PM). The Clean Air Scientific Advisory Committee (CASAC) Secondary NAAQS Review Panel for Oxides of Nitrogen and Sulfur reviewed the Second Draft ISA at a public meeting held on September 5-6, 2018. At the meeting the Panel received a briefing on the Second Draft ISA, provided an opportunity for the public to present comments, and deliberated on responses to the EPA's charge questions. Draft consensus responses to the charge questions were developed by Panel subgroups based on the deliberation. At a teleconference meeting held on <<*Insert Date*>> the Panel discussed its consensus responses and the Chartered CASAC conducted a quality review and approved the Panel's report.

The EPA's charge questions and CASAC responses are presented below. The individual comments of members of the CASAC Secondary NAAQS Review Panel for Oxides of Nitrogen and Sulfur are included in Appendix A of this report.

Charge to the CASAC

The Second Draft ISA includes revisions developed in response to the comments and advice provided by the CASAC on the first external review draft of the ISA. The EPA has incorporated information from relevant studies published through May 2017 (the first draft ISA was current through December 2015). The revisions in the Second Draft ISA focus on several overarching recommendations previously provided by the CASAC:

- Reorganize the document by moving the detailed discussion of subject matter to appendices and focus on key messages in the Executive Summary and Integrative Synthesis (Chapter 1);
- Add additional references suggested by the CASAC; and
- Improve cross-referencing among chapters and appendices on key topics.

The Executive Summary and Integrated synthesis now serve as the main body of the document. The Executive Summary is intended to be a concise synopsis of key findings targeted to a broad audience, whereas the Integrated Synthesis is a more detailed synthesis of the ISA's most policy-relevant scientific findings.

Do the revised Executive Summary and Integrated Synthesis convey the main scientific findings of the ISA? Please comment on how effectively the revisions to the ISA reflect the recommendations and comments received from CASAC and the public comments. Please identify any additional revisions to *the ISA that will substantively strengthen the identification, evaluation and communication of the main scientific findings in these sections:*

1. *Executive Summary and Connections, concepts, and changes (Chapter 1.2)*
2. *Emissions and atmospheric chemistry (Chapter 1.3)*
3. *Gas-phase direct phytotoxic effects (Chapter 1.4)*
4. *Terrestrial nitrogen enrichment and acidification (Chapter 1.5)*
5. *Freshwater nitrogen enrichment and acidification (Chapter 1.6)*
6. *Estuarine and near-coastal nitrogen enrichment and nitrogen-driven acidification (Chapter 1.7)*
7. *Wetland ecosystem nitrogen enrichment (Chapter 1.8)*
8. *Freshwater and wetland ecosystem sulfur enrichment (Chapter 1.9)*
9. *Ecological effects of particulate matter other than nitrogen and sulfur deposition (Chapter 1.10)*
10. *Recovery, climate modification, key scientific uncertainties and ecosystem services (Chapters 1.11, 1.12, and 1.14)*

Review of the Executive Summary

The Executive Summary presents a concise review of the science underlying the current NAAQS review, with particular emphasis on areas of improved understanding since the last (2008) oxides of nitrogen[1] (NO_x) /oxides of sulfur[2] (SO_x) ISA. It is well-organized, clearly written, directly responsive to previous CASAC comments, and should serve as a useful model for future ISAs for these or other pollutants. Key findings are concisely summarized in Table ES-1 in the form of carefully crafted causal statements with links to the more detailed supporting discussions in the appendices.

Several topics, which are discussed thoroughly in the Integrated Synthesis and appendices, and which would benefit from more emphasis or clarity in the Executive Summary, include: the importance of ammonia (reduced nitrogen), concepts of chemical and biological recovery, and a brief definition of ecosystem services. Another minor suggestion is to add links in Table ES-1 to the appropriate sections of the Integrated Synthesis (in addition to the links to the appendices).

The causality discussion of direct effects of gaseous SO_x and total oxides of nitrogen[3] (NO_y) on vegetation is modified by the observation that there is no evidence that such effects are continuing at current, lower levels of exposure. It seems to be implied, but not always clearly stated in the Executive Summary or in the Integrated Synthesis, that all other causal associations are occurring at current levels of nitrogen (N) and sulfur (S) deposition. If true, this could be more clearly stated for the other causal statements. An alternative approach might be to follow the statement of "no effects from current gaseous exposures" with an observation like "Effects for all other causal associations identified in this ISA are currently occurring in some areas as a result of past and continuing S and/or N deposition."

[1] NO_x (oxides of nitrogen) is the sum of nitric oxide (NO) and nitrogen dioxide (NO_2.)
[2] The Second Draft ISA defines SO_x (oxides of sulfur) as the sum of SO_2 and particulate sulfate SO_4^{2-}.
[3] The Second Draft ISA defines NO_y (total oxides of nitrogen) as nitric oxide (NO) and nitrogen dioxide (NO_2) and all other oxidized N containing compounds formed from NO and NO_2.

Chapter 1.2 - Connections, Concepts, and Changes

Chapter 1.2, a discussion of connections, concepts and changes (since the 2008 ISA), is well organized, clearly worded, and directly responsive to previous CASAC comments. The summary figures are excellent and help introduce and explain some of the complex concepts and connections discussed later in the ISA which may be unfamiliar to many readers.

Some additional discussion of the concept of ecosystem services would be helpful and appropriate to include in this chapter. This might also be a good place to add an introductory discussion of the concepts of chemical and biological recovery, as these concepts relate to all the subsequently discussed ecological effects areas (Chapters 1.5 through 1.9), but are not really discussed until Chapter 1.11. The expected nature, asymmetries, and time scales associated with recovery seem especially important for effects associated with a combination of (larger) historical deposition followed by (typically smaller) continuing levels of S and N deposition.

Suggested minor wording changes in Chapter 1.2 include adding “in Understanding” after “Changes” in the title of Chapter 1.2. In the title of Section 1.1.2, “Source Apportionment” should be changed to “Source Contribution,” as the term source apportionment has a common, different, and more specific meaning in association with atmospheric modeling studies. As suggested for Table ES-1 in the Executive Summary, it would be helpful to add links in Table 1-1 to the appropriate sections of the Integrated Synthesis, in addition to the links to the appendices.

Chapter 1.3 - Emissions and Atmospheric Chemistry

Chapter 1.3 of the Second Draft ISA, Emissions and Atmospheric Chemistry, reflects previous recommendations of the CASAC. The chapter is an accurate condensation of material presented in Appendix 2 and has a generally appropriate level of detail. The CASAC recommends the following clarifications and additions (minor) to both Chapter 1.3 and Appendix 2, specifically on the topics of $PM_{2.5}$ speciation and the treatment of ammonia (NH_3) emissions and measurements.

- In the second paragraph of the chapter on page 27, expand the sentence beginning “NH_3 may account for 19-63% of total observed inorganic N deposition” by including the geographic information in the discussion in Appendix 2 (p. 2-5, lines 1-3): “…the contribution of NH_3 to total inorganic N deposition ranged from 19% in locations in the Northwest U.S. to 63% in 2 locations in the Southwest U.S., and was generally higher in summer than in winter (Li et al. 2016).”

- In this same paragraph, clarify what is meant by “most” in the sentence on lines 11-14 (also page 28 line 9), “…PM impacts discussed in this document are also mainly focused on N and S containing species, which together usually make up most of the $PM_{2.5}$ mass in most areas of the U.S…”

- Clarify the rationale for the chapter’s emphasis on inorganic constituents of PM by bringing forward the explanation from Appendix 2, which notes that the lack of information on speciated composition of carbonaceous matter makes it difficult to assess its ecological impacts.

- Section 1.3.1 (page 28 line 4) should note the relative magnitude and different spatial distributions of reduced nitrogen[4] (NH_x) emissions from animal waste (concentrated animal feeding operations - CAFOs) and fertilizer application. It should also note that animal waste and fertilizer application are two distinct and separate operations.
- Section 1.3.1 (page 28 line 16) states that "In the eastern U.S., NO_3^- and SO_4^{2-} make up an even greater portion of $PM_{2.5}$ mass in areas were $PM_{2.5}$ mass is the highest…" but this statement is not supported by the figure presented. In locations like Pennsylvania, New Jersey, and New York, it would appear that these two indeed make up close to 50%, but nitrate (NO_3^-) percentage is smaller than in areas like southern California or the Midwest.

- Section 1.3.2 should include more detailed information on NH_3 measurements, including remote sensing and the Southeastern Aerosol Research and Characterization (SEARCH) study (Hansen et al. 2003). The remote sensing section itself (Appendix 2.4.3.2) needs to include the Cross-track Infrared Sounder (CrIS), which arguably has the best remote sensing of NH_3 to date, since 2011. See for example the remote-sensing measurements of N dry deposition in Kharol et al. (2018). If the main uncertainty is NH_3 seasonality (page 29 line 12), remote sensing could help. This section should also mention studies that use statistical models, machine learning, land use regression, and global models to estimate the distribution of NO_2; the options go beyond regional models and satellite data. It is also recommended that the discussion of satellite NO_2 data be enhanced and that the capability of satellites to detect components of NO_y be clarified.

- The introductory sentence of Section 1.3.3 makes a strong statement about dramatically declining SO_x and NO_x emissions. This should be accompanied by an equally strong statement noting the concomitant increase in NH_x emissions.

Chapter 1.4 - Gas-phase Direct Phytotoxic Effects

The current understanding of gas-phase direct phytotoxic effects has been well summarized in Chapter 1.4 of the Second Draft ISA. Several revisions are provided to strengthen the chapter and supporting appendices. This brief chapter documents that little new work has been done on the direct effects of sulfur dioxide, nitrogen dioxide, nitrogen oxide, peroxy acetyl nitrate, and nitric acid (SO_2, NO_2, NO, PAN, and HNO_3 respectively) on plants. Early conclusions, however, had been quite clear that there is a causal relationship between exposure to all of these compounds and injury to vegetation.

The levels of NO_2 and SO_2 have declined below regulatory secondary standard levels across most of the country, and there is little evidence that these low levels have direct gas-phase effects on vegetation. One exception, cited in Section 1.4.3, is that some lichen species have declined or were extirpated under past and current ambient HNO_3 levels. The lichen studies document the known sensitivity of lichens to gas-phase phytotoxic effects, compared to vascular plants that tend to be more tolerant.

[4] NH_x (reduced nitrogen) is NH_3 plus NH_4^+

A logical conclusion that readers may draw is, if current levels of SO_2 and NO_2 are below regulatory standards and there is no evidence of phytotoxic impacts, there is no further need to review the science of S and N deposition. To help readers understand the potential for additional kinds of impacts, a brief statement could be included that describes the range of effects of ambient N + S including plant injury, physiology, and growth. Such a statement would be a good lead-in to Chapter 1.5.

Although the current understanding of gas-phase direct phytotoxic effects is well summarized in this chapter, there is no clear demarcation of how much of the atmospheric N or N + S effects are due to direct gaseous uptake by plant canopies versus deposition to external canopy or ground-level surfaces that is washed off by precipitation and transported into the soil (i.e., leading to potential soil-mediated deposition effects).

Likewise, canopy uptake of atmospheric N and S can cause phytotoxic effects – manifesting itself as visual injury or nonvisual harm. Phytotoxic effects appear to be uncommon in the U.S., except possibly near some point sources. But physiological effects from direct canopy uptake of atmospheric N and S is likely widespread, and may or may not be causing what could be called harmful effects. Studies that look at canopy uptake of gaseous N and S pollutants suggest stomatal uptake is widespread, but quantification of uptake and the physiological processing, effects, and importance of this uptake is a "black box" – it is not well understood.

The CASAC suggests that discussions of direct effects of SO_x and NO_x in Chapter 1.4, or supporting Appendix 3, not be limited to phytotoxic effects, but that a wider range of effects be considered such as phytotoxicity and visual injury, nonvisual physiological harmful effects, and possible fertilizer effects (e.g., growth changes, altered root:shoot ratios, or biodiversity impacts, that may or may not be considered ecologically desirable). These are discussed in other chapters of the ISA (e.g., Chapter 1.5) and could be referenced here.

Finally, there is no summary of the effects of NH_3 on vegetation, either in Chapter 1.4 or supporting Appendix 3. Chapter 1.3 reports that NH_3 concentrations have been increasing across the U.S. Research on NH_3 fumigation has been done in Europe (Sheppard et al. 2008; Cape et al. 2009; Sheppard et al. 2009) but not in the U.S. This has included observed growth decline of a heathland shrub (*Calluna vulgaris*) and death of a lichen (*Cladonia*) at ammonia critical levels of 8 $\mu g/m^3$. In its review of the first draft of the ISA (U.S. EPA CASAC 2017), the CASAC wrote "In Chapter 3, the ISA should note that there is a need for research on NH_3 fumigation effects on plants to fill a key knowledge gap." The CASAC recommends that the potential role of NH_3 on vegetation be discussed in Chapter 1.3 of the Second Draft ISA because it is relevant to atmospheric N deposition and ecological response. Although NH_3 is not currently a criteria pollutant, a literature review on this topic would inform the reader about potential NH_3 impacts on vegetation at current atmospheric concentrations and be an incentive to promote research on this topic in the U.S.

Chapter 1.5 - Terrestrial Nitrogen Enrichment and Acidification

Chapter 1.5 of the Second Draft ISA reflects previous CASAC recommendations. The chapter is well written and provides a clear summary of the effects of N and acidifying deposition on terrestrial ecosystems. The CASAC notes that a recent publication by Horn et al. (2018) on individual forest tree species mortality and growth responses to N and acidifying deposition across the U.S. should be included in the ISA. Their synthesis would make a major contribution to the document and to the understanding of NO_x and SO_x effects on forest growth and mortality, on an individual tree species basis, across the entire country. The following revisions to the Second Draft ISA are provided to further strengthen Chapter 1.5.

- On page 42 (Section 1.5.1.2), lines 13-14, a valid point is made that it is difficult to assign a single national critical load (CL) value. However, the Clark et al. 2018 publication (cited below Figure 1-7; and now published), reports on a minimum CL that was relatively uniform across the country (200 – 400 µeq/ha). It could be useful to point out the findings of this study as a potential single value "rule of thumb" depending on the intended application.

- On page 56 (Section 1.5.3.3), lines 1-2, the CASAC recommends modifying the last part of the sentence to read: "...base cation uptake, and the type and accuracy of deposition estimates (i.e., wet or bulk, vs. total; measured or modeled)".

- When referring to soil indicators and specific values, it is important to be explicit about the soil material that is being discussed. In most instances, the reference is to mineral soils, often B horizons, which are very different from the surface O horizons in forest soils. The likely resolution is to use the term "mineral soil" in all instances unless the O horizon is included or intended based on the supporting references. Most values for B horizon mineral soils would likely be incorrect if applied to organic surface O horizons.

- Compared to Chapter 1.4, there is a large amount of detail in Chapter 1.5. If the revisions to Chapter 1.5 offer the opportunity to move some of the detail to Appendix 4, or eliminate it if it already is in Appendix 4, the CASAC would recommend doing so. For instance, the explanation of belowground C allocation or the role of N deposition in decomposition (page 41) could go into Appendix 4.

Chapter 1.6 - Freshwater Nitrogen Enrichment and Acidification

Chapter 1.6 of the Second Draft ISA is an excellent summary of the known effects of atmospheric deposition of S and N on acidification and nutrient enrichment in freshwaters. The chapter reflects previous CASAC recommendations and effectively conveys the main scientific findings of the ISA. The text refers to the conclusions from the 2008 ISA (U.S. EPA 2008a) and adds new information where relevant. For example, in Chapter 1.6 there is expanded discussion of differences in biological responses between glaciated and unglaciated regions, freshwater algal blooms, and the role of climate modification – all with regard to ecosystem response to S and N. Moving much of the detailed text and citations to appendices means that the thorough reader must often shift back-and-forth between the main text and appendices, however this change is effective at improving the flow and succinctness of the chapter. The bolded conclusions are well supported by evidence provided in the text and appendices and the addition of individual growth effects is well justified. The CASAC also notes that the modeling section has been improved.

The CASAC notes that the term dissolved organic carbon or "DOC" used in the ISA refers to measurement of just part of the dissolved organic matter (DOM) that is the carbon. There is also organic nitrogen, phosphorus (P), and S as well as other elements in DOM, which is important in many ecosystems. These other forms of DOM can serve as nutrients, and also promote acidification when oxidized. While DOC is the principal form of DOM that comprises organic acids, and therefore affects pH and acid neutralizing capacity (ANC) of surface waters, the term "DOM" should be used in this chapter and throughout the ISA when referring more broadly to the role of organic matter.

Minor suggestions, corrections, or points of clarification

The CASAC provides the following minor suggestions, corrections, or points of clarification to consider when finalizing Chapter 1.6 of the Second Draft ISA.

- Section 1.6.1.1, page 59, line 12 should also mention dry deposited forms such as NO_2, NO, etc.

- In Section 1.6.1.1, page 59, line 14 it is not clear what is meant by elevated NO_3^- concentrations. Does this mean any measurable concentration of NO_3^-?

- Section 1.6.1.1, page 59, line 28 could be generalized by removing the word "iron" since there are many varieties of sulfide minerals.

- In Section 1.6.1.1, page 61, Table 1-3, for surface water sulfate it should be stated that preindustrial estimates are being compared to modern "measurements."

- In Section 1.6.1.1, page 61, Table 1-3, the surface water pH row could be updated to reflect the trends to more recent times such as 2015-16. It is not clear why references are cited here but not in other sections of this table.

- In Subsection 1.6.1.1.1, page 62, line 3, "conditions" should be "acidification."

- In Subsection 1.6.1.1.1, page 62, lines 10-14, the point being made is not clear. There is a shift away from focusing on chronic vs. episodic conditions to what?

- In Subsection 1.6.2.1.1, page 67, line 27, it would be preferable to write out words rather than use HAB on first use.

- In Section 1.6.2.2, page 69, line 16, the plus signs here should be plus or minus signs.

- In Section 1.6.3.2, page 75, line 7, something is missing from the sentence. The ANC units do not appear to make sense.

- In Section 1.6.3.2, page 75, lines 3 to 19, the CL values do not have much meaning unless compared with deposition values. It would be more useful to discuss CL exceedances rather than just raw CL values.

Chapter 1.7 - Estuarine and Near-Coastal Nitrogen Enrichment and Nitrogen-Driven Acidification

In the Second Draft ISA, the treatment of estuarine and coastal atmospheric nitrogen enrichment impacts (Chapter 1.7 and related appendices) is greatly improved. The Chapter reflects previous CASAC recommendations. Despite being lengthy, the related ISA appendices are comprehensive and the length is justified because of the great number of environmental drivers and internal controls of primary production, biogeochemical cycling, and associated water quality and habitat responses and conditions.
Emphasis on atmospheric deposition of nitrogen

The emphasis and detail in the Second Draft ISA on roles of atmospheric deposition of nitrogen (ADN) in eutrophication and altered biogeochemical cycling and biodiversity in estuarine and near coastal systems is excellent and appropriate. It is important to emphasize reduced forms of ADN (NH_3/NH_4), as these are an increasingly important fraction of ADN that may be linked to an observed increase in harmful algal blooms and declines in water quality, including hypoxia. The CASAC notes that organic N is an appreciable fraction (~20% of ADN) and that its role in eutrophication and biogeochemical cycling is an important issue to be addressed.

The suggestion in the Second Draft ISA that increasing levels of ADN may be changing nutrient limitation from N to N + P co-limitation or P limitation is largely speculative. There are however a few coastal systems where excessive N loading may periodically alter nutrient limitation from N to P or Si limitation. A prime example is the Mississippi plume region of the northern Gulf of Mexico (Sylvan et al. 2006) but this is largely driven by land-based N inputs during the spring high runoff period, which includes chemical fertilizers and wastewater.

The CASAC also notes that external organic matter (OM, which includes organic carbon + organic nitrogen + organic phosphorus) loading to estuarine and coastal waters appears to be increasing. The role of external OM relative ADN in total N loading, in productivity and biogeochemical cycling, and possibly in acidification responses is an important issue to be addressed.

Estuarine and near coastal water acidification

The CASAC finds that there is a need to deemphasize acidification in association with ADN inputs to estuarine and near-coastal waters. It should be removed from the heading in the Second Draft ISA and paragraphs emphasizing it should either be deleted or greatly toned down. The proposed “connections” between N enrichment and acidification are largely speculative and not supported by long-term monitoring of pH and related environmental variables in estuarine ecosystems. There is new literature to support this conclusion (Baumann and Smith 2018). The recent study by Baumann and Smith (2018) of long-term data bases of pH and trophic state as chlorophyll-a (Chla) on numerous Environmental Protection Agency - National Estuary Program (EPA-NEP) and National Oceanic and Atmospheric Administration - National Estuarine Research Reserve System (NOAA-NERRS) estuarine sites, shows no clear relationship between nutrient (nitrogen) inputs and acidification

The longest water quality data sets on U.S. estuaries (Chesapeake Bay and the Neuse River-Pamlico Sound system) indicate no significant trends in pH in either surface or bottom waters (see H. Paerl, Individual Comments, Appendix A, page A-49). Acidification is controlled by multiple interacting factors including rates of primary production (CO_2 fixation) which have been increasing due to eutrophication, tending to drive pH up, and mineralization of autochthonous (within system) and allochthonous (imported to system) organic matter, driving pH down. The net results are highly variable. One important fact is that with regard to autochthonous processes, it is impossible to mineralize more organic matter (driving pH down) than what is produced by autotrophs (algae and higher plants) (driving pH up). Therefore, with regard to eutrophication, one might expect pH to rise, unless every C molecule that is fixed is mineralized, in which case one would expect no net change in pH.

Models

The modeling discussion in the Second Draft ISA is improved over the last version, and the new section on uncertainty is informative. There are some new models added in the new version. These models highlight that a number of model inter-comparison activities, such as the International SCOPE Project on Nitrogen Transport and Transformation, the Northeast Nitrogen Synthesis, and the U.S. Geological Survey National Water Quality Assessment Program (Howarth 1996; Alexander et al. 2001; Alexander et al. 2002; Boyer and Howarth 2003; Driscoll et al. 2003), show that the various models, which are developed for different goals/places/timeframes, are generally consistent with one another (and with observations) with regard to the importance of various N sources, and the relationships between N sources (atmosphere, others) and ecosystem effects.

Subsection 1.13.2.2.3, pages 109-110, on eutrophication modeling is weak, and is not necessarily about eutrophication modeling – it largely addresses coastal N loading models. The treatment of the SPAtially Referenced Regressions on Watershed Attributes (SPARROW) model has some errors; in the other integrated synthesis chapter modeling sections and related appendices the treatment of SPARROW is better. The Dynamic Land Ecosystem Model (DLEM) (Tian et al. 2012) is also widely used and compared to SPARROW at regional scales across the U.S. and could be added to the modeling sections to strengthen the coastal N loading models described. Both DLEM and SPARROW are very different approaches and represent the continuum from statistical or empirical models to physically based/deterministic models – and these two approaches come to similar conclusions about N loads in any given region of the U.S. The SPARROW model has the advantage for this particular study/need of being able to estimate uncertainties of the time-averaged riverine loads, as well as the source shares ascribed to atmospheric deposition, and is probably the best approach to be used for the purpose of

EPA's Risk and Exposure Assessment. DLEM has the advantage of being able to simulate temporal variability of N loads (but without the source apportionment or same level of uncertainty).

Chapter 1.8 - Wetland Ecosystem Nitrogen Enrichment

Chapter 1.8 of the Second Draft ISA highlights potential effects of excess N, and often related acidification, on a wide variety of wetland ecosystem types, from bogs and fens to coastal wetlands/estuaries. This chapter is a clearly-written, succinct summary of a previously well-written chapter in the first draft of the ISA.

The chapter begins with an important distinction between most wetlands and their terrestrial counterparts – atmospheric deposition of N and S does not elicit acidification response in wetlands. Conversely, excess N can bring about numerous changes in many components that threaten the structure and function of wetland ecosystems. These include wetland biogeochemical processes and the ecophysiology of plants.

As stated in this chapter, the outcome of the ISA is quite clear – "the body of evidence is sufficient to infer a causal relationship between N deposition and the alteration of biogeochemical cycling in wetlands." On the other hand, both spatial variation and variation among types of wetland often preclude broad generalizations, wherein they can serve as either a source or a sink for a variety of N compounds. The figure referenced in the chapter (Figure 11-2 in Appendix 11) is a useful summary of recent N-manipulation studies examining N processing in contrasting wetlands, including coastal marshes, mangroves, riparian wetlands, and bogs.

The Second Draft ISA is similarly clear regarding the effects of excess N on wetland biota. It indicates that "the body of evidence is sufficient to infer a causal relationship between N deposition and the alteration of growth and productivity, species physiology, species richness, community composition, and biodiversity in wetlands." A conclusion of responses highlights what is generally known about unimpacted wetlands, regardless of type: they are typically highly N-limited. Thus, the initial responses to added N is that they exhibit characteristics along a gradient toward N saturation. Initially, there is enhanced growth and net primary productivity, plant tissue N increases, along with profound shifts in plant and microbial communities. The rates of these changes decrease over time as these systems move toward N saturation.

Similarly, biodiversity, particularly of wetland plant communities, declines in the face of N enrichment. This can be especially serious because a hallmark of wetlands is high plant species richness. Moreover, numerous endangered species can be threatened by excess N. Ultimately, this chapter highlights yet another parallel with terrestrial ecosystems regarding N enrichment. Excess N decreases abundance/richness of sensitive species, whereas it increases abundance/richness of tolerant species.

Chapter 1.9 - Freshwater and Wetland Ecosystem Sulfur Enrichment

Chapter 1.9 of the Second Draft ISA reflects previous CASAC recommendations. Discussion of the topic "freshwater and wetland ecosystem sulfur enrichment" is much improved by the adoption of the new organizational format. Further, EPA has added substantial new, policy-relevant, peer-reviewed literature into the revision of this chapter, such as a discussion of the relationship between mercury (Hg) and DOC, and Hg and S sources. This particular topic is one that has not received a lot of research until relatively recently, with a focus on sulfide toxicity and links to formation of methylmercury. The causal statements are now made in a more direct and succinct way, and they are well supported by the available literature. For example, the causal determination about sulfide phytotoxicity has been expanded from the first draft of the ISA to include growth and productivity as end points; and the causal determination about Hg has been reworded from the first draft to highlight that evidence is sufficient to infer a causal relationship between S deposition and increased methylation of Hg in surface water, sediment, and soils in aquatic environments.

In Chapter 1.9 on page 91, the point is made that where S inputs are similar in magnitude to rates of microbial sulfate reduction, the products of microbial sulfate transformation such as sulfide may be more reliable indicators of S enrichment effects than surface water sulfate concentrations. This is an important lesson that can also be applied to the N cycle, where N uptake by microbes or plants, or N removal by denitrification, may limit the ability of nitrate concentrations in water to quantify ecological effects of atmospheric inputs of reactive nitrogen.

The decline in sulfate inputs to terrestrial systems has been found to result in S deficiency, or potential S deficiency, in crops (Ketterings et al. 2012; Grant et al. 2012; Elkin et al. 2016). Similar S deficiency has not been reported in wetlands or aquatic ecosystems, as noted in the ISA.

Chapter 1.10 - Ecological Effects of Particulate Matter other than Nitrogen and Sulfur Deposition

Chapter 1.10 of the Second Draft ISA and the detailed Appendix 15 are well written and responsive to the input of CASAC on the first draft of the ISA. The chapter is a good review of a vast amount of literature on this expansive subject, including direct and indirect effects, effects on the solar radiation, and effects of trace metals and organics. The ISA also covers the effects on fauna and, for ecosystem level effects, looks into the gradients of response with increasing distance from a PM source (i.e., smelter). It notes only limited evidence for recovery around former smelters due to the continued presence of metals in the soil.

One general suggestion to improve Chapter 1.10 is to include a brief description of the role the "crustal material" component of PM plays as a significant source of cations. While soil is a relatively minor component of $PM_{2.5}$, it is typically the largest component of coarse particle mass ($PM_{10-2.5}$), and larger particles dry deposit more efficiently than small ones. Airborne soil could be a significant source of cations (e.g., Ca^{+}, Mg^{++}, K^{+}, Na^{+}) that may partially buffer acidifying deposition. It is also the one component of PM that appears to be increasing – at least in some regions of the U.S. and during some seasons.

Chapter 1.10 repeats the finding in the 2009 PM ISA (U.S. EPA 2009) that "the body of the evidence is sufficient to infer a likely causal relationship between deposition of PM and a variety of effects on individual organisms and ecosystems." The ISA notes that "the body of research since the 2009 PM ISA strengthens the finding on likely causal relationships and is based on information from the previous review and new findings in this review." Figure ES-2, however, notes in a footnote that these new findings in this review are "limited" in scope. Chapter 1.10 should also note that these new findings are limited in scope.

Chapter 1.11 - Recovery of Ecosystems from Nitrogen (N) and Sulfur (S) Deposition in the U.S.

Chapter 1.11 is responsive to previous CASAC comments concerning reorganization of the ISA. However, the CASAC suggests that the EPA consider whether the definition of ecological recovery (page 98, lines 22 - 25) as mimicking pre-industrial conditions is overly narrow. Alternatively, one might consider a definition more attuned to interdependencies between humans and ecosystems. In this regard, a system that has recovered could be defined as one that generates ecological processes and functionality similar to those found in the latter half of the 19^{th} century. This may better reflect concerns about recovering processes and functionality than duplicating conditions that existed 150 years ago.

Chapter 1.12 - Climate Modification of Ecosystem Response to Nitrogen (N) and Sulfur (S) Addition

Chapter 1.12 of the Second Draft ISA is brief with detailed material included in appendices. This revision reflects the CASAC's comments on the first draft of the ISA and is appreciated. However, the appendices on this topic are quite extensive, full of qualitative and quantitative discussion of dozens of recent works. The CASAC thus suggests revisions that go beyond pointing to the existence of research in particular areas in favor of providing a summary of specific research findings. For example, on page 101 (lines 1-3 and 6-9) the existence of works by Pinder et al. (2012) and Greaver et al. (2016) are mentioned, but not their contents. Then on line 10, it seems that the few studies on the effects of climate on ecosystem response to S that are identified in the appendix (Mitchell et al., 2011; Rice et al., 2014) could be readily discussed. Ultimately, this lack of detail in the Chapter 1.12 leaves a plausible interpretation that our understanding of climate modification of ecosystem response to N and S addition is too immature and uncertain to make this a component of this round of secondary criteria analysis. The expanding literature on this subject suggests otherwise.

The CASAC also finds that there is a need for explicit clarification in Chapter 1.12 of the scope of what is considered as "climate," such as meteorological factors over a five-year horizon, in contrast to long-term climate change, or associated changes to CO_2 concentrations and impacts on biogeochemistry.

Chapter 1.13 - Key Scientific Uncertainties

The CASAC finds that, while the introduction and overview of uncertainty in Chapter 1.13 is interesting, this framework for defining and viewing uncertainty would be better served by more thorough incorporation throughout the ISA. This would indicate: (1) which system (e.g., atmospheric science, ecological science, etc.) contributes the most uncertainty to the results, (2) the type of uncertainty that dominates in that system (e.g., statistical, scenario, etc.), and (3) ways that uncertainty can be reduced for each system. This could include some introduction to the adopted uncertainty framework within the concepts reviewed in Chapter 1.2. It is further suggested that Chapter 1.13 include a guidance paragraph on what one should do with data on system uncertainty, addressing questions such as: How should it affect analysis of the secondary standard? How should uncertainty affect judgments on the relative ecological health and integrity of ecosystems?

The CASAC also recommends including in Chapter 1.13 the following uncertainty elements that are specifically identified in the related appendix:

- Soil NO_x (See Appendix 2.2.3, page 2-14).

- SO_x partitioning (See Appendix 2, page 2-60), and how this is so uncertain despite certain SO_2 emissions.

- Relative importance of N deposition from the air vs. land for aquatic systems.

Chapter 1.14 - Ecosystem Services

Chapter 1.14 reflects the CASAC's previous comments on reorganization of the first draft of the ISA. However, the chapter is somewhat brief, and is just an abbreviated copy of the Appendix 14 summary (Section 14.6). As such, it comes across as a bit disconnected from the broader needs of the ISA, and does not adequately connect the topic to the previous chapters. The chapter could be improved by including an expanded introduction of how ecosystem service literature provides an understanding of why N and S matters for public welfare. This introduction could be included in Chapter 1.14 or within the definition of key concepts in Chapter 1.2.

The CASAC also notes that the placement of this chapter, coming after the discussion of uncertainty, seems odd, and suggests the chapter be placed before the uncertainty review. The latter would thus cover ecosystem services.

Chapter 1.14 would also benefit from a general summary of the extent of ecosystem services considered, bringing forward some of the relevant affected areas. Acidification and eutrophication are summarized in Chapter 1.14, but the section on nitrogen and climate modification in Appendix 14 (Section14.3) is not. There should also be a distillation of quantitative results (e.g., Table 14-1) in Chapter 1.14 to summarize (qualitatively) what are likely some of the most impacted ecosystem services, or the most prevalent types of pathways. The CASAC provides the following notes on three specific references to include in the ISA: Wang et al. (2017), Jenkins et al. (2010), and Keeler et al. (2016).

- One measure of the economic value of removing N from the landscape is \$5.91 / kg per year (mean) or \$10.50 / kg per year (high end). These values are based on the cost to remove a kg of N from a community water system. Wang et al. (2017) used U.S. EPA (2008b) to get these values. Wang et al. (2017) does not give the year of the dollar estimate.

- Estimated "annualized" value of N mitigation service (\$/kg N) in Arkansas in 2008 \$ is \$25.27 (mean), \$22.82 (low), and \$106.09 (high) (Jenkins et al. 2010).

- Keeler et al. (2016) measured the social cost of nitrogen (SCN) in Minnesota. They note that each kg of N applied to a field generates four compounds: NO_3^-, N_2O, NH_3, and NO_x. The total annual damage done by the four compounds measured in dollars/kg of N applied to a field is \$2.62 (mean), \$0.44 (low), \$10.79 (high). To convert annual values to a net present value the authors assume a twenty-year time horizon and a 3% rate of discount. This conversion generates values of \$40.15 (mean), \$6.74 (low), \$165.34 (high) per kg of N applied. These values account for the damage done to water quality (from N as NO_3-), changes in climate (from N as N_2O), and changes in air quality (from N as NO_x, NH_3, NH_4NO_3, and $(NH_4)_xSO_4$). All dollar values are in 2010 dollars.

Minor suggestions, corrections, or points of clarification in Chapter 1.14

The CASAC provides the following minor suggestions, corrections, or points of clarification to consider when finalizing Chapter 1.14 of the Second Draft ISA.

- Regarding the sentence on page 97, lines 20-21: "Overall N emissions and deposition have been increasing or relatively steady; consequently, there has been little reported on N enrichment recovery." One would assume there are some regions where N enrichment recovery has occurred. Can regional analysis be used to become more informed about N enrichment recovery? For example, later it is stated that recovery has occurred to some extent in parts of the northeast U.S.

- Regarding the sentence on page 97, lines 26-29: "For acidification caused by N and S deposition, chemical recovery of aquatic and terrestrial ecosystems is characterized by trends in water quality indicators…" Does this sentence indicate that chemical recovery from acidification caused by other sources of N and S are characterized by different trends in water quality indicators? It could be assumed that, no matter the original source(s) of acidification, recovery is characterized by the same "trends in water quality indicators (NO_3^-, 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."

- The following edit is suggested on page 98, lines 4-6: "When evaluating ecosystem recovery from ~~to~~ acidification, it is important to note that different chemical pools within the soil or water column may recover at different rates with the same decreases in ~~declining~~ atmospheric deposition ~~at different rates~~."

- On page 110, line 27 replace "valuation" with "values"

- On page 110, line 29 and 111, lines 1-3: Replace sentence that begins "Aside from valuation…" with "Aside from valuation studies, there is an improved understanding of the numerous causal pathways by which N and S deposition may affect ecosystem services, supported by studies that relate deposition to final ecosystem services under the FEGs-CS classification system."

- On page 111, line 14: delete "In their work" and insert parenthetical at end of previous sentence.

- On page 111, lines 15-17: these references refer to the language above at 111, lines 1-3.

- On page 111, lines 25-27: replace "…and (3) thousands of…" with "and (3) there is an improved understanding of the numerous causal pathways by which N and S deposition may affect ecosystem services, though most of these causal relationships remain to be quantified."

Minor suggestions, corrections, or points of clarification in Appendix 14

- On page 14-1, lines 6-11: replace the following existing text "The term "ecosystem services" refers to a concept that ecosystems provide benefits to people, directly or indirectly (Costanza et al. 2017), and these benefits are socially and economically valuable goods and services deserving of protection, restoration, and enhancement (Boyd and Banzhaf, 2007). The concept of ecosystem services recognizes that human well-being and survival are not independent of the rest of nature, humans are an integral and interdependent part of the biosphere (Costanza et al. 2017)" with "Generically, the term "ecosystem services" refers to the concept that ecosystems provide benefits to people, directly or indirectly (Costanza et al. 2017) and that ecosystems produce socially valuable goods and services deserving of protection, restoration, and enhancement (Boyd and Banzhaf 2007)."

- On page 14-1, lines 13-15: replace the sentence that begins "However…" with "However, because ecosystem services are often public goods their benefits can be difficult to monetize. We emphasize that the practical difficulty in no way implies that ecosystem service benefits are small."

- The following edit is suggested on page 14-1, lines 17-18: "Explicitly linking ecosystem services <u>to social and economic welfare measures</u> has proven difficult."

- On page 14-2, lines 1-3: replace "thousands of causal relationships…" with "new work identifies a large number of scientifically and economically plausible causal relationships that link N and S air pollution to changes in Final …"

- On page 14-2, line 12: replace "develop" with "quantify"

- On page 14-5, lines 5-9: replace "A few years later, Chestnut and Mills (2005) compared the actual benefits of reducing emissions of NO_x and SO_x in Title IV of the Clean Air Act Amendments (CAAA) by the estimate of benefits made in 1990. They conclude that quantitative assessment was problematic at that time due to a lack of..." with "A few years later, Chestnut and Mills (2005) derived new estimates and compared them to the 1990 estimates of the benefits of reducing emissions of NO_x and SO_x in Title IV of the Clean Air Act Amendments (CAAA). They conclude that the 1990 quantitative assessment was limited by a lack of..."

- The following edit is suggested on page 14-6, line 1: "…dose-response <u>relationships</u>"

- On page 14-6, line 22: replace "valuation" with "values"

- The following edit is suggested on page 14-6, line 27: "…dose-response <u>relationships</u> between deposition, ecological effect<u>s</u>, and services <u>is</u>…"

- On page 14-6, line 28: replace "in developing specific data on" with "to quantification of"

- On page 14-9, line 6: replace "In other words" with "According to the study"

- On page 14-9, line 12: add "<u>via</u> air deposition…"

- The following edit is suggested on page 14-9, line 13: "alters services<u>. They did this by</u>…"

- The following edit is suggested on page 14-9, line 16: "…then applied per-unit…"

- On page 14-18, line 5: replace "purchase of" with "greater reliance on…" and "leading to" with "associated with"

- On page 14-9, line 14: replace "success" with "spread"

- On page 14-22 line 28: replace "valuation" with "values"

- On page 14-22 lines 30-31: replace the sentence "Aside from valuation, …" with "Aside from valuation efforts, studies using the FEGS CS have improved understanding of the numerous causal pathways by which N and S deposition may affect ecosystem services." Also note that the original language was duplicated on 14-23 lines 10-12. It is recommended that the citations from page 14-23 lines 10-12 be moved up to this spot.

- On page 14-23, line 9: delete "In their work" and insert parenthetical at end of previous sentence.

- On page 14-23, lines 13-15: delete the sentence that begins "In these analyses…"

- On page 14-23 lines 29-31: replace "…and (3) thousands of…" with "and (3) there is an improved understanding of the numerous causal pathways by which N and S deposition may affect ecosystem services, though most of these causal relationships remain to be quantified."

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U.S. EPA (U.S. Environmental Protection Agency) 2009. *Integrated Science Assessment (ISA) for Particulate Matter (Final Report, December 2009)*. EPA/600/R-08/139F. U.S. Environmental Protection Agency, Office of Research and Development, National Center for Environmental Assessment- RTP Division, Research Triangle Park, NC. [available at: http://cfpub.epa.gov/ncea/cfm/recordisplay.cfm?deid=216546]

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Appendix A

Individual Comments from Members CASAC Secondary NAAQS Review Panel for Oxides of Nitrogen and Sulfur on *EPA's Integrated Science Assessment for Oxides of Nitrogen, Oxides of Sulfur, and Particulate Matter – Ecological Criteria (Second External Review Draft)*

Dr. Edith Allen **A-2**
Dr. Praveen Amar **A-6**
Dr. James Boyd **A-12**
Dr. Douglas Burns **A-17**
Ms. Lauraine Chestnut **A-19**
Dr. Mark Fenn **A-20**
Dr. Ivan Fernandez **A-26**
Dr. Frank Gilliam **A-35**
Dr. Daven Henze **A-36**
Dr. Donna Kenski **A-39**
Dr. William McDowell **A-41**
Dr. Erik Nelson **A-42**
Dr. Hans Paerl **A-49**
Mr. Richard Poirot **A-53**

Dr. Edith Allen

The revised ISA is well written and in large part the revisions to the first draft ISA reflect the CASAC recommendations, but I have some recommendations for revisions and additions. All of the summary information from Chapters 1-14 of the 1st draft ISA has been revised and expanded, and moved to the Chapter 1 Integrated Synthesis in the 2nd draft ISA. The new longer, more detailed summaries are a good approach to helping readers (especially non-specialists in the field) grasp a large amount of complex material, with references to relevant Appendices. Many additional citations have been included.

The reorganization has resulted in only one Chapter in the entire document. This is a bit odd, as I initially found myself looking for Chapter 2, etc. Instead of calling this Chapter 1, perhaps it could simply be called "Integrated Synthesis" with 14 chapters. Or divide Chapter 1 into several chapters by topic with sub-chapters.

The major omission I note in the revised ISA overall is the lack of reference to reduced nitrogen in the Executive Summary, Chapters 1.4 and 1.5 and Appendix 3, which was among the recommendations and comments from CASAC. These are detailed below.

Executive Summary--Emissions, Ambient Air Concentrations, Deposition
There is no mention of reduced N in the Executive Summary, although reduced N is discussed throughout the ISA. I realize this ISA is titled "Oxides of N and S…." but the document makes a strong scientific contribution to reporting the atmospheric concentrations and understanding the role of reduced N air pollutants in natural systems. It seems that a statement can be added to the Executive Summary that reflects the ISA contribution to reviewing our knowledge on reduced N atmospheric concentrations and impacts. Reduced N is not part of the regulatory framework at this time, but we have this opportunity to demonstrate with scientific evidence that the current regulations for oxidized N only control about one-half (regionally variable) of atmospheric N pollutants. With respect to reduced N, the Executive Summary does not convey the all of the main scientific findings of the ISA.

Chapter 1 Integrated Synthesis

Chapter 1.1.1, p. 3, l. 7-12, states the aims of this ISA with a focus on oxidized N and S and PM. However, reduced nitrogen is not mentioned in these aims (again, because this is the "Oxides of N and S…." report), but in fact data on reduced N (NHX) is discussed in every section of Chapter 1 and in the appendices. The fact that this ISA also presents new information about reduced N could be included in the aims, to highlight the importance of reduced N as an air pollutant. With respect to reduced N, Chapter 1.1.1 does not convey all of the main scientific findings of the ISA.

Chapter 1.2.3, p. 3, l. 27: states that agriculture is the main source of reduced N. However, automobiles are now also known as a source of reduced N (Sun et al. 2017 Env Sci Tech). Furthermore, agriculture is a source of oxidized N (Almaraz et al. 2018). Additional research to quantify these sources is an important task for setting future clean air regulations. These citations could perhaps be included in

Chapter 1.3 Emissions and Atmospheric Chemistry (although perhaps new 2018 citations will not be included at this late date. There are older citations on NOx emissions from agriculture).

Sun, K., Tao, L., Miller, D.J., Pan, D., and Golston, L.M., et al. 2017. Vehicle emissions as an important urban ammonia source in the United States and China. Environ. Sci. Technol. 51: 2472-2481.

Almaraz M et al. 2018. Agriculture is a major source of NOx pollution in California. Science Advances 4. doi: 10.1126/sciadv.aao3477

Chapter 1.4 and Appendix 3

Chapter 1.4 is brief compared to the other chapters, but has a new summary of oxidized N and S gas phase pollutants and their impacts. The explanatory information is reviewed in Appendix 3. This is appropriate because little new supporting research has been done in this area, the levels of NO2 and SO2 have declined below regulatory secondary standard levels across most of the country, and there is no evidence that these low levels have direct gas-phase effects on vegetation.
However, there is no summary of gas-phase effects of NHx on vegetation, either in Chapter 1.4 or supporting Appendix 3. See below for literature updates on impacts of gas-phase reduced N on vegetation.

Appendix 3 presents a comprehensive summary of new research since 2008 on the phytotoxic effects of gas phase NOx and SOx. Atmospheric concentrations of both are generally below levels known to cause phytotoxic effects on vegetation. Exceptions are noted (e.g., p. 3-10, l. 26-28, “nearly continuous exposure to 0.1 ppm NO2 for 8 weeks significantly reduced growth of Kentucky blue grass;” p. 3-14, l. 25--some species of lichens are sensitive at HNO3 of 8-10 ppb).

Appendix 3 does not report on gas-phase effects of NHx on vegetation, although there have been studies on NHx effects on vegetation in the US and Europe. Several of these are cited in Appendix 6 Terrestrial Ecosystems: p. 6-37 , l. 34 reports studies on NHx pollution impacts in the US (Jovan et al. 2012 in California, Schirokauer et al. 2014a in Alaska). The effects could be from gas phase or total deposition, which is why these studies are reported in Appendix 6. The studies conclude that the combined deposition of NOx and NHx are related to lichen decline, not a particular form of N. NOx was the predominant air pollutant in the California study, and the two forms were approximately equal in Alaska.

By contrast, Sheppard et al. (2011) p. 6-37, l. 35, showed that NH3 gas phase dry fumigation caused high shrub and lichen mortality at 20 kg N/ha/yr. Atmospheric concentrations are not reported in this article, but are in an earlier article from the same experiment, with CL of 8 ug NH3/m3 (Sheppard et al. 2008, citation below). Wet deposition promoted shrub growth. This article could be cited in Appendix 3.

The CASAC wrote the following statement in the 1st ISA Review Draft Report 8-4-17, p. 7 l. 22-23: “In Chapter 3, the ISA should note that there is a need for research on NH_3 fumigation effects on plants to fill a key knowledge gap”

As there was no report of the phytotoxic effects of NHx in this 2nd draft ISA, the revision does not reflect the recommendations and comments from CASAC with regard to reduced N.

In the CASAC response to the first ISA (8-4-12) I suggested a few citations from Europe on NH3 fumigation to be cited in Chapter 3 of the 1st ISA, now Chapter 1.4 and Appendix 3 of the 2nd ISA draft. None of these are cited either in the current draft ISA or the 2008 ISA:
Cape et al. recommend a CL of 1 ug/m3 NH3 for lichens and 3 ug/m3 NH3 for higher plants.
Cape, J. N., L. J. van der Eerden, L. J. Sheppard, I. D. Leith, and M. A. Sutton. (2009). Evidence for changing the critical level for ammonia. Environmental Pollution **157**:1033-1037.
Sheppard et al. 2008 observed growth decline of a heathland shrub (*Calluna vulgaris*) and death of a lichen (*Cladonia*) at ammonia critical levels of 8 ug/m3 NH3:
Sheppard, L.J., Leith, I.D., Crossley, A., van Dijk, N., Fowler, D., Sutton, M.A., Woods, C., (2008). Stress responses of *Calluna vulgaris* to reduced and oxidised N applied under 'real world conditions'. Environmental Pollution 154 (3):404–413.
Sheppard, L.J., Leith, I.D., Crossley, A., van Dijk, N., Fowler, D., Sutton, M.A., 2009. Long-term cumulative exposure exacerbates the effects of atmospheric ammonia on an ombrotrophic bog: implications for critical levels. In: Sutton, M.A., Reis, S., Baker, S.M.H. (Eds.), Atmospheric Ammonia – Detecting Emission Changes and Environmental Impacts. Springer, Berlin, pp. 49–58.

The six studies discussed above suggest to me that NH3 atmospheric concentrations in the US are at a high enough level in some areas where they may cause direct phytotoxic effects on plants, but more research is needed. Chapter 1.4 and Appendix 3 could make this recommendation regarding reduced N.

An editorial change to Appendix 3:
p. 3-6 l. 27 "In a more recent, Geiser and Neitlich (2007) "
CHANGE TO
More recently, Geiser and Neitlich (2007)

Chapter 1.5
Compared to Chapt. 1.4, there is a large amount of detail in Chapt. 1.5. This is fine with me, but if there is a general consensus to reduce the length of Chapt. 1.5 some of this detail could be moved to, or already is in, Appendix 4. For instance, the explanation of belowground C allocation or the role of N deposition in decomposition (p. 41) could go into Appendix 4.

Appendix 4—Biogeochemistry
This Appendix is improved with additional text for each section explaining or hypothesizing why certain changes under N deposition may or may not occur.

Some editorial changes to Appendix 4:
Table 4-1 "**Section of ISA that Discusses Each Endpoint"** Be sure to state that the section numbers refer to Appendix sections (e.g., Appendix Sec. 4.3.2).

p. 4-40 l. 15 "dentification"
CHANGE TO
denitrification

Table 4-13 the + and – signs are too small to see

4-92 l. 9 Comparisons are is summarized

CHANGE TO
Comparisons are summarized

4-98 l. 12 there was is no single deposition….
CHANGE TO
there was no single deposition…..

p. 4-108, l. 4-5--N addition can increase increased litter decomposition,
N addition can increase litter decomposition,

Appendix 5 Biological Effects of Acidification

Reviews literature on levels of N deposition at which effects occur. The studies typically include NOx plus NH_x deposition or fertilization. The studies show the importance of both combined sources or reduced and oxidized N contributing to vegetation and ecosystem responses. No changes needed regarding additional discussion about reduced N, as this form of N is included here.

Appendix 6 Terrestrial Ecosystems Enrichment
Reviews literature on levels on N deposition at which N enrichment effects occur. Studies include both NOx and NHx deposition or fertilization, alone or combined. Some studies show different ecosystem responses to N form, others do not. No changes needed regarding additional discussion about reduced N, as this form of N is included here.

Table 6-11 p. 6-85-- Change second occurrence of Rao and Allen (2010) to Rao et al. (2010). Rao et al. (2010) refers to the 5 yr duration field study.

Appendix 13 Climate modification

Some sections on climate modification appear in individual Appendices, while some are included in Appendix 13. Perhaps it would be best to be consistent, e.g., move all sections on Climate Modification into Appendix 13. But I don't feel strongly about this.

General comment: There are many long tables, especially in the Appendices, that do not seem to have any format for order of citations. These could be ordered by type of response where applicable (e.g., positive, negative, neutral), or by some other category appropriate to the topic of the table, or simply by alphabetical order of author. As listed, it is hard to find citations in these large tables.

Dr. Praveen Amar

The charge to the CASAC focuses on the Executive Summary and Integrated Synthesis of the ISA. The CASAC has been asked to: (a) comment on whether the revised Executive Summary and Integrated Synthesis convey the main scientific findings of the ISA; (b) comment on how effectively the revisions to the ISA reflect the recommendations and comments received from the CASAC and public comments; (c) identify any additional revisions to the ISA that will substantively strengthen the identification, evaluation, and communication of the main scientific findings. These main scientific findings are included in the following sections of the second draft ISA:

1. Executive Summary and Connections, Concepts and Changes (Chapter 1.2)
2. Emissions and atmospheric chemistry (Chapter 1.3)
3. Gas-phase direct phytotoxic effects (Chapter 1.4)
4. Terrestrial nitrogen enrichment and acidification (Chapter 1.5)
5. Freshwater nitrogen enrichment and acidification (Chapter 1.6)
6. Estuarine and near-coastal nitrogen enrichment and nitrogen-driven acidification (Chapter 1.7)
7. Wetland ecosystem nitrogen enrichment (Chapter 1.8)
8. Freshwater and Wetland ecosystem sulfur enrichment (Chapter 1.9)
9. Ecological effects of Particulate Matter other than nitrogen and sulfur deposition (Chapter 1.10)
10. Recovery, climate modification, key scientific uncertainties and ecosystem services (Chapter 1.11, 1.12, 1.13 and 1.14)

My comments include comments on Executive Summary, Chapter 1.2, Chapter 1.3 and Chapter 1.10 (and Appendix 15)

<u>Comments on Executive Summary:</u>

The revised Executive Summary in the second draft ISA is concise and very well-written and reflects the main scientific findings included in the ISA. The specific comments below are provided to improve the presentation and to hopefully improve the communication of findings to the scientific community and the general public.

Page lxiii (Lines 1 to 21): Though this second draft does a much better job of paying attention to the role of ammonia and reactive reduced nitrogen (NHx) in overall acid deposition and nitrogen enrichment, it would be helpful to explicitly name ammonia gas in Lines 6-7 as an important gas that contributes to the gaseous and particle phase deposition of reduced nitrogen (even though it is NOT a criteria pollutant). It would be also useful to add the term "NHx" or ammonia (as appropriate) every time we use the terms "NOy, SOx, PM…" together when and where it strengthens the overall context. For example, throughout the document when we note "NOy, SOx, and PM…" emissions contributing to total N and S deposition, we need to replace this with "NOy, NH3, SOx, and PM…" emissions.

Page lxiii (Line 5): When we use the term "N" for the first time in Line 5, I suggest we define it here (perhaps in the footnote) as "total reactive nitrogen (oxidized and reduced, organic and inorganic, etc.)". Same comment for "S" deposition (SO_2 and SO_4).

Page lxiv (Lines 31 32): What is meant by the word, "Connectivity" as in "these ecosystems are linked by the CONNECTIVITY…"? Does the word "connectivity" have the usual meaning in the English language or does it have a specialized meaning in the ecological literature?

Page lxv (Lines 16 18): EPA needs to be commended for following up on CASAC's recommendation on the first draft of ISA for by modifying Appendix 16 by adding Adirondacks as a separate and stand alone case study area that is now one of the six potential candidates for additional analysis under the Risk and Exposure Assessment (REA).

Page lxvi (Lines 1-10): Please note that "N and S containing species" do NOT make up "most" of the $PM_{2.5}$ mass in "most" areas of the U.S. Many areas of the U.S. have substantial mass fractions of EC and OC (POA, SOA) in the measured $PM_{2.5}$ mass (50 percent or higher). Please provide reference here (perhaps, a footnote) for the statement that NH_3 contributes 19 to 63% of total inorganic nitrogen deposition. Also, do you mean NH_3 here or do you mean NH_x? The sentence in Lines 7-8, starting with "therefore, the contribution….in this ISA" is rather awkward and should be rewritten.

Page lxvii (Lines 6 to 20): Please clearly state that EGU emissions of SO_2 are going down as in "coal-burning EGUs" since EGUs burning natural gas emit negligible emissions of SO_2 and also that very few EGUs burn fuel oil. Also, the Executive Summary needs to state clearly that animal waste operations (CAFOs) and fertilizer application are two distinct and separate agricultural operations and the first one (CAFO) is a much larger source of ammonia emissions than the second one. I think it is more like that 60 to 70 percent of the total NH_3 emissions come from CAFO/animal waste operations and about 15 to 20 percent of ammonia emissions come from fertilizer applications. Please provide this emissions split here as well as in Chapter 1 and Appendix 2 (Table 2-1) by listing separately emissions of NH_3 from CAFO/animal waste and fertilizer applications under the "agriculture" category, based on the latest EPA inventories (year 2015 or later, perhaps?). Please note either in Executive Summary or Integrated Synthesis as well as in Appendix 2 that agricultural sources contribute a very large fraction of total ammonia emissions (87%; Table 2-1). Also, it would be useful to provide information on projections of *future* NH_3 emissions here and other parts of the ISA (Chapter 1 and Appendix 2) clearly stating the magnitude of this *increase* in ammonia emissions and contrasting this with overall projected *decrease* in NO_x and SO_2 emissions and implication of this important finding on relative contribution of reduced and oxidized nitrogen to future overall reactive nitrogen emissions as well as ecological effects of nitrogen enrichment and total acid deposition of N and S. (Please note that the SO_2 emissions are summed up incorrectly in Table 2-1 in Appendix 2. The correct total for SO_2 should be 2.84 teragrams of S/year instead of 2.3 teragrams.).

Page lxvii (Lines 25-27): Please provide, similar to NO_2 concentrations decrease, information on decrease in ambient SO_2 concentrations. Also, it would be useful to provide data on lowered concentrations of nitrate and sulfate fractions of $PM_{2.5}$, recognizing fully well that they will not be linear or proportional to decrease in SO_x and NO_x emissions.

Page lxviii (Lines 16 17): It would be useful to provide the data on measured H equivalents in 1990s and compare that to the value of 1500 H equivalents in Midwest and Atlantic states in the years 2011-2013.

Page lxviii (Lines 24-34): Please provide the reference to this five-level hierarchy of causality evidence. Not sure what reference to "Sir Bradford Hill" means in Chapter 1. Also, please note the explanatory language for level-4 evidence here is not the same as in Chapter 1. Finally, in Line 24, do you mean "two orders of magnitude" from current concentrations (as in 10 to 100 to 1000) or do you mean "a factor of 2" from current concentrations?

Page lxxv (Lines 1 and 2): This is the first reference to "biological recovery" in this document under Executive Summary. Please briefly describe here what is meant by this term and how it differs from "chemical recovery" or "geochemical recovery."

Page lxxv (Lines 32 to 35): I think we need to state that "calcium addition" at a few lakes or aquatic watersheds or at a few tree species levels does not provide rigorous evidence that there is "potential for recovery" that is scalable to very large number of lakes, aquatic watersheds and terrestrial ecosystems.

Page lxxvii (Lines 12 and 27): I think we need to avoid the word "New" as in "new CLs" (line 27). What do new CLs mean? Are there old CLs? The Line 12 reference to CLs is more appropriate as it notes critical loads established for watersheds after the 2008 ISA. This comment applies to the whole ISA document and we must avoid the term "new CLs" and replace with language such as "CLs established after the 2008 ISA…".

Page lxxx (Line 12): Please replace "1,104 unique chains…" with easier to grasp language ".. just over 1,100 unique chains….". The reference to "1,104 unique chains" just confuses the issue.

Page lxxx (Lines 23-35): The final concluding section on "Integrating across Ecosystems" is beautifully written and makes the main points just right.

<u>Comments on Chapter 1.2 (Connections, Concepts, and Changes):</u>

This new section on "Connections, Concepts, and Changes" is a welcome addition as it describes in one place how various important components of ISA are organized. A general comment I have is about the Title of the section itself. The word "Changes" does not clearly describe what is in the Text itself. For example, the subtitle under "Changes" notes "New Evidence and Causal Determinations" (Section 1.2.3; page 20). Also, well- written Text under the section clearly notes our enhanced understanding of causal determinations since 2008 and higher levels of weight of evidence since 2008, etc. In summary, "Changes" clearly outlines "Changes since the 2008 ISA." I suggest we use a slightly modified Title "Connections, Concepts, and Changes in our Understanding" since the 2008 ISA.

Page 7 (Line 15): At the risk of stating the obvious, please say "Emissions of NOy, AMMONIA, SOx and PM cause an accumulation of N and S…."

Page 7(Lines 22 23): Please give a brief description (two or three sentences) on how the concept of cascading also applies to Sulfur. And, how it is different (and less complex, perhaps?) than the cascading of nitrogen in the environment.

Page 8 (Figure 1-2) The term “VOC” is defined twice.

Page 8 (Lines 11 to 16): The analogy between “dose-response relationships” in human health assessments and “critical loads” in environmental health assessments is well placed here.

Page 8 (Lines 33 34): The sentence states “REDUCTION in ENTROPY through energy flow from autotrophs to top predators…” I thought ALL processes in natural and physical systems *always* result in an increase in entropy and never a decrease in entropy (“Second Law of Thermodynamics”). Am I missing something?

Page 11 (Title of Section 1.2.2) The Title “Deposition and Source Apportionment of N and S to Ecosystems”: The words “source apportionment” are used here incorrectly. The term “source apportionment” is used in the atmospheric sciences and emission sources/source categories literature to apportion contribution of various sources to measured atmospheric concentrations. This section, 1.2.2.2 notes varying contributions of various sources (atmospheric deposition, agriculture, transport from watersheds, etc.) for different ecosystems without detailed apportioning. I suggest the Title “Deposition and Source *Contributions* of N and S to Ecosystems” which is more reflective of what follows in this section.

Page 11 (Lines 21 22): If atmospheric deposition is the second largest human-mediated N source, it begs the question “what is the largest source of N deposition?”. Please note the largest source of N deposition.

Page 12 (Line 12): It notes that field observations have shown that draught conditions result in an increase in lake load of 5 kg/ha in S. What was the baseline S lake load without draught?

Pages 12-15 (Section 1.2.2.3): The write up on critical loads is excellent. However, the “standard” definition of “critical load” is repeated three times (Pages 12, 13, 15). Also, it should be useful to say a few more things about “target loads” or a single “target load” and how they are set in the policy context of regulations. This is expected to be an important topic in the Risk Exposure Assessment (REA) and Policy Assessment (PA).

Pages 17-18 (Section 1.2.2.5): This section on “Reduced versus Oxidized Nitrogen Effects across Ecosystems” provides a good summary of the state-of-the science and is very helpful and timely. It should be extremely useful in future REA and PA as EPA tries to address the role of (increasing) ammonia emissions versus roles of (decreasing) emissions of NOx and SO2.

Page 18 (Section 1.2.2.6): This section on AAI is well written. It appears that the second draft ISA (June 2018) and the REA would evaluate the role of “critical loads” as an “organizing principle” for all ecosystem types instead of AAI.

Page 19 (Line 1) starting at the bottom of page 18: Please clearly state “and, ammonia, the non criteria pollutant”

Page 21 (Line 18): Please add NH3 to “it is clear that NOy, NH3, SOx and PM..”

Page 23 (Line 26): Replace "new CLs" with "CLs established since the 2008 ISA".

Comments on Section 1.3: Emissions and Atmospheric Chemistry (Pages 27-32):

This section covers the major points reasonably well but needs to improve to clearly state a number of technical subjects, as noted below:

Page 27 (Lines 6 to 14): on line 14, the words "rather than NOx or PM, therefore it is discussed in the ISA" are rather awkward. We should simply say that "in some places, NH3 may account for a larger fraction (19-63%) of observed inorganic N deposition compared to smaller contributions of NOy and PM." Lines 9 to 12 need to be rewritten for clarity. Also, we need to state here and at other places in the ISA that sulfates and nitrates do NOT contribute to "most" of the PM2.5 mass at "most" of the sites. As I noted in my comments on Executive Summary, EC, OC and trace metals contribute about half or more to the PM2.5 mass.

Page 27 (Lines 18-19): Needs to be rewritten. At line 18, start the new sentence as "NO3 and NH4, and in some cases organic nitrogen….".

Page 28 (Lines 4 5): Here and at other places in the draft ISA, we need to clearly state that animal waste (CAFO) and fertilizer applications (in that order) are two distinct and separate operations resulting in substantial ammonia emissions. Since these two agricultural sources are very different in nature, it would be helpful to split these two categories under "Agriculture" (for example, Table 2-1 in Appendix 2) and provide separate emission estimates. Please provide here and other appropriate locations (Table 2-1) the magnitude of these emissions, noting that CAFOs are a much larger source of ammonia emissions than fertilizer applications; more like 4 to 1. Please see other comments I have provided above under my comments on the "Executive Summary".

Page 28 (Lines 29 to 32): Please say something about why the data from IMPROVE and CSN are not used for estimating deposition rates.

Page 29 (Line 4): Please explain what is meant by "compensation points". A clear definition in parentheses should help.

Pages 29 (Line 14), Page 32 (line 5) Do we have a descriptive scale for describing uncertainties in this ISA? At these two places, we use the words "highly uncertain" and "sizeable uncertainty". Do they have the same meaning? What is meant by "inherent" uncertainties?

Page 29 (Line 42): What was the magnitude of SO2 concentrations in the air in response to 72% reduction in SO2 emissions from 1990 to 2011?

Page 29 Line 22 and other places in this ISA) : Similar to the *quantitative decreases* in SO2 and NOx emissions over the years which are clearly stated in this ISA, we need to provide data on *quantitative increases* in NH3 emissions over the years. Here we simply say "emissions of NH3 have increased in many years" without saying by how much. Similarly, we need to include EPA estimates of *future increases* in ammonia emissions under "business as usual" scenarios taking into account potential increases in animal waste operations (CAFOs) and increased food production (fertilizer application).

Comments on Section 1.10 (Ecological Effects of Particulate Matter other than N and S Deposition) and Appendix 15 of the same Title (Pages 15-1 to 15-27)

The CASAC is asked to comment on the adequacy of characterization of these ecological effects for the non-nitrogen and non-sulfur particles associated components in this second draft ISA, and on whether and how Section 1.10 (and Appendix 15) includes CASAC recommendations on the first draft ISA. Finally, we are asked to identify any additional revisions to ISA (Section 1.10 and the Appendix 15) that will substantively strengthen the identification, evaluation, and communication of the main scientific findings included in this ISA.

Section 1.10 (and the detailed Appendix 15) is well written and responds to the input of CASAC on the first draft ISA. It does a good job of reviewing a vast amount of literature on this expansive subject (including direct and indirect effects, effects on the solar radiation, effects of trace metals and organics). The ISA also covers the effects on fauna and, for ecosystem level efforts, looks into the gradients of response with increasing distance from PM source (smelter). It notes only limited evidence for recovery around former smelters due to the continued presence of metals in the soil.

This section repeats the finding from the 2009 PM ISA that **"the body of the evidence is sufficient to infer a likely causal relationship between deposition of PM and a variety of effects on individual organisms and ecosystems."** This section and the Executive Summary note that this causal determination was not included in the first draft of 2018 NOx/SOx/PM ISA. The ISA notes that "the body of research since the 2009 PM ISA strengthens the finding on likely causal relationship and is based on information from the previous review and new findings in this review. Figure ES-2 in Executive Summary, however, notes in the footnote that these new findings in this review are "limited" in scope. To be consistent, Section 1.10 should also note that these new findings are "limited" in scope.

Dr. James Boyd

<u>Executive Summary</u>

lxvii, line 11
…dominated by dust and the combustion products of fires…

lxix, line 1
Unclear what “judgments of error” means or refers to. Also, the word “judgement” implies subjectivity, as opposed to an objective assessment of “uncertainty.”

lxix, line 7
Define what is meant by an “endpoint category.”

lxxiv, line 8
Would be useful to provide examples of what is meant by injury. “Injury refers to a range of effects including…”

lxxiv, line 13
Use a more broadly understood term (eradicated, eliminated, extinguished) be used instead of “extirpated.”

lxxiv, line 13
Replace “services to humanity” with “socially valuable ecosystem services.”

lxxv, lines 4 &6
Define and distinguish, or consolidate use of the terms “species richness,” “biodiversity.” Are they the same thing? If not, explain.

(Comment applies to the use of the terms throughout the document.)

lxxiv, line 27
Add “culturally <u>and commercially</u> important tree species”

lxxvii, lines 26 and 30
Can/should “nutrient ratios” be instead “nutrient concentrations”?

lxxviii, line 15
“…shown to elicit biological responses, <u>such as …</u>”

lxxix, line 12
What does it mean to “negatively effect competition”?

lxxx, line 12
This should be explained more fully. In particular "chains" and "unique" will not be understandable. Suggested language: "new work identifies a large number of scientifically and economically plausible causal relationships that link …"

lxxvii, line 16
"…services and their social benefits."

lxxvii, line 19
Change "over a thousand relationships" to "numerous, but still inadequately quantified relationships…"

<u>Chapter 1.2</u>

Page 6, line 5
Replace "extirpation" with "loss"

Page 6, line 6
"unique species, but also several decades…"

Page 6, line 21
More specific about what "resource collapse" means. Collapse of fish populations, water quality, etc.?

Page 6, line 30
"… and services on which humanity…"

Page 7, line 1
"…it is…"

Page 7, line 9
Meaning of "multifunctionality" is unclear.

<u>Chapters 1.11, 1.12, 1.13, 1.14</u>

98, line 6
Confusing. Delete "at different rates"?

110, line 27
Replace "valuation" with "values"

110, line 29 and 111, lines 1-3

Replace sentence that begins "Aside from valuation… with

"Aside from valuation studies, there is an improved understanding of the numerous causal pathways by which N and S deposition may affect ecosystem services, supported by studies that relate deposition to final ecosystem services under the FEGs-CS classification system."

111, line 14
Delete "In their work" and insert parenthetical at end of previous sentence.

111, lines 15-17
These references refer to the language above at 111, lines 1-3.

111, lines 25-27
Replace "…and (3) thousands of…" with
"and (3) there is an improved understanding of the numerous causal pathways by which N and S deposition may affect ecosystem services, though most of these causal relationships remain to be quantified."

<u>Appendix 14</u>

14-1, lines 6-11

Replace the existing text: "The term "ecosystem services" refers to a concept that ecosystems provide benefits to people, directly or indirectly (Costanza et al. 2017), and these benefits are socially and economically valuable goods and services deserving of protection, restoration, and enhancement (Boyd and Banzhaf 2007). The concept of ecosystem services recognizes that human well-being and survival are not independent of the rest of nature, humans are an integral and interdependent part of the biosphere (Costanza et al. 2017)"

With

"Generically, the term "ecosystem services" refers to the concept that ecosystems provide benefits to people, directly or indirectly (Costanza et al. 2017) and that ecosystems produce socially valuable goods and services deserving of protection, restoration, and enhancement (Boyd and Banzhaf 2007)."

14-1, lines 13-15
Replace the sentence that begins "However…" with "However, because ecosystem services are often public goods their benefits can be difficult to monetize. We emphasize that that practical difficulty in no way implies that ecosystem service benefits are small."

14-1, lines 17-18
"Explicitly linking ecosystem services <u>to social and economic welfare measures</u> has proven difficult."

14-2, lines 1-3

Replace "thousands of causal relationships…" with

"new work identifies a large number of scientifically and economically plausible causal relationships that link N and S air pollution to changes in Final …"

14-2, line 12
Replace "develop" with "quantify"

14-5, lines 5-9
Replace "A few years later, Chestnut and Mills (2005) compared the actual benefits of reducing emissions of NOX and SOX in Title IV of the Clean Air Act Amendments (CAAA) by the estimate of benefits made in 1990. They conclude that quantitative assessment was problematic at that time due to a lack of..." with

"A few years later, Chestnut and Mills (2005) derived new estimates and compared them to the 1990 estimates of the benefits of reducing emissions of NOX and SOX in Title IV of the Clean Air Act Amendments (CAAA). They conclude that the 1990 quantitative assessment was limited by a lack of..."

14-6, line 1
"…dose-response relationships"

14-6, line 22
Replace "valuation" with "values"

14-6, line 27
"…dose-response relationships between depositions, ecological effects, and services is…"

14-6, line 28
Replace "in developing specific data on" with "to quantification of"

14-9, line 6
Replace "In other words" with "According to the study"

14-9, line 12
Add "via air deposition…"

14-9, line 13
"alters services. They did this by…"

14-9, line 16
"…then applied per-unit…"

14-18, line 5
Replace "purchase of" with "greater reliance on…"
And replace "leading to" with "associated with"

14-9, line 14
Replace "success" with "spread"

14-22 line 28
Replace "valuation" with "values"

14-22 lines 30-31
Replace the sentence "Aside from valuation, …" with "Aside from valuation efforts, studies using the FEGS CS have improved understanding of the numerous causal pathways by which N and S deposition may affect ecosystem services."

Also note that the original language was duplicated on 14-23 lines 10-12. Recommend taking the citations from 14-23 lines 10-12 and moving them up to this spot.

Also delete the sentence at 14-23 lines 13-15 that begins "In these analyses…"

14-23, line 9
Delete "In their work" and insert parenthetical at end of previous sentence.

14-23 lines 29-31
Replace "…and (3) thousands of…" with
"and (3) there is an improved understanding of the numerous causal pathways by which N and S deposition may affect ecosystem services, though most of these causal relationships remain to be quantified."

Dr. Douglas Burns

In general, I am pleased with the changes that have been made in this version of the ISA by providing an Executive Summary, succinct and shorter chapters that summarize effects and causal relationships, and the relegation of much of the detailed supportive evidence to relevant appendices. The flow and readability of the text are improved for a reader who would not want to wade through the detailed evidence provided in the numerous citations.

Chapter 1.6 – Freshwater Ecosystem Nitrogen Enrichment and Acidification This chapter does a good job of summarizing known effects of atmospheric deposition of S and N on acidification and nutrient enrichment in freshwaters. The text effectively refers to the conclusions from the 2008 ISA and adds new information where relevant. Moving some of the detailed text and citations to appendices is effective in improving the flow and succinctness of this chapter. The bolded conclusions are well supported by evidence provided in the text and appendices. I have only a few generally minor suggestions, corrections, or points of clarification as described below.

Section 1.6.1.1, page 59, line 12 – should also mention dry deposited forms such as NO2, NO, etc.

Section 1.6.1.1, page 59, line 14 – not clear what is meant by elevated NO3 concentrations. Do you mean any measurable concentration of NO3?

Section 1.6.1.1, page 59, line 28 – could generalize by removing word “iron” here since there are many varieties of sulfide minerals.

Section 1.6.1.1, page 61, Table 1-3 – for surface water sulfate, should state that preindustrial estimates are being compared to modern “measurements.”

Section 1.6.1.1, page 61, Table 1-3 – could update the surface water pH row to reflect that trends to more recent times such as 2015-16. Why are references cited here but not in other sections of this table?

Subsection 1.6.1.1.1, page 62, line 3 – “conditions” should be “acidification.”

Subsection 1.6.1.1.1, page 62, lines 10-14 – I am not certain about the point being made here. There is a shift away from focusing on chronic vs. episodic conditions to what? I disagree with this statement.

Subsection 1.6.2.1.1, page 67, line 27 – would be good to write out words rather than use HAB on first use.

Section 1.6.2.2, page 69, line 16 – the plus signs here should be plus or minus signs.

Section 1.6.3.2, page 75, line 7 – something missing from this sentence, ANC units do not make sense.

Section 1.6.3.2, page 75, lines 3 to 19 – the CL values do not have much meaning unless compared with deposition values. Would have a lot more value here to discuss CL exceedances rather than just raw CL values.

Chapter 1.8 – Wetland Ecosystem Nitrogen Enrichment and Acidification. This brief chapter summarizes current knowledge on the effects of atmospheric N deposition on a variety of wetland end points including biodiversity, endangered species, N leaching, and links to other chemical constituents including greenhouse gases. The chapter effectively builds on the results of 2008 ISA and cites studies that expand the scope of effects, including at least two review/synthesis studies. The two bolded causal statements are well supported by the evidence provided in the chapter and related appendices.
Subsection 1.8.2.2, page 89, line 14 – change "that show that" to "show that"

Chapter 1.9 - Freshwater and Wetland Ecosystem Sulfur Enrichment
This chapter and the accompanying appendix is effective in summarizing the results of the 2008 ISA and updating these results with significant new studies that have been published since release of the previous ISA. A new causal statement on the role of atmospheric S deposition and the ecological effects of sulfide phytotoxicity is warranted by recent published studies. The 2008 causal statement on the role of S deposition and enhanced rates of Hg methylation and bioaccumulation is strengthened by additional evidence published since the previous ISA. I have only one small item for consideration that reflects a growing body of research on S deficiency in crop production.
Appendix 12 – One issue related to sulfur cycling with links to atmospheric deposition is an increasing number of reports that with declines in deposition in recent years, evidence of sulfur crop deficiency is being reported with increasing frequency. While this is not an enrichment issue, it is worthy of at least a brief mention because there are economic implications. As far as I know, there have been no comparable results reported for natural ecosystems. Examples of S deficiency studies include some agricultural reports in the "gray" literature as well as a few papers in the peer-revised literature (I have not performed a comprehensive literature review of this topic):

- Ketterings, Q.M., Godwin, G., Gami, S., Dietzel, K., Lawrence, J., Barney, P., Kilcer, T., Stanyard, M., Albers, C., Cherney, J.H. and Cherney, D., 2012. Soil and tissue testing for sulfur management of alfalfa in New York State. *Soil Science Society of America Journal*, *76*(1), pp.298-306.
- Grant, C.A., Mahli, S.S. and Karamanos, R.E., 2012. Sulfur management for rapeseed. *Field Crops Research*, *128*, pp.119-128.
- Elkin, K.R., Veith, T.L., Lu, H., Goslee, S.C., Buda, A.R., Collick, A.S., Folmar, G.J., Kleinman, P.J. and Bryant, R.B., 2016. Declining Atmospheric Sulfate Deposition in an Agricultural Watershed in Central Pennsylvania, USA. *Agricultural & Environmental Letters*, *1*(1).

Ms. Lauraine Chestnut

Overall: The new organizational structure for the document is a great improvement over the first draft. It makes the massive amount of information assembled from multiple relevant disciplines much easier to digest and helps a great deal in highlighting the policy relevance of the information.

Executive summary: This is a very good short overview of the ecosystem effects of concern related to these pollutants. The focus on evidence of causal relationships is helpful at this level of detail. The graphics are also helpful in communicating the big picture.

Chapter 1: Integrated Synthesis

Overall: Creating this chapter as the main body of the ISA is very helpful. It has enough detail to understand the underlying scientific evidence without being overwhelming. It does a good job integrating the information across the many disciplines that are relevant to this review.

Chapter 1.2.2 Connections, Concepts, and Changes

This section does a good job of explaining some of the key concepts that were identified as important and cross cutting across disciplines in the review of the first draft of the ISA. It gives the necessary background for understanding key concepts used in the ISA that are important for subsequent policy assessment. It is also helpful in highlighting changes in information available since the previous ISA.

What is missing is a short section on ecosystem services and how these are relevant to understanding the significance of the effects on ecosystems presented throughout the ISA.

Chapter 1.14: Ecosystem Services

This chapter is fine as a short summary of the literature, but it does not adequately connect the topic to the previous chapters. It is not clear from what is presented here why this topic is included.

Dr. Mark Fenn

EXECUTIVE SUMMARY:

The executive summary is appropriately concise now, giving a well written summary of the key points of the ISA.

---Table ES-1: For the first 3 causal statements, it is worth considering adding a modifier to the table to the effect that although these causal effects occur, they are rare in occurrence across CONUS under current atmospheric conditions. If that isn't acceptable, could add this as a footnote to the table.
---p 74 (the pdf page #), lines 6-8: Here and elsewhere, it seems greater acknowledgement should be given that although evidence suggests that phytotoxicity from these gaseous pollutants is not widespread in CONUS, plant uptake of N gases and S gases is occurring at subtoxic levels, and almost certainly affecting plant nutrition and physiology to some degree across wide regions without visible symptoms, although this is an understudied area of research. Certainly plant uptake of such pollutants is well documented, even if the physiological effects are not well understood.
---p 80 (the pdf page #), line 29-30: I don't understand why this statement says that the gas-phase direct phytotoxic effects were not included in this diagram (Figure ES-3). The gas-phase effects are shown in Figure ES-3 just as they are in Figure ES-2.

INTEGRATED SYNTHESIS (CHAPTER 1):

---Chapter 1.4, **Gas-phase direct phytotoxic effects**: The current understanding of gas-phase direct phytotoxic effects is well summarized. I would only point out that there is no clear demarcation in our current understanding of how much of the atmospheric N or atmospheric N + S effects in a given area are due to direct gaseous uptake by plant canopies versus deposition to external canopy surfaces or ground-level surfaces that are washed off by precipitation and transported into the soil (i.e., leading to potential soil-mediated deposition effects).

Likewise canopy uptake of atmospheric N and S can cause phytotoxic effects—manifesting itself as visual injury or nonvisual harm. Phytotoxic effects appear to be uncommon in the U.S., except possibly near some point sources, or within some urbanized areas. But physiological effects from direct canopy uptake of atmospheric N and S is likely widespread---and may or may not be causing what we would call harmful effects. Studies that look at canopy uptake of gaseous N and S pollutants suggest stomatal uptake is widespread, but quantification of uptake and the physiological processing, effects and importance of this uptake is more of a black box---it is not well understood.

What I am suggesting is that discussions of direct effects of SOx and NOx in the ISA not be limited to phytotoxic effects, but at least somewhere in this section, the wider range of effects be considered---including phytotoxicity and visual injury, nonvisual physiological harmful effects, to possible fertilizing effects that may or may not be considered ecologically desirable.

---Chapter 1.5, **Terrestrial nitrogen enrichment and acidification**: This section is very nicely written, providing a clear summary. I would just note, as the authors of the ISA may be aware, that the Horn et al. publication on individual forest tree species mortality and growth responses across the U.S. will likely be published within the next 4-8 weeks. If so, and this analysis can be included in the ISA, this will make a major contribution to the ISA and to our understanding of NOx and SOx effects on forests across the entire country.

---page 42, (Section 1.5.1.2) lines 13-14: Although the valid point is made here that it is difficult to assign a single national CL value---nonetheless, I would just mention that in the Clark et al. 2018 publication (cited below Figure 1-7; and is now published), the minimum Cl was relatively uniform across the country (200 – 400 μeq/ha). Thus, this may function as a general rule of thumb and is an intriguing summary point from this study.

---pp 56 (Section 1.5.3.3), lines 1-2: I suggest editing the last part of this sentence describing deposition something along these lines: "…..base cation uptake, and the type and accuracy of deposition estimates (i.e., wet or bulk, vs. total; measured or modeled)".

APPENDIX 4:

---pp 4-4, lines 4-5: There are a couple of more recent studies on this that can be cited:

Houlton, B.Z., Morford, S.L. and Dahlgren, R.A. 2018. Convergent evidence for widespread rock nitrogen sources in earth's surface environment. Science 360, 58-62.

---Table 4-2: Consider citing the following study:

Avila, A., Aguillaume, L., Izquieta-Rojano, S., García-Gómez, H., Elustondo, D., Santamaría, J. M. and Alonso, R. 2017. Quantitative study on nitrogen deposition and canopy retention in Mediterranean evergreen forests. Environ. Sci. Pollut. Res. 24, 26213-26226.

---pp 4-11, lines 11-13: Seems this sentence would be more clear if the word 'decreasing' is removed.

---pp 4-13, lines 34-35: A recent paper on the importance of on-road or mobile sources of NH3 could be cited here as well:

Fenn, M.E., Bytnerowicz, A., Schilling, S.L., Vallano, D.M., Zavaleta, E.S., Weiss, S.B., Morozumi, C., Geiser, L.H. and Hanks, K. 2018. On-road emissions of ammonia: An underappreciated source of atmospheric nitrogen deposition. Sci. Tot. Environ. 625: 909-919.

---pp 4-65, lines 24-26 discuss increasing DOC in streamwater in Hubbard Brook, while lines 34-35 mention decreasing DOC in Hubbard Brook, apparently in soil solution. It would be helpful to further clarify this potential discrepancy and make it more explicit that lines 34-35 are referring to DOC in soil solution. If the link is possibly due to recovery from soil acidification maybe this should be briefly mentioned here?

---pp 4-117, Table 4-19 (Continued): For the first entry in the table on this page (Decomposition/mineralization), it mentions new studies. There is a recent study showing evidence that the old paradigm that lignin degrading enzymes are suppressed by high N is not correct:

Albright, M.B.N., Johansen, R., Lopez, D., Gallegos-Graves, L.V., Steven, B., Kuske, C. R., and Dunbar, J. 2018. Short-term transcriptional response of microbial communities to nitrogen fertilization in a pine forest soil. Applied and Environmental Microbiology. 84: e00598-18. 18 pp.

In this study by Albright et al 2018, they state: "We found little support for the conventional view that high N supply represses the expression of genes involved in decomposition."

EDITORIAL SUGGESTIONS for Appendix 4:

---Tables in Appendix 4: The heading for the last column (literature references) refers to HERO ID. Is this necessary?
---Table 4-3, in the text and in the References section: The surname Kopacek is distorted, presumably because a foreign language special character did not translate into the pdf correctly.
---pp 4-65, line 26: acidification is misspelled. Also, line 26 needs editing.
---In several places the surname Kopacek is distorted, presumably because a foreign language special character did not translate into the pdf correctly.
---pp 4-66, line 11: Needs editing because of redundant wording.
---pp 4-92, line 9: Delete the word “is”.
---pp 4-97, acronym definitions listed at the end of the table. AOSR is defined, but I don’t see that the Athabasca Oil Sands are mentioned in this table.
---pp 4-109, line 26: Insert ‘a’ before ‘brief’.
---pp 4-112, line 3: Change ‘and’ to ‘a’?

<u>APPENDIX 5:</u>

---pp 5-8, lines 23-25: Seems that this sentence should also mention that Table 5-2 more specifically refers to Ca, and sometimes Al, addition studies.
---pp 5-28, lines 21-22: Seems that something should also be said in summary in relation to the fact that sapling or tree growth wasn’t affected, at least not yet, by the N + S treatments in these two studies---this seems important to note here.
---pp 5-39, line 3: The wording ‘and plots that received higher rates of N’ is too vague even though median values are given; be good to edit to show more specificity as to how high are these higher rates of N.
---pp 5-42, line 1: Although target loads are briefly defined very early on in the ISA it might be useful to define what is meant by a target load at the beginning of this section and paragraph, especially because here it says that target loads were calculated; even though it becomes more clear later what is meant by target loads in this study. Target loads generally consider policy and economic factors, for example. Are those ‘calculated’? Some clarification or definition might be needed up front here as to what these target loads refer to.

As further context, I see that the document does give a more specific definition of target loads for the study discussed on pp 5-46 (see lines 3-4 for the definition).

EDITORIAL SUGGESTIONS for Appendix 5:

---pp 5-41, lines 4-5: Change “…when exposed to wet deposition….” to something like “….based on wet deposition fluxes….”.
---pp 5-47, lines 9-11 and lines 16-17, 21: in lines 9-11 the wording is awkward. Maybe ‘although’ could be replaced with the word ‘with’. In lines 16-17 the wording needs serious editing. In line 21 the word ‘and’ is missing before pH. In line 27 a space is needed after the literature citation.

<u>APPENDIX 6:</u>

---pp 6-25, line 31: Some editing is needed to specify the direction of response for tree ring width and xylem conduit density.
---p 6-42, lines 16-17: Might not want to generalize too much from one study; I'd suggest rewording this sentence to say that this is enhanced herbivore feeding may be one mechanism by which N may alter lichen community composition.
---p 6-46, (Fig. 6-3): I'm not clear why the studies listed in grey text (N-induced C sequestration studies) are included in the figure when no data points are shown for them.
---p 6-64, lines 17-26: Adding 200-300 kg N/ha/yr for 7-11 years to these low productivity alpine systems seems excessive if one wants to relate responses to realistic N deposition fluxes.

---case study: SEKI is more like central California. Maybe rename this case study "southern/central California".

---pp 6-160, line 26: Presumably this should refer to "average" or "seasonal average" NO2 concentrations.
---p 6-161, lines 1-4: But wasn't the N deposition range quite narrow for this study? Might mention it was only in southern Ontario.
---p 6-161, lines 18-21: Maybe add "low biomass systems" to this list of characteristics affecting ecosystem sensitivity to added N (e.g., arid shrublands versus temperate forests).

---p 6-165, lines 15-35 (LIMITATIONS/ISSUES ASSOCIATED WITH EMPIRICAL CLS):

---p 6-168, lines 18-21: I think the actual CL values were 10-11 (not 10-12; see also table 6-24).
---p 6-173, Table 6-25: Linder et al. 2013 is listed as a southern California reference, but this study is for work in arctic Alaska. Is this properly cited in the table?

EDITORIAL SUGGESTIONS for Appendix 6:

---pp 6-31, lines 24-25: This portion of the sentence needs reformatting: "…. the results of Treseder (2008) meta-analysis…".
---pp 6-90, line 14: Ceanothus is misspelled.
---pp 6-138, lines 7-8 (also pp 6-140, lines 13-14; pp 6-157, line 35; pp 6-163, line 25 & 27): There are two dark dot symbols in this sentence, that seem out of place for punctuation. Possibly happened when the pdf file was created.
---pp 6-186, line 14-15: Here it refers to CMAQ simulated N deposition as "deposition data". I suggest rewording this to make it clear these are not empirical deposition data, but simulated deposition.

<u>APPENDIX 16, SOUTHERN CALIFORNIA CASE STUDY:</u>

---pp 16-149, lines 19-21: This is a classic case of issues associated with relying too much on simulated data that hasn't been validated with empirical data, particularly at smaller spatial scales. As you can see from the NADP data shown in figure 16-48, it seems unlikely that oxidized N is that much greater than reduced N deposition in JOTR. As further evidence, we also measured throughfall deposition in the park in 2006 and found NH4-N and NO3-N deposition in throughfall to be nearly equivalent---but of course this is complicated by canopy effects on throughfall fluxes. Generally canopy retention of NH4-N is greater than for NO3-N (so throughfall underestimates NH4-N deposition), especially in arid climates, so these data also argue for a much greater proportion of NHx deposition than TDEP simulations suggest. These throughfall data were used by Leela Rao et al. in their publications, but I don't think data on proportions as NH4 vs. NO3 were included.

In summary, seems that some text should be added to clarify lines 19-21 to the effect that the NADP data (not sure how or if you can refer to our additional throughfall evidence---but see Fenn et al. 2018 below) strongly suggest that the TDEP model as shown in Figure 16-47 is greatly underestimating the importance of reduced N in JOTR. See Figure 7 in Fenn et al. 2018 (citation below). If we had included the data for throughfall under juniper and pinyon pine trees in JOTR to Fig. 7 you can see how this spatial pattern of the importance of both reduced and oxidized N extends from the LA Basin eastward into the desert. The Leifer et al. 2017 study that we cite in this paper also documents the significant transport of NH_3 from the LA Basin, including from some dairy farms east of LA, eastward towards Palm Springs (and JOTR).

Fenn, M.E., Bytnerowicz, A., Schilling, S.L., Vallano, D.M., Zavaleta, E.S., Weiss, S.B., Morozumi, C., Geiser, L.H. and Hanks, K. 2018. On-road emissions of ammonia: An underappreciated source of atmospheric nitrogen deposition. Sci. Tot. Environ. 625: 909-919.

---pp 16-155, Figure 16-51: Be useful to mention in the caption that site CA-75 is in SEKI and that CA-99 is in Yosemite NP.

---pp 16-156, lines 1-5: I think these statements may need some modification. Again, caution must be used in applying simulation models such as TDEP or CMAQ to understand spatial variability in deposition over relatively small scales (compared to regional or national scale) or to estimate with confidence absolute deposition values.

The statement that N deposition is greater in SEKI than in JOTR may be true as TDEP indicates, considering the strong N emissions source area in the Central Valley (major ag emissions and highways and cities). Wet deposition is greater at SEKI of course---JOTR is a desert. However, I've measured throughfall in SEKI and JOTR and for both parks the highest throughfall deposition values were very similar (10-12 kg N/ha/yr). CastNet doesn't do a good job of estimating dry deposition, so you can't rely on that for comparison, at least not with high confidence. Simulated deposition is highly uncertain for various reasons as well---they are better for broad scale. So maybe the statement that SEKI deposition is higher than Joshua Tree should be couched with more uncertainty.

Thus, regarding the following statement on spatial variability, as a minimum a clause should be added at the end such as: "….according to the TDEP simulations".
"There is also much greater spatial variability in N deposition in SEKI than JOTR, according to the TDEP simulations."

---pp 16-156, lines 4-5: Regarding this statement on oxidized versus reduced N, see my comments above and note that NH4 in wet deposition (NADP/NTN data) is considerably greater than NO3 in wet deposition at all 3 parks shown in the figures---casting doubt on the accuracy of this statement. This is empirical data that I would trust more than TDEP output, although admittedly these are only wet deposition data.

---pp 16-156, lines 16-18: Lee et al. 2016 acknowledge in their paper that on-road NH3 emissions are not "shown here" (I assume they mean they weren't included in their simulations); but they acknowledge their greater influence near urban areas. Plus they probably don't account for the concentration of dairy emissions east of LA (Chino/Norco area). In any case, simulation outputs should be interpreted in light of empirical data when possible.

---pp 16-158, lines 22-23: This sentence should be reworded slightly for clarity; When I first read it, I understood it to say that this may be occurring in JOTR. So I found this sentence surprising considering the several publications documenting this phenomenon of N deposition enhancing the growth and fuel buildup from invasive grasses leading to more frequent fire in Joshua Tree NP, which can burn and destroy the namesake species. See studies by Rao et al. and also summarized in the Fenn et al. 2015 book chapter.

---pp 16-164, Table 16-31: The first two CL values given in the table (32 and 39 kg N/ha) based on the study of Rao et al. 2010 are off by a factor of 10; The conversion from the units given Rao et al. 2010 (0.32 and 0.39 g N/m2) to kg N/ha may have been done wrong or the decimal points were left off. The correct CL values are 3.2 and 3.9 kg N/ha for CB and PJ, respectively (as also given in the text on page 16-165).
NOTE: Seems this error is also propagated into Figure 16-54 (see the last 2 CLs shown in the figure).

---pp 16-167, lines 19-21: Repeats the same statement as found on page 16-166, lines 26-28.
---pp 16-169, line 4: Chaparral is misspelled.
---pp 16-169, lines 5-6: Soil acidification in soils of mixed conifer forests in the San Bernardino Mountains should also be included in this sentence (described also in Fenn et al. 2011a).
---pp 16-173 (Table 16-33): I suggest changing the year of the Grulke et al. reference to 2009 (also change in the list of References). I am a coauthor and have a copy of the book—it was published in 2009.
---pp 16-175; Fig. 16.54: The third CL from the top shown in the figure is for the NO3 leaching CL for the Sierra Nevada Mountains (value given is 2 kg N/ha/yr). The citation given is Fenn et al. 2011a, but I'm assuming this is referring to subalpine watersheds and the study by Baron et al. 2011a (see pp. 16-171, lines 21-24). Would be good to indicate that this is for subalpine watersheds or high elevation lakes.
By comparison, the empirical CL value for mixed conifer forests in the southern Sierra Nevada and San Bernardino Mountains (mid-elevation foress) is 17 kg N/ha/yr (see Table 13.5 of Fenn et al. 2011a).
---Fig. 16.54 continued: Likewise, the last two CL values (Rao et al. 2010) also have the incorrect values (correct values are 3.2 and 3.9 kg N/ha/yr). See comments from page 16-164, Table 16-31 above for details).

<u>ADDITIONAL COMMENTS:</u>

---pp R-189): The same reference by Schirokauer et al. 2014 is cited twice (as 2014a and 2014b).
---R-133: The reference by Linder et al. 2013 is missing the name of the journal: Open Journal of Air Pollution

Dr. Ivan Fernandez

I applaud the work of EPA in addressing the major restructuring of the second draft ISA. The Executive Summary and Chapter 1 lay out the primary scientific findings of this assessment, identifying linkages with the 2008 ISA and determinations of causation. The reduced length of the presentation embodied in Chapter 1 provides greater accessibility to the key findings while linkage to the appendices supports the discussion in the primary literature. The inclusion of the effects of changing precipitation and temperature through grounds the discussion in the contemporary realities of ecological responses to nitrogen and sulfur.

There are a number of minor issues and editorial improvements throughout the draft that should be carefully evaluated and improved to be sure the quality of the final document is worthy of the high quality of scientific work done by EPA in developing this second draft. There are times when it is not clear why a primary reference versus a reference to an appendix are used, or sometimes no specific reference is given for a point made, although all statements appeared to be supported by the supporting appendix. There are also times when the attempt to condense the findings of a paper into a single sentence results in ambivalence as to the meaning. In the final version the extra sentence to clarify should be included where the authors feel it is necessary.

Specific comments follow.

PDF Page	Line	Comment
PREFACE		
56	20	The dot likely should be a hyphen in this line and this appears to be a typographic issue from the title of the document and throughout.
EXECUTIVE SUMMARY		
63	6	refer(s)
65	13	Delete comma
69	Table ES-1	Excellent summation table. A minor cosmetic suggestion is to adjust columns in the final format to avoid word wrapping ‘causal relationship’ or other terms for the Current Draft ISA column, allowing both the 2008 and current ISA to look the same when they are the same.
72	2	‘…integrated (in)to a single…’
75	24	‘…availability of (base) cations…’
77	23	I propose rewording this sentence to clarify as ‘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.
78	22	Here sulfur is spelled out, and in other places calcium or aluminum was spelled out, just about carbon was not, and regularly N or Hg are not. Is there a convention being followed as to when elements or ions are spelled out or abbreviated?
80	11	Replace ‘leaked’ with ‘lost’? The loss referred to here largely includes ‘leaching’ losses of N from agricultural fields, but also water and wind erosional losses of N-rich surface soils. Thus, ‘lost’ seems a better term.
CHAPTER 1		

83	27	Delete the second 'related'
	Footnote 1	The constituents listed for NOy here are inconsistent with the species listed for NOy in the legend for Figure 1-2. This is also the issue with p. 84 line 8.
84	28	Delete 'are'
85	15	The term 'laboratory and field additions' here might be open to interpretation for some readers. Perhaps something like 'experimental laboratory and field additions of the pollutants'?
92	7	I wonder if here or elsewhere where the first causal determinations are stated if it is useful to identify the period of literature encompassed. That is, literature supporting this causative determination goes through May 2017, correct?
93	10	'sulfate' spelled out and inconsistent with format of other instances using the symbol
93	11	'…that drought 'can' increase lake S load…'
93	18	Table 7-6 appears to be water quality criteria, not deposition data for estuaries.
93	24	Change 'biogeochemistry' to 'biogeochemical'
93	29	Change 'It' to "This definition…"; insert 'a' before 'better'
96	1-2	I do not know what 'the long-term sustainable deposition is indicated.' Means as used here. Clarify?
100	Footnotes 7, 8	As stated here, it refers to a transference ration as converting deposition of NOy or SOx to air concentrations. It would seem that should be the reverse. The 2011 PA states (ES-9) "F3 and F4 reflect transference ratios that convert ambient air concentrations of NO_y and SO_x, respectively, into related deposition of nitrogen and sulfur."
108	11	The wording seems a bit awkward. What about "…it is discussed in the ISA to better describe how the criteria pollutants NOy and the PM component of NH4+ along with NH3 determine N deposition."
110	18	It's not clear to me how 'ISA' fits into the sentence. As reported in the ISA? If so, sentence should be modified to say so. It otherwise, please clarify.
111	4	This Figure 2-4 link takes me to a sulfur figure? Ditto the Figure 2-32 link below.
113	2-9	This section refers to the uncertainty of deposition, notably with regard to surface layer turbulence and surface characteristics. I presume this does not include the magnification of deposition by canopy capture in vegetated landscapes, and as such, might that be made clear here?
114	23	'…from (the) 2008 ISA…'
115	17	As used, 'Padgett et al.' should be before and outside of the brackets.
116	18	Change 'and' to 'that are'
117	2	Comma after 'consequence'
117	11	Change 'chemistry' to 'chemical'
117	12	'…and some models are well established.' Refers to what? This statement is vague.
118	9	Seems 'biogeochemical' is better than 'geochemical' here given the preponderance of N transformations in soils as part of most of the mechanisms described in Table 1-2.

118	10	How about 'Base cations (can) counterbalance…' since acid cations can as well.
118	11	Delete 'solution' as those inputs are to the whole soil system, not just directly to a water substrate
118	13	Change 'this' to 'atmospheric'
120	Table 1-2	For the Decomposition entry, I believe it should be plural as 'meta-analyses'.
121	20	Punctuation dot issue
122	4-6	Suggest revise to "N deposition to soils can decrease surface soil C:N ratio, which can stimulate nitrification when C:N ratios fall below 20 to 25. The NO3- created by nitrification may be leached, biologically immobilized, or denitrified."
122	26	'…increased (+18%) [with N additions], suggesting…'
124	20	Consequence(s)
130	4	Change to '…in all ecosystems, limitations other than N tend to be more marked…'
138	7	Change 'occurs as' to 'results in'
138	14	Change to '…biogeochemistry [with] subsequent…'
142	Table 1-3	For Surface Water (base cation), the Effect text could be modified to say '…have decreased [primarily] in response…' Base cations would have decreased even if deposition did not decline because soil exchangeable reserves would be depleted and replaced by Al. So declining cation leaching is due to both soil depletion and reduced strong acid anion leaching.
142	3	'Traditionally[,]…'
143	3	What does 'chronic conditions' mean here?
143	6	It's not drainage water until it drains, so this should be revised. Change 'enter' to 'exits'?
143	6-8	Is this a quote from a particular study? Seems odd to suggest 'two' parameters are influenced, since most of the time many would change. Is there a point being made here that does not come through from the wording?
143	10-14	This section seems to suggest there is a shift in chronic vs episodic conditions for evaluative purposes, but then gives an example based on ANC with no mention of how the 'shift' is related. This needs clarification.
143	26	Change 'with' to 'that have'
145	19-21	This section uses the terms surface as well as drainage waters. Are they meant to refer to the same thing, or is one focusing on lakes and the other the streams draining the watersheds that feed into the lake? Not clear the intent of the terminology.
145	28-32	This is a long and complicated sentence that loses clarity along the way. Should be correlated 'with'. Gradients of atmospheric and N deposition, but isn't N part of atmospheric? Does atmospheric mean only acidic atmospheric? Is the gradient only high elevation systems, or is that a second concept in the same sentence?
147	22	Does this mean only 'atmospheric' deposition? If so, this should state that so it is clear.

148	1	I presume this means N concentrations in the water column but it could also mean in the biomass itself, so please clarify.
149	20	'…in the U.S. [since the 2008 ISA].'
150	4	Change to kg N/ha/yr.
150	14	Pardo et al should not be in the brackets, only the date. Ref formats throughout the document should be checked.
150	16	What does the '+1.0' mean in these data?
150	22	Change to CL.
150	33-34	Models such as MAGIC project more than just ANC, and include Al or pH. Therefore, this statement appears unsubstantiated as stated.
153	31	Figure 1-9 is blue but not hyperlinked in my pdf
156	4	'The CLs for deposition [for aquatic acidification] are expressed…'
156	32	Delete 'kg'
156	34	Change to 'Adirondack'
157	2	Change 'have' to 'has'
157	28	I would change 'understood' to 'known'. One implies we know little of the mechanisms, while the other says we have not done the work to quantify the contribution.
159	35	'…particularly those that [are] receiving high inputs…'
164	15	Comma after 'waters'
164	15-17	Excess organic matter could come from changing land use as well as increased erosion that might be a climate signal rather than land use change. The contribution from living algae and seagrasses I believe should be balance by photosynthesis as to the effect on net CO2 change in the water column.
166	6	'e'?
166	28	'…[does] not…'
166	29	Period after S in U.S.
167	26	For consistency, change nitrogen to N here and all other instances in the document.
171	3	This sentence should be clear that the CL is for N, and if it relates to total N inputs and not just atmospheric deposition, that should be explained in the statement.
174	1	Change to '…given (specific) iron and DOC…'
174	5	Omit comma
174	11	Since SRB is defined 3 lines above, SRP should be defined here
175	11	Which ISA?
178	20-21	As stated, it suggests there are no regions with decreased N deposition and only increased or steady everywhere. While this is clearly the case for the most part, are there areas like New England with absolute declines? I can't tell from Fig 2-44 if it is suggested by the coloration on these maps. However, given the focus on this area of the country for acid deposition concerns historically, if there is evidence for declines in total N deposition then the statement should be modified to include the concept that some regions show declines in total N deposition although most of CONUS shows increases or steady state. Publications such as Beachley et al. 2016-JSM Env Sci Ecol 4(2):1030; Sullivan et al. 2018. Env Sci Pol 84:69-73, suggest areas of decline.

178	27	Change 'are' to 'is'
179	4-6	This sentence does not read correctly. If you drop the end (at different rates.) it would be fine.
179	7	It would be clearer if the sentence was explicit as to the measurement being discussed. Are these extractable SO4 and NO3 or soil solution phases?
179	11	The distinction here may be supported in the appendix, but equating slow with soil seems an oversimplification. Cation exchange is a fast reaction and changes in the exchangeable base cation composition happen quickly, for example. Where soil solution reflects a dynamic equilibrium with soil, they respond on similar time scales.
179	31	While the statement that only partial biological recovery may be possible is correct assuming a static concept of 'normal', it seems like this is a place to indicate that only partial biological recovery may be possible and given other stressors, the biological characteristics of recovery from acidification and N enrichment may be redefined.
180	35	Change 'described the' to 'demonstrated that'
180	36	Delete 'adjacent'
181	4	Change 'content' to 'concentrations'
181	7	The parenthetical phrase pointing to more details on Al and DOM is fine, but this could be done for many of the mechanistic details housed in the appendices. Why was this one singled out?
182	15	…data (are) insufficient…
182	1-16	Section 1.12 adequately highlights the relevance of changing temperature and precipitation in understanding ecosystem response to changing N and S deposition. However, it neglects to also refer to the importance of rising atmospheric CO_2 in understanding responses and recovery, from the physiological stimulation of vegetative growth to the acidification of surface and marine waters. Is this somewhere else? If not, that linkage should be included in this climate summary.
183	13-14	Something is wrong with the wording. Change '…and…' to '…to the…'?
184	30	Omit comma after CASTNet
184	32	Omit the word 'for'
184	35	Omit the word 'of'
185	22	'from 15% or less to 99%' is confusing. Was there an actual lower number and if not, then would '<15%' accurately and more clearly reflect that boundary, and avoid the phrase in between numbers?
186	32	'of' or 'on' NOx? 'of' suggests that the focus is on the canopy that is altered, 'on' suggests the interaction with the canopy as it affects NOx. Which is intended here?
187	16	There is a word missing. Perhaps '…community compositional (conditions)…'?
189	22	Omit comma after BCw as Li and McNulty used the term 'BCw base rate'
189	33	Omit period after)
189	31-34	The Bonten et al reference is a valuable resource but as used here, this simply says it exists, not what the reference found. This adds little to the message of Chapter 1, but certainly is a valuable resource for the Appendix.

190	27	Add comma after PROPS
191	5	Should this sentence read 'The SPARROW model uses only wet N deposition. A large….'?
192	13	What does '…leaked out of its application…' mean?
Appendix 4. Soil Biogeochemistry		
344	1	Fix brackets on refs in all instances of the document, such as here.
344	28	Change 'an' to 'a'
345	8	'…mobilization of aluminum (Al) cations [of varied speciation], several of which…'
345	10	'determinant[s]' and eliminate the period after 'acidification'
345	12	'The [accelerated] loss of base cations…'
352	Fig 4-3	In this draft pdf, this is an unacceptably poor quality reproduction of the figure.
352	3	I would insert the word 'chronically' before 'exceeded' since some N always leaches but that is different from the concept referred to here of saturation.
353	18	Are those superscript commas supposed to be del notations in this line?
353	9	Replace the comma with the word 'of'
353	11-13	Besides saying the studies exist, can something be said about how they inform the issue of N leaching?
353	32	What does 'where N demand is not indicated' mean?
354	4	Fix spelling of Kopacek throughout document, including Table 4-10
354	17	Delete second 'with'
356	Table 4-3	I wonder if table titles can be improved or more descriptive. Table 4-2 is Pathways and Pools, and yet this table includes 'leaching' which might be considered a pathway. What are the intended conceptual divisions in these table data?
362	17	Change 'is' to 'can be', since slow release is a function of deposition. This is stated implicitly as a recovery phase statement, but not explicitly.
363	13-14 (also Table 4-4 for base saturation)	This statement is probably the opposite of how these two properties interact during acidification. This is true, I believe as reflected in the references regression, when considering future correlations. Soils with high SO4 adsorption capacity will, during the recovery period, have more SO4 release for longer periods of time, thereby having a faster rate of base cation depletion compared to soils with low SO4 adsorption capacity during the recovery period. This mechanism can be more clearly described here.
367	3	I would delete 'until stores become depleted' since a 'store' is poorly defined and if it includes mineral base cation pools they will never be depleted. Nor are they truly depleted, but rather approach a dynamic equilibrium of increasingly lower base saturation during the acidification phase.
368	6	Change to Bear Brook Watershed in Maine
368	30	Change 'to' to 'for'
368	31	Delete first 'sources of'
375	26	Delete first 'dissolved'
376	5	Delete 'water'

380	8-10	The Mitchell reference seems to indicate that sugar maple influences organic matter quality related to increased rates of nitrification. The paper does not appear to report a positive correlation between organic matter and nitrification.
380	10-12	The Russow ref is presented as a contrast to Mitchell, yet they were about two different things. Russow indeed described high organic matter soil as
		challenging process differentiation because of high NH4 adsorption, largely focusing on denitrification pathways. This appears to be at least an oversimplification of the findings that leaves a possibly incorrect impression.
380	33	forest(s)
380	35	'…ratio(s) may also have negative relationship(s) with…'
388	2	Delete period after 'matter'
388	18-21	The meaning of this sentence is a bit obtuse. It reads like litter decomp is from different microorganisms, and then lists roots? And heterotrophs? Seems like it wants to say these are the sources of soil respiration, but rather points to litter decomposition? Clarify.
395	30	forest(s)
396	7	The Mineau findings are not the same as Fatemi (SBB 98:171-179)
397	5	Change 'are' to 'were' and 'is' to 'was'
397	7	forest(s)
405	25	Delete the 'a', and also, why 'In contrast'? Driscoll say DOC goes up as pH goes up, and Fuss reports DOC goes up as Hubbard Brook recovers (as in, pH goes up).
406	11	Delete 'caused'
408	28	Delete 's' from roots
409	7-8	Change 'has' and 'is' to have' and 'are'
414	5	summarize[s]
414	10	'…[of] N…'
414	13	'…[to] six…'
420	6	Delete 'the reversal'
429	1	Delete 'in'
431	11	Period after 'leaching'
431	13	No superscript for 14.
431	15	Superscript comma? Issue throughout this section.
447	5	An eastern forest fire and N reference perhaps not captured in this review would be Kahl et al. 2007 Env Monit Assess 126:9-25.
448	28-29	Something is missing in this section?
448	3-5	The wording in this sentence seems off. Do you really mean the adsorption capacity of P, or the adsorption capacity of the soil for P? And 'uptake by plants for productivity' appears to be about increasing productivity?
448	5	Delete 'increased'
448	17-18	Change to 'As precipitation and runoff patterns change with a changing climate, this important process will be affected.' Changing hydrology in the Northeast is well documented and underway, not just a future effect.
448	20	switch 'in' and 'is'

449	14	Relevant to the weathering mechanism here besides Belyazid is a paper by Kopacek (EST 51:159-166) showing the effects of precip on contributions from scree in some watersheds.
450	10-19	Contosta et al (GCB 23:1610-1625) offers an useful analysis of the changing winter on ecosystem dynamics including N cycling for this section.
450	11	What effects of snow depth, increasing or decreasing, relevant to the changes noted in the rest of the sentence?
450	20-21	Study the effects of effects? Study different pools and processes related to pools and fluxes? Revise wording.
451	13	Delete 'The'
451	14-16	Revise this sentence to read 'It is important to consider net ecosystem flux because consumption may be offset by production.'
451	30-32	To clarify this important point, suggest '…global terrestrial carbon sink, [the GHG benefits of an atmospheric] CO2 reduction could be…'
453	3	Change 'and' to 'a'
453	9	Insert 'atmospheric N deposition is' before text in brackets
454	7	I would encourage clarity in reference to 'upper soil horizon' here and anywhere forest soil depth inferences are made. If this phrase is code for the O horizon or forest floor, it should be explicit. If it is meant to refer to the upper mineral soil, that should be clear as well. In many of the forests studied for S and N impacts, particularly in the northeaster US, there is a dramatic difference between the surface O horizon and underlying mineral soil horizons in most response mechanisms and values on this subject.
458	24	'match' in what way?
458	27	Fix brackets.
458	27-32	For the Yanai reference it would be good to state what the starting point was as well as describe the final 8 kg N/ha/yr sink status for comparison. For the Mitchell reference it would be useful in the summary to say what they found for retentions from the data rather than just saying the study exists. The next ref of Lieb gets an extensive description of findings.
459	6-8	Change to 'relation[ship]' and to '…leaching [in] regions…'
461	26	Fix brackets on this McNulty as it is part of the sentence.
462	33	To clarify this important point, suggest '…global terrestrial carbon sink, [the GHG benefits of an atmospheric] CO2 reduction could be…'
Appendix 13		
1258	22	pool(s)
1258	28-31	There is no issue with the text here, but this identifies issues of note that literature was, as yet, unavailable for in this review. While the Greaver ref adequately addresses possibly effects of N deposition on C cycling, there is little explicit discussion of the potential for the physiological response of plant communities to rising CO2 (the fertilization effect) to modify the response to N deposition, although the text recognizes a fertilizer effect. If this is not covered elsewhere in the ISA, then here might be a place to discuss it. The concept is addressed at some level in Galloway et al. 2014.

1263	25-28	I recommend that quoted text from Greaver et al. 2016 be quoted exactly as published (thus the concept of quoting), even when there may be an error in the published text. The statement here appears to say 'while' while the actual text I believe says 'whereas'. The original text says 'though' (which I think was supposed to be 'through'?), but the text here uses 'although', which I believe also both provides a different meaning and does not make sense in the sense. Please verify all quoted text for this chapter. A cut and paste from the Greaver publication is:
		Some studies show that climate change will mitigate acidification through increased weathering67, whereas others show that climate change will aggravate acidification though increased nitrification outpacing enhanced weathering68.
1263	32	Missing end quotes.
1263	2	Increase(s)?
1263	10	Missing end quotes.
1263	24	Missing end quotes.
1265	Sect. 13.2	This section on estuaries does not mention the contribution of atmospheric N sources to coastal acidification. References like Gledhill et al. 2015, Oceanography 28(2):182-197 touch on this linkage but I suspect there is a literature on this now available that is beyond my familiarity.

Dr. Frank Gilliam

Section 1.8 of the Integrated Assessment highlights potential effects of excess N and, often related, acidification on a wide variety of wetland ecosystem types, from bogs and fens to coastal wetlands/estuaries. Full disclosure, I am not a wetland ecologist, so my comments need to be taken with that in mind. The section begins with an important distinction between most wetlands and their terrestrial counterparts—atmospheric deposition of N and S does not elicit acidification response in wetlands. Conversely, excess N can bring about numerous changes in many components that threaten the structure and function of wetland ecosystems. These include biogeochemistry and the ecophysiology of plants.

As stated in this section, the outcome of the earlier ISA is quite clear—*the body of evidence is sufficient to infer a causal relationship between N deposition and the alteration of biogeochemical cycling in wetlands*. On the other hand, both spatial variation and variation among types of wetland often preclude broad generalizations, wherein they can serve as either a source or a sink for a variety of N compounds. The figure referenced herein (Figure 11-2) is an extremely useful summary of quite recent N-manipulation studies examining N processing in contrasting wetlands, including coastal marsh, mangroves, riparian wetlands, and bogs.

The ISA is similarly clear regarding the effects of excess N on wetland biota—*the body of evidence is sufficient to infer a causal relationship between N deposition and the alteration of growth and productivity, species physiology, species richness, community composition, and biodiversity in wetlands*. A conclusion of responses highlight what is generally known about unimpacted wetlands, regardless of type: they are typically highly N-limited. Thus, the initial responses to added N is that they exhibit characteristics along a gradient toward N saturation. Initially, there is enhanced growth and net primary productivity, plant tissue N increases, along with profound shifts in plant and microbial communities. These rates of these changes decrease over time, as these system move toward N saturation.

Similarly, biodiversity, particularly of wetland plant communities, declines in the face of N enrichment. This can be especially serious, considering the hallmark of wetlands as have such high plant species richness. Moreover, numerous endangered species can be threatened by excess N. Ultimately, this section highlights yet another parallel with terrestrial ecosystems regarding N enrichment. Excess N decreases abundance/richness of sensitive species, whereas it increases abundance/richness of tolerant species.

Comments on Section 1.4 Gas-Phase Direct Phytotoxic Effects

This brief section documents that little new work has been done the direct effects of SO_2, NO_2, NO, PAN, and HNO_3 on plants. Early conclusions, however, had been quite clear that that is a causal relationship between plant exposure to all of these and injury to vegetation.

Dr. Daven Henze

Chapter 1.3

27.13 (and similarly, 28.9): This summary of PM2.5 composition seemed a bit vague. What is meant by most (51%? 90%?) and what is meant by most areas (based on land area? Or where people live? Or number of counties?)? Further, as noted in Appendix 2 and referenced works, a significant fraction (~50%) can be carbonaceous matter, and yet the inorganics are the focus not so much because they dramatically dominate the carbonaceous matter mass, but because they can be chemically identified. Note it is stated (2-3, line 9): "As a result, the main contributors to PM2.5 mass for which ecological impacts can be readily assessed are limited to sulfate and nitrate". Lastly, 28.16 states that "In the eastern US, NO_3^- and SO_4^{2-} make up an even greater portion of PM2.5 mass in areas were PM2.5 mass is the highest…" isn't supported by the figure presented (or my general familiarity with the topic); in locations like PA, NJ, NY it would appear that these two indeed make up close to 50%, but NO_3-percentage is smaller than in areas like southern CA or the midwest. Highest nitrate levels are usually associated with combination of transportation and agricultural (NH3) sources, which isn't necessarily where $SO4^{2-}$ is highest.
28.28: AMoN

29 – 29: Summary of NH3 measurements is thin. Should include SEARCH, and remote sensing. SEARCH is covered in 2.4.1 but not 2.4.3.1, which is odd. Should individual measurement campaigns for NH3 be discussed (CAMNet, aircraft from CalNex…)? These sorts of measurements provide valuable information about diurnal variability of NH3 (SEARCH), vertical profiles (aircrafts), and widespread seasonality and long-term trends (remote sensing) that are not necessarily evident from AMoN. The remote sensing section itself 2.4.3.2 needs to include CrIS, which arguably has the best remote sensing of NH3 to date, since 2011. See for example the remote-sensing measurements of N dry deposition in Kharol et al. (2018). If a main uncertainty is NH3 seasonality (29.12), this could help.
- Note: this discussion important, as ISA highlights (p102, lines 24-26) that the largest source of uncertainty in atmospheric science section is NH3 deposition and emissions.

29.5: Studies have also used statistical models, machine learning, land use regression, and global models to estimate the distribution of NO2; so, the options go beyond regional models and satellite data. Summary of satellite NO2 data also thin.

Further, I think more could be said about remote sensing. There isn't any summary here of 2.2.4.2, and also 2.2.4.2 doesn't even mention remote sensing of NH3. If the main uncertainty is NH3 seasonality (29.12), remote sensing could help.

29.10: This is a rather subtle distinction, but I'm not sure the CTMs alone do well at predicting long-term changes, in absolute value, of concentrations of these species. Really it is the accuracy of emissions estimates that drive these. The models are used for predictions of the relative change in concentrations over long periods of time following e.g. some future policy scenario.

29.15: A topic sentence or two that transition from the title of 1.3.3 (deposition) to the initial discussion of emissions would be useful.

1.11: Recovery

No comment

1.12: Climate modification of ecosystem response to impacts of N and S addition

Overall, section 1.12: Overly brief and not providing much detailed information. The appendices on this topic are quite extensive, full of qualitative and quantitative discussion of dozens of recent works. However, the summary is rather cursory, and more points to the existence of research in these areas rather than a summary of research findings. For example:
- 101. 1-3 and 6-9: This summary presents the existence of recent work on climate impacts by Pinder et al. (2012) and Greaver et al. (2016). However, the actual findings of these works are not provided. It would be useful if they could be briefly summarized here.
- 101.10: It seems like the few studies of S on climate that are identified (Mitchell et al. 2011; Rice et al. 2014) would be easy to summarize.

1.13: Uncertainty

The introduction and overview of uncertainty is interesting. It does though seem to setup the framing of uncertainty discussion through these specific definitions (e.g., ontic vs epistemic), which, however, are not used in the subsequent discussion (unlike, e.g., the framework of uncertainty characterization in IPCC assessments that is used throughout). Should the generic definitions of uncertainty thus be shortened, or should this language be incorporated into the rest of this section? Or these definitions of uncertainty should be moved to 1.2 (Concepts)?

103: Soil NOx is cited as a particularly large source of uncertainty in Appendix 2.2.3 (page 2-14) but it is not mentioned here in the summary of NOx emission uncertainties, which focuses on combustion / transportation.

104.12: There are also remote sensing measurements for NH3.

103/104: Partitioning of SOx between SO2 and sulfate is an important model uncertainty that is mentioned in the Appendix (2-60) but not in the summary; this should probably be included, as it may help explain why uncertainties cited for the emissions of SO2 are so much smaller (10-15%) than uncertainties in the concentration estimates (up to 47%).

1.14: Ecosystem Services

This overview also seemed a bit brief, and is just an abbreviated copy of section 14.6. Acidification and eutrophication are summarized, but the section on nitrogen and climate modification (14.3) is not. Section 14 contains tables of quantitative results and summaries that could serve as a valuable source of information to distil and present in an overview, such as the estimated range of ecosystem services costs per N in table 14-2 from Sobota et al. (2015) [note formatting issue in citing this paper in the table].

Also, why does this section come after the discussion of uncertainty? Seems a bit out of place, and should go before uncertainty review, and the latter should cover ecosystem services.

Reference:

Kharol, S. K., Shephard, M. W., McLinden, C. A., Zhang, L., Sioris, C. E., O'Brien, J. M., ... Krotkov, N. A. (2018). Dry deposition of reactive nitrogen from satellite observations of ammonia and nitrogen dioxide over North America. Geophysical Research Letters, 45, 1157‐1166. https://doi.org/10.1002/2017GL075832

Dr. Donna Kenski

Charge: *(a) comment on whether the revised Executive Summary and Integrated Synthesis convey the main findings of the ISA; (b) comment on how effectively the revisions to the ISA reflect the recommendations and comments received from CASAC and public comments; (c) identify any additional revisions to the ISA that will substantively strengthen the identification, evaluation, and communication of the main scientific findings.*

Executive Summary and Chapter 1.2, Integrated Synthesis and Connections, Concepts, and Changes: In general, this is another great job by EPA staff tackling a tremendously complex subject. The Executive Summary and Integrated Synthesis are generally accurate and concise condensations of the much more comprehensive Appendixes. I especially appreciate the addition of Sec. 1.13 summarizing key uncertainties—it is helpful to have this discussion pulling together the varied aspects of uncertainty in one place.

The 'Connections, Concepts, and Changes' is a useful addition, since the scope of this ISA is so broad. It serves as a nice review of the basics, as well as an introduction for readers who may not be well versed in every aspect of deposition science. All efforts to bring order to this very complex topic and convey a sense of the bigger picture that arises from the multiplicity of N/S/PM deposition effects are much appreciated. Toward that end, Figure ES-2, ES-3, and 1-4 are excellent visual summaries.
The inclusion and discussion around NHx have improved since previous reviews, but sometimes its treatment is still spotty. For example, on page 3, the top bulleted list should have a bullet for reduced nitrogen and the text should specifically mention NHx (like on p. lxvi, lines 7-8). The third bullet in this list (PM) should elaborate a little more, in parallel with the previous 2 bullets, to call out the components of PM.

Section 1.2.2.6 is titled Scientific Advancements of the Aquatic Acidification Index (AAI). It is a nice summary of the AAI and its history, but doesn't describe any new application or advancement of AAI since the 2011 PA. I suggest retitling the section 'Aquatic Acidification Index'.
Appendix 2: Thanks for the careful attention and responsiveness to our comments on the last draft ISA. I found the revised discussion of the monitoring network strengths and weaknesses for estimating deposition much improved. Similarly, the additional information on transference ratios, bidirectional exchange of NHx, and model uncertainty adds valuable and relevant detail that was previously lacking. Figures 2-18, 2-26, and 2-32 (2001 county-level emissions) are hopelessly outdated and apparently there just as placeholders for more current data. Please replace with gridded emissions, not county-level, as the variation in county size makes meaningful spatial comparisons next to impossible. With those exceptions, the maps are great and having emissions, concentration, and deposition closer together makes the spatial connections easier to see.

Chapter 1.3: Emissions and Atmospheric Chemistry: This chapter was a model of brevity, given that it condenses 130 pages of Appendix 2 into 6. I am tempted to ask for more information to be included here on transference ratios and modeling methodology and uncertainty, but probably the document as a whole is better served by keeping this summary short. One minor revision is needed: Section 1.3.2, paragraph 2 (p. 29 lines 4-6) says that "unmeasured component species of NOy and concentrations of all NOy species in data-sparse regions must be provided by regional models in conjunction with satellite data." So far, NO2 is the only component species detected by satellites, and the only one described in Appendix 2.4.2. This paragraph should be reworded to be more precise about the abilities of satellite data to detect NO2 vs. NOY and more accurately convey the information from App. 2.4.2.

Chapter 1.10: Ecological Effects of Particulate Matter other than N and S Deposition: I concur with the addition of the 'likely causal' statement that was added to this section. The studies cited and summarized in Appendix 15 provide sufficient evidence to support this determination.

Chapter 1.13 Key Scientific Uncertainties: Thanks for adding this section and also Sec. 2.2.3 on Emissions Evaluation and Uncertainty. I found it quite helpful to have this information gathered together in one place. It highlights the varying quality and quantity of uncertainty information in different disciplines. In Section 1.13.1.1, please note that activity estimates are also a large source of uncertainty in mobile source emissions.

Minor comments, typos:

What are the weird little dots after some words in the Executive Summary (p. lxiii, line 2 and footnotes, for example, but many other places as well)?

p.13, lines 2-7: the Nilsson-Grennfelt definition of CL was defined in the previous paragraph, doesn't need to be repeated here. It is also repeated on p. 15, lines 30-31; probably not needed here either.

p. 15, line 9: values -> value

p. 17, line 1: is it -> it is

p. 18, line 19: DON and DIN have not been previously defined

p. 20, line 34: delete 'in'

p. 22, first subheading in Table, phototoxic -> phytotoxic

p. 28, line 28: the network is typically abbreviated AMoN, not AmoN. Also, it should appear in the list of abbreviations but does not.

p. 103, line 32: delete 'for'

p. 103, line 35: delete second 'of'

p. 104, line 11: 'cloud top pressure' or 'cloud height' would be better than 'cloud pressure'

p. 2-34, line 13: should this be 'direct measurements of NOy' ?

pp. 2-77, 2-78, 2-84, 2-85, 2-90, figure captions: Clear -> Clean

p. 2-56, line 23: add a period after NHx

p. 2-104, line 20: Add 'background concentrations' after 'Estimated PM2.5'

p. 2-104, line 34: Adjoint

Dr. William McDowell

Additional comments on ISA

The revised discussion of effects of changing atmospheric deposition on DOC and TOC concentrations (Appendix 7.1.2.9) effectively describes the latest literature on the effects of changing acid deposition on surface water DOC.

Dr. Erik Nelson

Executive Summary

- I am having trouble understanding the placement of the four boxes on the bottom of Figure ES-1 (e.g., Ecosystem Services). Are these four dynamics placed here because they are informed by the exposure and biological effects information above? The relationship between these 4 dynamics and the exposure and biological effects information could be made clearer.
- Figure 1-2 is very informative and helpful for understanding the basic science that this ISA covers. Does it belong in the Executive Summary as well? As I read the chemistry alphabet soup on pages lxvi – lxviii I was hoping for a figure that tied everything together. Figure 1-2 might serve this purpose.
- Lines 35- 36, page lxvii to line 2 of page lxviii: Does this mean that **inorganic** N species are routinely monitored but **organic** N species are not? If this is the distinction please make clearer.
- I wonder if Table ES-1 is necessary given the causal relationships are also summarized in Figure ES-2. A table that covers over 3 pages is hard to read and digest. Figure ES-2 presents Table ES-1's data in one page. Therefore, why present the harder to read Table as well? I understand that the Table shows the causal determination in the previous ISA. But is the ES the place to show the causal determination in the previous ISA? That is a detail that could be presented later (in fact, it is presented later in Table 1-1); I do not think previous ISA information is important enough for the ES. By deleting that column of previous ISA determination in Table ES-1 the table and Figure ES-2 become redundant and the more cumbersome Table ES-1 could be deleted.
- Does the ISA ever explain how the strength of a causal relationship between the criteria pollutants and the ecological effect is determined? Or is this algorithm described in US EPA 2015e? The ES needs to better explain where the algorithm for creating the five-level hierarchy of ecological effect evidence can be found.
- The sentences found on lines 10-13 of page lxxv are confusing. I think the ES would read much better of you simply deleted those two sentences. As of now the two sentences muddle the essential point: ANC is a measure of the buffering capacity of natural waters against acidification and that waters with low ANC cannot avoid the effects of acidification and associated ecological effects.
- In the section entitled "Nitrogen (N) Enrichment/Eutrophication of Terrestrial, Wetland, and Aquatic Ecosystems" should we acknowledge the issue of separating N enrichment/eutrophication due to deposition from N enrichment due to agricultural and storm water runoff? Does the impact of enrichment/eutrophication on ecosystems differ across the two sources? The public is likely to be more aware of N enrichment/eutrophication problems due to run off. Do we have to take pains to highlight this ISA covers N enrichment/eutrophication due to deposition only while the impacts of N enrichment/eutrophication are driven by both processes?
- Does the ES need to quickly define what an ecosystem service is? I am not sure if the ecosystem service concept is that well known yet.

- The ES says the following: "New valuation studies for ecosystem acidification pair biogeochemical modeling and benefit transfer equations informed by willingness-to-pay surveys, especially for the Adirondacks and Shenandoah regions." (lines 36, page lxxix – line 1, page lxxx). I think it should say: "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."
- The Ecosystem Services section is missing a very important paper:

  Keeler, Bonnie L., Jesse D. Gourevitch, Stephen Polasky, Forest Isbell, Chris W. Tessum, Jason D. Hill, Julian D. Marshall. 2016. The social costs of nitrogen. Science Advances, e1600219.

  According to the abstract: "Despite growing recognition of the negative externalities associated with reactive nitrogen (N), the damage costs of N to air, water, and climate remain largely unquantified. We propose a comprehensive approach for estimating the social cost of nitrogen (SCN), defined as the present value of the monetary damages caused by an incremental increase in N."

  With their method a researcher can generate a number akin to the Social Cost of Carbon (SCC) but for Nitrogen. If, for example, a policy causes N deposition on a landscape to increase by 100 kg then the monetary damage caused by that policy in terms of N would be SCN x 100 where SCN is measured in the present value of damages in $ per kg of N. Of course, much of SCN accounts for damage to human health so we would have to remove this damage component from SCN for the purposes of the secondary NAAQS assessment (the assumption is that the remaining SCN is due to N's impact on ecosystem services).

  In my opinion Keeler et al. (2016) is just as important as the other ecosystem service research mentioned on lines 5 – 13 of page lxxx.

Chapter 1.2

- Page 1, lines 5 – 7: The sentence "This ISA communicates critical science judgments of the ecological criteria for oxides of nitrogen, oxides of sulfur, and particulate matter" does not make sense. Judgments of the ecological criteria for oxides…? Maybe "judgments on the ecological effects of oxides…"
- Page 1, lines 9 – 12: "Welfare effects" refers to the impacts on the well-being of people and animals. The "welfare" of soil, water, etc. is not impacted by pollution. Instead soil, water, etc. processes, integrity, and functionality are impacted by pollution. Please only use the word "welfare" when referring to the well-being of people and animals.
- Figure 1-1 does not include a box for literature suggested by experts or the public at the 2014 and 2015 meetings.
- Page 7, line 15: Why is the subscript on the "O" in "NO" "Y"? Does that need to be explained?

- Page 7, lines 15 – 17: A proposed line edit: "Emissions of NOY, SOX, and PM ~~cause~~ **contribute to** an accumulation of N and S in the environment that creates a multitude of effects on terrestrial, wetland, and aquatic ecosystems." This edit accounts for other sources of N and S, such as runoff, that are contributing to accumulation of N and S in the environment.
- Page 9, lines 8 – 10: A proposed line edit: "The importance of preserving biodiversity and ecosystem function contributes to the sustainability of ecosystem services that benefit human**'s** welfare and society **in general** (Chapter 1.2.2.4 and Appendix 14)."
- Page 9, lines 11 – 20: We should clarify that loads **above** a CL in an ecosystem will lead to a change in the ecosystem properties or processes. In most cases these changes will negatively affect the well-being of humans that interact with the ecosystem. Conversely, loads **below** a CL in an ecosystem will likely mean maintenance of current ecosystem properties or processes and associated ecosystem services.
- Page 9, lines 16 – 18: A proposed line edit: "~~Use of~~ **For** CLs **to be used** in evaluating the effects of **N and S** deposition upon ecosystems **they must be able to differentiate** N and S ~~consider how~~ deposition **from** ~~compares to~~ other anthropogenic and ambient sources of N and S to these ecosystems (Chapter 1.2.2.2.)."
- Page 10, lines 18 – 20: A proposed line edit: "However, only some organism-level endpoints such as growth, survival, and reproductive output have been definitively linked to **pollution** ~~effects~~ at the population level and above (U.S. 19 EPA, 2013b)."
- Page 11, lines 2 – 3: A proposed line edit: "Other ecosystems may be profoundly altered if a single attribute is affected **by pollution**."
- Page 11, lines 18 – 21: We cite studies that "find human-mediated watershed N inputs that range from <1.0 to 34.6 times the rate of 20 background N input (Appendix 4.2)." But what fraction of this is due to deposition and what fraction is due to other sources of input (e.g., runoff)? In lines 17 – 18 we say that atmospheric deposition is the main source of anthropogenic N to unmanaged terrestrial ecosystems. However, the quoted sentence above does not support our "main source" claim, it just states the extent of overall N input to ecosystems.
- Page 11, lines 21 – 23: Regarding the sentence "Across all watersheds, atmospheric N deposition is the second largest overall human-mediated N source and the largest N source to 33% of watersheds." To be complete we should mention the largest overall human-mediated N source.
- Page 12, line 16. A proposed line edit: "Freshwater inflows to estuaries ~~may contain~~ **transport** N from agriculture, urban, wastewater and atmospheric deposition sources."
- Page 12, line 16: A proposed line edit: "In fresh surface waters and wetlands, ~~sources of~~ S ~~that contribute to~~ enrichment ~~effects 3 are the same sources of S that~~ induce**s** acidifying effects**. Sources of S** ~~these sources~~ include weathering of minerals in sediments and rocks, leaching from terrestrial S cycling, internal cycling, and direct atmospheric deposition."
- Page 12, line 20 – 22: Regarding the sentence "The importance of atmospheric deposition as a cause of estuarine eutrophication is determined by the relative contribution of the atmospheric versus non-atmospheric sources of N input." This is the case for **ALL** ecological effects we are looking at! In every system we need to determine the relative contribution of the atmospheric versus non-atmospheric sources of N and S input! A version of this sentence needs to be prominently featured earlier in the chapter.

- Page 13, lines 2 – 8: Much of this language repeats what was said in page 12, lines 25 – 30. Please avoid unnecessary repetition.
- Page 13, lines 2 – 8: Again, we need to emphasis that whether or not an ecosystem has N and S loads or concentrations above a CL will be a function multiple sources. Therefore, critical question for researchers are: 1) how much does atmospheric deposition contribute to a CL exceedance and 2) if the atmospheric deposition was decreased by X% would N or S load or concentration fall below the CL threshold? This last question is critical to designing effective policy interventions.
- Page 14, lines 23 – 27. Doesn't this paragraph apply to S inputs as well? Only N inputs are mentioned.
- Page 15, lines 9 – 11. The sentence in these lines can be deleted. The previous sentence demonstrates the point we are making sufficiently.
- Page 15, lines 14 – 15. Why? Because systems can more readily adapt to perturbations in the long run?
- Page 15, lines 30 – 33. This is the third time this definition has been given! Please delete this repetitive text!
- Page 15, lines 33 – 36. You may want to move this alternative definition to the point in the text where you first write out Nillson and Grennfelt's definition.
- Page 16, lines 3 – 4. A proposed line edit: "There are causal relationships between **additions of** N and/or S **to a system** and biodiversity loss in terrestrial, freshwater, wetland, and estuarine ecosystems in the U.S. (Chapter 1, Table 1-1)."
- Page 17, lines 8 – 11. Do we mean "**ecosystem process**" instead of "**ecosystem service**" in the following sentence: "Accelerating ecosystem service declines in response to species loss may be due to multifunctionality, which suggests that different ecosystem functions require the presence of different sets of species (Isbell et 10 al., 2015; Reich et al., 2012; Zavaleta et al., 2010)."
- Page 17, after line 19. I feel like we need a summary sentence or two here explaining why we devoted a whole section to the links between biodiversity decline and N and S additions. Is this here because we believe this is the key ecological response to N and S additions? In other words, are we arguing 1) N and S additions to an ecosystem reduce or at least change the biodiversity found in a system, 2) these changes to biodiversity in a system affect its ecological processes and functionality, and 3) some of these changes affect human welfare? Is that why we have this section in here? If so we need to explicitly say this. Or is this section in here simply because biodiversity was one of the ecological features that the secondary standard covers? Either way, we need to tell the reader why this section exists.
- Page 17. I suggest we change the title of section 1.2.2.5 to "Reduced versus Oxidized Nitrogen Effects across Ecosystems" to "Reduced versus Oxidized Nitrogen Impacts on Ecological Processes across Ecosystems."
- Page 17, line 22. Please reiterate the main source of NO_Y and the main source of NH_X.
- Page 17, line 26. Please reiterate the main source of NO_3^- and the main source of reduced forms of N.
- Page 17, line 28. Is NH_3 / NH_4^+ a reduced form of N?

- Page 17, lines 28 – 35. It seems like we are suggesting that reduced forms of N are added to an ecosystem via atmospheric deposition. Is this true? If so, please be more explicit about this.
- Pages 17 – 18. A table that indicates what ecological processes and what species are affected by reduced form N (atmospheric N?) versus those affected by oxidized N would be helpful. The same table could indicate the sources of reduced form and oxidized N.
- In section 1.2.2.5 most of the discussed impacts involve changes in biodiversity structure in an ecosystem. Is this way we had a section on biodiversity before this section? But on page 18 we read about the how the form of N affects biogeochemical processes as well. So why do we have a chapter section on biodiversity and how it is impacted by N and S additions but we do not have a chapter section biogeochemical processes and how it is impacted by N and S additions?
- Page 18, lines 13 – 15. Suggested line edit. "This result suggests that terrestrial community diversity is also generally not **differentially** affected **by the form of N**, possibly because plant uptake of N is mediated by soil biogeochemical cycles that often rapidly transform N between oxidized and reduced forms."
- Page 18, lines 30 – 31. How were the percentage of water bodies to protect selected? I do not understand how we go from a distribution of CLs to some sort of selection algorithm.
- Pages 18 – 20. Can the AAI differentiate between atmospheric sources of N and S and land-based sources of N and S? If it can't how useful is it to our endeavors?
- Page 20, lines 19 – 21. So there is no new understanding of the sources of **S** deposition and in the relationship between atmospheric concentration and deposition.
- As I argued above, given Table 1-1 I suggest we delete Table ES-1.
- Page 26, lines 2 – 6. Suggested line edit: "This new research confirms the causal relationship between N **loading**, **either via atmospheric** deposition**, runoff, or both,** and ecological effects documented in the 2008 ISA**. Further, the new research** ~~and~~ improves our understanding of the mechanistic links that inform causal determinations between N **additions via atmospheric** deposition, biogeochemistry, and biota in terrestrial ecosystems (Chapter 1, Table 1-1)."
- Page 26, after lines 6 – 10. Suggested line edit: **"Therefore, assuming we can differentiate between atmospheric loading of N and other sources across ecosystems, we can determine how rates of atmospheric deposition impacts whether or not these newly identified CLs are exceeded.**"
- Page 26, after lines 11 – 15. Suggested line edit: **"Again, assuming we can differentiate between atmospheric loading of N and other sources across ecosystems, we can determine how rates of atmospheric deposition impacts whether or not these newly identified soil CLs are exceeded.**"
- Page 26, lines 30 – 32. Suggested line edit: "With increasing N inputs to coastal waters, **both due to atmospheric deposition and land-based runoff,** CO_2 in the water column is produced from degradation of excess organic matter from changing land use, as well as respiration of living algae and seagrasses, which in turn can make the water more acidic (Appendix 10.5)."
- Chapter 1 barely discusses PM. We need some language that explains why this is.

Recovery, climate modification, key scientific uncertainties, and ecosystem services (Chapters 1.11, 1.12, 1.13, and 1.14) Lead discussants are: *Drs. Daven Henze, James Boyd, Stephen Schwartz, Lauraine Chestnut, Erik Nelson*

- Page 97, lines 20-21. Regarding the sentence: "Overall N emissions and deposition have been increasing or relatively steady; consequently, there has been little reported on N enrichment recovery." I would assume there are some regions where N enrichment recovery has occurred. Can't we use regional analysis to become more informed on N enrichment recovery? For example, I think later on we say that recovery has occurred to some extent in parts of the northeast US.
- Page 97, lines 26 – 29. Regarding the sentence: "For acidification caused by N and S deposition, chemical recovery of aquatic and terrestrial ecosystems are characterized by trends in water quality indicators…" So are we saying that chemical recovery from acidification caused by other sources of N and S are characterized by **different** trends in water quality indicators? I assume no matter the original source(S) of acidification, recovery is characterized by the same "trends in water quality indicators (NO_3^-, 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."
- Page 98, lines 4 – 6. Suggested line edit: "When evaluating ecosystem recovery **from** ~~to~~ acidification, it is important to note that different chemical pools within the soil **or water column** may recover at different rates with the same decreases in declining atmospheric deposition at different rates.
- Page 98, lines 22 – 25. Is this the only definition for recovery? Is this the best definition of ecological recovery? A mimicking of pre-industrial conditions seems like a very narrow definition of recovery. How about this for a definition: "a system that generates ecological processes and functionality similar to those found in the latter half of the 19th century has recovered"? Isn't this last definition more attuned to the interdependencies between humans and ecosystems? I would assume we care more about recovering processes and functionality than duplicating conditions that existed 150 years ago.
- Page 99, lines 9 – 12. Suggested line edit: "In areas where N and S deposition has decreased, chemical recovery must first create physical and chemical conditions favorable for growth, survival, and reproduction **of the pre-industrial assemblage** in order for biological recovery to occur."
- Page 101, lines 12 – 16. The text in this section is a bit unclear. Are we simply saying that our understanding of climate modification of ecosystem response to N and S addition is too immature and uncertain to make this a major focus of this round of secondary criteria analysis?
- Section 1.13. I like the introduction to the uncertainties section. As we go through the different systems that affect our endeavor could we indicate 1) which system (e.g., atmospheric science, ecological science, etc.) contributes the most uncertainty to our results, 2) the type of uncertainty that dominates in that system (e.g., statistical, scenario, etc.) and 3) ways uncertainty can be reduced for each system?

- Section 1.13. What about the uncertainty in determining how much N in a system came from atmospheric deposition and how much came from land-based inputs? I assume determining the relative contribution of atmospheric deposition to loading in a system is important. I assume there is uncertainty when calculating this relative number?
- Section 1.13. I would like a concluding paragraph that gives some guidance on what we should do with all this data on system uncertainty. How should it affect analysis of the secondary standard, for example? How should uncertainty affect our judgments on the relative ecological health and integrity of ecosystems? Any help on these questions would be greatly appreciated.

Chapter 1.14

- One measure of the economic value of removing N from the landscape is $5.91 / kg per year (mean) or $10.50 / kg per year (high end). These values are based on the cost to remove a kg of N from a community water system. Wang et al. (2017) used USEPA (2008) to get these values. Wang et al. (2017) does not give the year of the $ estimate.
- Estimated <u>annualized</u> value of N mitigation service ($/kg N) in Arkansas in 2008 $ is $25.27 (mean), $22.82 (low), and $106.09 (high) (Jenkins et al. 2010).
- Keeler et al. (2016) measured the social cost of nitrogen (SCN) in Minnesota. They note that each kg of N applied to a field generates four compounds: NO3-, N_2O, NH_3, and NO_x. The total annual damage done by the four compounds (gases?) measured in $ / kg of N applied to a field is $2.62 (mean), $0.44 (low), $10.79 (high). To convert annual values to a net present value the authors assume a twenty-year time horizon and a 3% rate of discount. This conversion generates values of $40.15 (mean), $6.74 (low), $165.34 (high) per kg of N applied. These values account for the damage done to water quality (from N as NO3-), changes in climate (from N as N_2O), and changes in air quality (from N as NO_x, NH_3, NH4NO3, and (NH4)xSO4). All $ are in 2010 $.

Wang, Yangyang, Shady Atallah, Guofan Shao. 2017. Spatially explicit return on investment to private forest conservation for water purification in Indiana, USA. Ecosystem Services 26, Part A: 45-57.

Jenkins, W. Aaron, Brian C. Murray, Randall A. Kramer, Stephen P. Faulkner. 2010. Valuing ecosystem services from wetlands restoration in the Mississippi Alluvial Valley. Ecological Economics 69 (2010) 1051–1061.

Keeler, Bonnie L., Gourevitch, Jesse D., Polasky, Stephen, Isbell, Forest, Tessum, Chris W., Hill, Jason D., and Marshall, Julian D. 2016. The social costs of nitrogen. Science Advances 2 (10): 10.1126/sciadv.1600219.

Dr. Hans Paerl

Please note: My comments mainly deal with impacts of atmospheric N enrichment on estuarine and coastal waters, specifically potential linkages of N enrichment on pH (acidification) of receiving waters. I am concerned that the "connections" between N enrichment are largely speculative and not supported by long-term monitoring of pH and related environmental variables in estuarine ecosystems, specifically the two largest systems in the US, Chesapeake Bay and the Albemarle-Pamlico Sound System. Below, I elaborate on this in my responses to what has been written in the *Review* as well as the *Integrated Science Assessment for Oxides of Nitrogen, Oxides of Sulfur, and Particulate Matter: Ecological Criteria (Second External Review Draft)*

3-23, lines 1-6. It is stated that "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."

I don't agree with this statement. The "new scientific information" is perhaps (but not for certain) mainly relevant to oligotrophic open ocean water, not estuarine and nearshore waters. A recent study by Baumann and Smith (2018)[1] of long-term data bases of pH and trophic state (as Chla) on numerous EPA-NEP and NOAA-NEERS estuarine sites, shown no clear relationship between trophic state and acidification. Furthermore, long-term (>20 year) data bases from Chesapeake Bay (Courtesy of Chesapeake Bay Program) and the Neuse River Estuary, NC (Courtesy of Neuse River ModMon Program), show a great amount of variability, and no clear trend in pH (Figs. 1 and 2). Acidification is controlled by multiple interacting factors including rates of primary production (CO_2 fixation) which have been increasing due to eutrophication, tending to drive pH up, and mineralization of autochthonous and allochthonous organic matter, driving pH down. The net results are highly variable. One important fact is that with regard to autochthonous (within system) processes, it is impossible to mineralize more organic matter (driving pH down) than what is produced by autotrophs (algae and higher plants) (driving pH up), So, with regard to eutrophication, one might expect pH to rise, unless every C molecule that is fixed is mineralized, in which case one would expect no net change in pH. In the Neuse R. Estuary, it looks like pH has risen at upstream station 70, while at downstream station 120 there is no significant trend (Fig. 1).

[1] Baumann, H. and E.M. Smith. 2018. Quantifying Metabolically Driven pH and Oxygen Fluctuations in US Nearshore Habitats at Diel to Interannual Time Scales. *Estuaries and Coasts* 41:1102–1117 DOI 10.1007/s12237-017-0321-3.

Figure 1: pH data from the eutrophic Neuse River Estuary, collected by the UNC-CH IMS ModMon project (http://www.unc.edu/ims/neuse/modmon/) below:

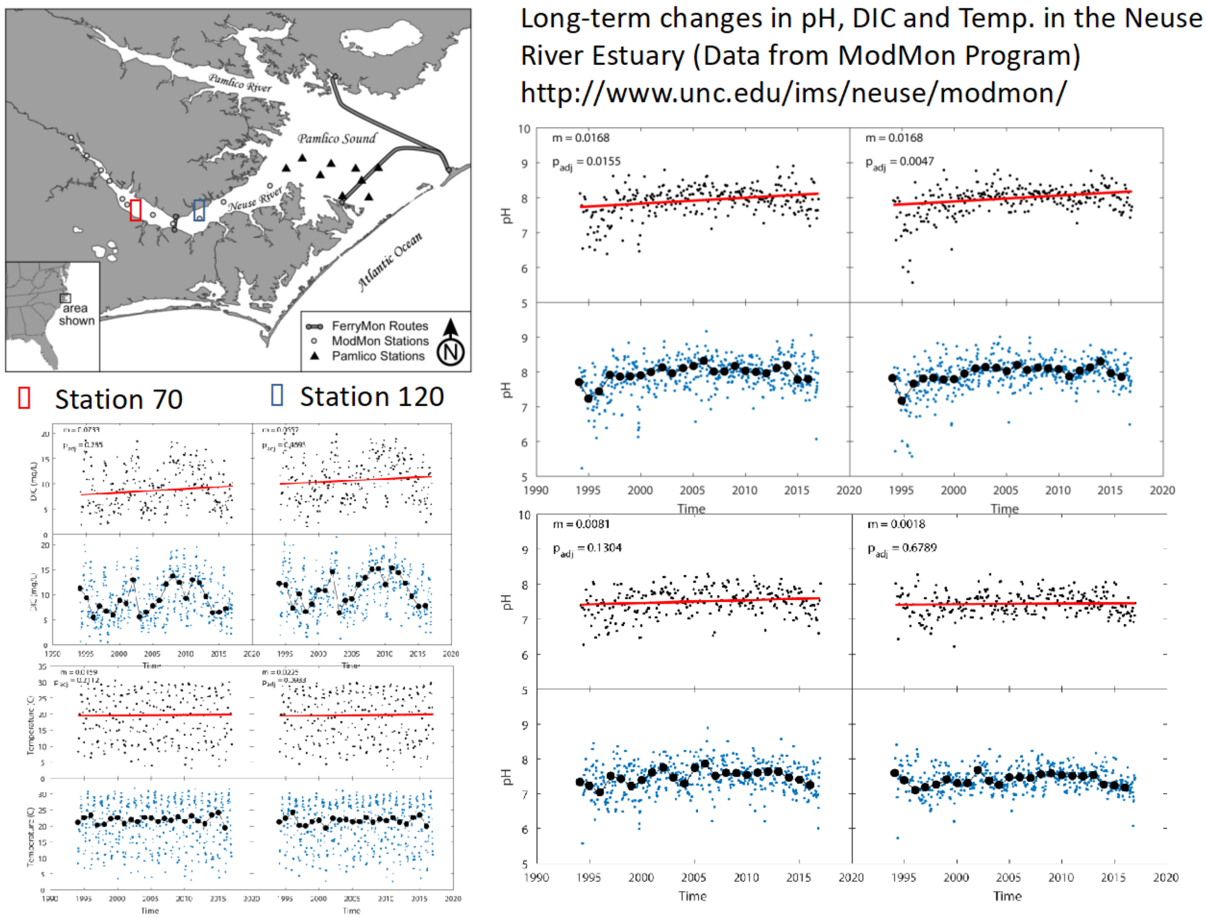
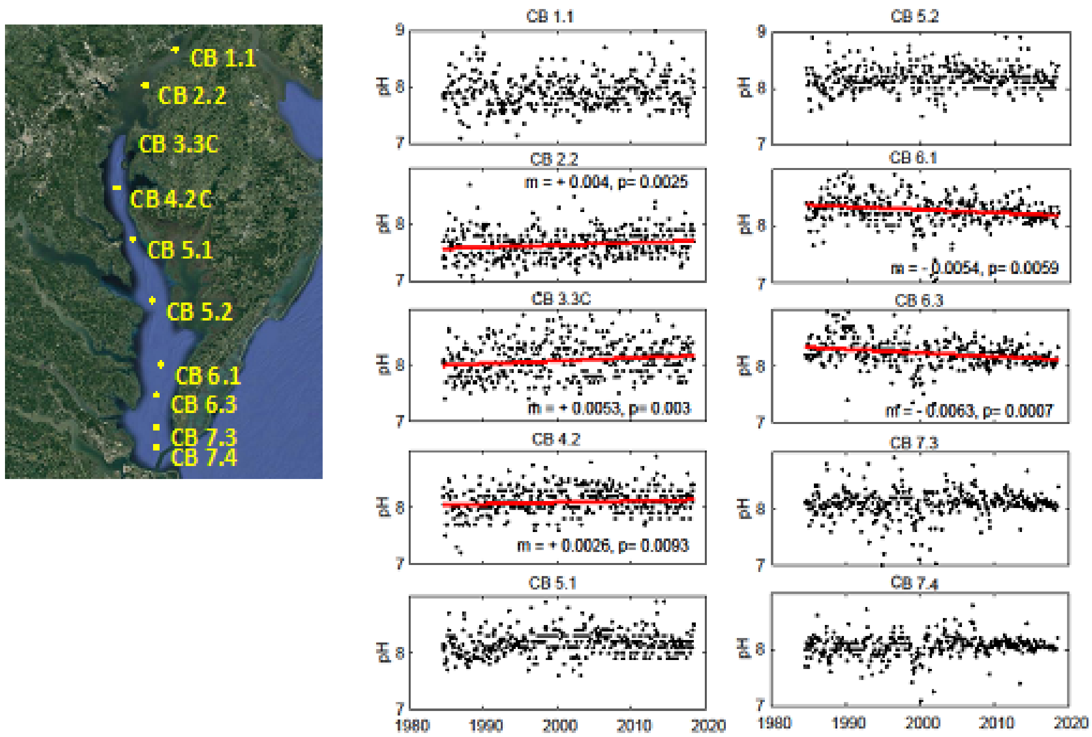


Figure 2: Long-term pH data from various locations in the Chesapeake Bay form 1984-2017. Data are courtesy of the EPA Cheapeake Bay Program, 2018 (https://www.chesapeakebay.net).



Results from a seasonal Kendal Test. m values are Sen slopes (units /year) and p-values have been adjusted for serial correlation.

I therefore caution against making the statement “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.”

This caution also applies to similar statements made in the Integrated Science Assessment for Oxides of Nitrogen, Oxides of Sulfur, and Particulate Matter: Ecological Criteria (Second External Review Draft), APPENDIX 10. BIOLOGICAL EFFECTS OF NITROGEN ENRICHMENT IN ESTUARIES AND NEAR-COASTAL SYSTEMS.

Section 10.1 **Introduction**
Line 8. There is no “hard data” to support the “role of N in nutrient enhanced coastal acidification (Appendix).” (More below on this topic).
P. 10-2, line 8. “altered growth, total primary production” could be changed to "altered (stimulated) primary production"
Lines 16-19, the statement is made “The body of evidence is suggestive, but not sufficient to infer, a causal relationship between N deposition and changes in biota, including altered physiology, species richness, community composition, and biodiversity due to nutrient-enhanced coastal acidification.” I do

not agree with this statement. The biggest problem isn't acidification, but rather eutrophication and associated detrimental effects (HABs, hypoxia, food web issues, etc.)
P. 10-7, lines 26-27, it is stated that “The authors suggest that increased N deposition may enhance primary production and potentially lead to a shift from N to P limitation in this region” (referring to upper North Pacific Ocean).
This is quite speculative, and there's no real evidence that this is taking place, especially if we invoke denitrification as an important N sink.
P. 10-9, line 22. What is meant by “physical”?

Section 10.5 **Nutrient Enhanced Coastal Acidification**
On P. 10-53, lines 32-34 state “several studies have suggested that the increased respiration caused by N enrichment may exacerbate coastal ocean acidification through alteration of the carbon cycle (Appendix 7.2.4).” However, at the same time, N-driven eutrophication (higher rates of primary production) has driven pH up. Therefore, the two processes have opposite effects on pH, with the net effect likely being no consistent trend in pH.

P. 10-54, in response to the text on lines 10-17, there is no conclusive evidence from intensive monitoring programs on the waters of Chesapeake Bay or the Neuse River Estuary, NC (largest tributary of the Pamlico Sound) that they have become significantly more acidic in the past several decades.
Also, on P. 10-54, in response to lines 18-26, Acidification is more likely observable in open ocean environments, but may be masked by enhanced primary production (eutrophication) in estuarine and coastal waters where it will lead to increases in pH. Furthermore, it is difficult to envision how more organic matter can get mineralized (depressing pH) than is produced photosynthetically (causing the pH to rise). I don't feel comfortable pushing the ocean acidification issue, especially not in coastal and estuarine waters, where no clear trends have been shown to exist.
See Baumann and Smith 2018, Estuaries and Coasts 41:1102-1117.

P. 10-55, Figure 10-10. The Effects of nutrient-driven eutrophication (increased rates of primary production, leading to increases in pH) are not included in this schematic.
This comment also applies to lines 12-27 on P. 10-57, which similarly omit the potential for "basification" of estuarine and coastal waters due to N-enhanced rates of photosynthesis.

Section 10.7. **Summary and Causal Determinations**
P. 10-59, lines 28-31 This is speculative and currently not supported with any long-term data set I’m aware of.
P. 10-64, lines 2-5. This is a very weak statement, and for good reason…..there is no convincing evidence for long-term acidification of US estuarine and coastal waters.
Again, see Baumann and Smith 2008, Estuaries and Coasts 41:1102-1116.
Lines 6-20, This is mainly based on discussions of open ocean water dynamics, but there is no long-term monitoring evidence showing a significant trend.

One last comment: P. 10-61, line 20. There's a difference between "seaweeds" and macroalgae. The term macroalgae is probably more appropriate.

Mr. Richard Poirot

Executive Summary and Connections, Concepts, and Changes (Chapter 1.2).

Generally, the Executive Summary presents a clear and concise review of the science underlying the current NAAQS review, with particular emphasis on areas of improved understanding since the last (2008) NOx/SOx ISA. Revisions to the 1st draft ISA and changes in the organizational structure are helpful and responsive to previous CASAC review comments. Figure ES-1 and Table ES-1 are especially effective at concisely summarizing key findings, conveying the organization of the document and providing links to the more detailed discussion in the appendices.

There were several occasions in the ES where it seemed like the writing got a bit careless. For example NOx is repeatedly redefined (in Chapter 1 as well) but is then occasionally used incorrectly (where NOy is intended). Another example is the (unnecessary) use of 3 different end dates (2011-2013) to describe "25 year trends - since 1990 - in emissions, concentrations or deposition - where current (2015 or 2016) data are readily available. See line-by-line comments for additional details. For the most part these inconsistencies are minor and easily correctable.

The Chapter 1.2 discussion of connections, concepts and changes (since the 2008 ISA) is well organized, clearly worded and directly responsive to previous CASAC comments. The summary Figures are excellent, and Table 1.1 provides a concise summary of the identified causality relationships, changes since 2008 ISA and linkages to the more detailed information in the appendices. It might be helpful to include some introduction to the concept of recovery – which is not much discussed until near the end of Chapter 1. If a critical load for terrestrial N enrichment had been exceeded in the past, but is no longer exceeded now, is there an expected ecological response? What are expected (or observed) time frames for chemical recovery of acidified surface waters; how do continuing, but lower levels of acidifying deposition affect the nature and timing of chemical recovery? What are some of the key factors and time frames that may lead to (or impede) biological recovery?

The causality discussion of direct effects of gaseous SOx and NOy on vegetation is uniquely modified by the observation that is little or no evidence that such effects are continuing at current, lower levels of exposure. It seems to be implied, but not always clearly stated, that all the other identified causal associations are occurring at current levels of deposition. If true, it would be helpful to state this more clearly (or to point out other effects that are likely not occurring at current levels of S & N deposition). A phrase like "…due to historical and continuing N deposition" might also be used, as I think it can often be difficult or impossible to separate out the influence past deposition.

Ecological effects of Particulate Matter other than nitrogen and sulfur deposition (Chapter 1.10)

It's a bit awkward to break out the ecological effects of S and N separately from other (visibility and materials damage) welfare effects of the same pollutants "considered in separate NAAQS reviews" - although I sort of get your logic. Still, it would require almost no effort to tack on a visibility module to any CMAQ SOx/NOx model results, and to quantify changes in $PM_{2.5}$ and visibility that might result if SOx and/or NOx emissions were controlled for ecological purposes. It also seems odd to include ecological effects of non-S, non-N PM here, but I think it is more or less "harmless" - and makes sense from an organizational standpoint. There's just not much literature on the ecological effects of other PM species, and your light treatment of this topic is reasonable.

One general criticism is that the soil or "crustal material" component of PM is often neglected or understated. While soil is a relatively minor component of $PM_{2.5}$, it's typically the largest component of coarse particle mass ($PM_{10\text{-}2.5}$), and larger particles dry deposit more efficiently than small ones. Airborne soil could be a significant source of cations (Ca^{+}, Mg^{++}, K^{+}, Na^{+}, etc.) that may partially buffer acidifying deposition, and it's the one component of PM that appears to be increasing - at least in some regions & seasons. You argue that recent widespread increases in P concentrations in lakes and streams might be due to climate induced increases in windblown dust, but don't say much about other soil components. Airborne soil can also provide a source of nourishment and (shade) protection for long-range transport and deposition of pathogenic bacteria & fungi, and there's some fairly extensive literature linking coral reef decline and other marine biological effects to pathogens routinely transported with Sahara dust to the Caribbean and SE US. See for example: Shinn et al. 2000 (https://doi.org/10.1029/2000GL011599) Garrison et al. 2003 (https://academic.oup.com/bioscience/article/53/5/469/241414), Griffin & Kellogg, 2004 (DOI: 10.1007/s10393-004-0120-8). Dust-related "Valley Fever" (Coccidioides) is well documented in dogs, cats and horses, and presumably affects other mammals as well. For example see: https://vfce.arizona.edu/valley-fever-dogs/valley-fever-other-animals

Executive Summary, line-by-line comments:

p lxiii, lines 6-17: The description of "oxides of nitrogen" is notably less comprehensive than that of "oxides of sulfur" (despite the footnote). You might at least add "gaseous and particulate" between "other" and "oxidized" on line 7, although I would list the major NOy species in the text and use the footnote to explain the "traditional" meaning of NOx. Also add an "s" to "refer" on line 6. Your definition of SOx on line 14 is different from that in the glossary (p. xlvii).

p lxiii, line 17: You could add "sea salt" to this list of PM components. At near-coastal/estuarian locations, it can be an important component of coarse and total PM mass (for which speciation is typically not determined) - and to a lesser extent of fine mass). Also, following reactions with HNO_3, "aged sea salt" ($NaNO_3$) - especially from larger particles - might be a significant contributor to particulate N deposition at near-coastal, near-urban sites.

p. lxiv, lines 27-29: As defined on the previous page, the terms NOy and SOx include the particulate forms of oxidized N and S species. So it's unnecessary to add "and particulate" here - or you could say "gaseous and particulate …" if you want to reinforce this.

p. lxvi, Figure ES-1: This an excellent graphic summary of the appendices and the logic of how they fit together! Just below the chart, you define the chemical formulae used in the chart - except you include NOx (not used in the chart) and exclude NOy (which is used in the chart).

p. lxvi, lines 1-9: You mention particulate NH_4 here, but seem to be omitting wet NH_4 deposition - a much more important contributor to total N deposition than PM NH_4 is. Also, please check if the statement "NH_3 contributes 19-63% of total inorganic N deposition" is correct. Are you sure you don't mean NHx (NH_3 + NH_4) contributes 19-63% of N? If not, that (NHx) contribution would be a more useful statistic to present.

p. lxvi, line 11 and p lxvii, line 1: This is an accurate summary of $PM_{2.5}$, but for PM in general (including coarse, which for some species is more important than fine in terms of deposition), you should add crustal material (or soil dust or minerals) and sea salt. At most rural IMPROVE sites, fine soil is typically a larger fraction of fine mass than EC is, and is likely the largest contributor to coarse ($PM_{10-2.5}$) mass at most sites. See for example Malm et al., 2007 (https://doi.org/10.1016/j.atmosenv.2006.10.077). Relatively alkaline crustal material may provide a significant source of Ca, Mg, K, etc., and appears be increasing in some regions & seasons.

Also, while carbon does exist in pure elemental form (EC) in PM, organic carbon (which is typically what's measured in PM) is always present in compounds that typically also include O & H. So "organic matter" or "particulate organic matter (POM)" are more accurate & comprehensive terms for an OC PM component. Fine OC is typically multiplied by a factor of 1.8 to represent fine POM in IMPROVE data analyses.

p. lxvii, line 6: You could add NH_3 to this list of "acidifying" gaseous precursors. At many sites, deposition of NHx equals or exceeds NOy on an equivalent basis. Also, here and on several other occasions throughout the document you keep re-defining "NOx (NO + NO_2)", but this is inconsistent, and sometimes you just use NOx. Pick one or the other, and if you really think constant redefinition is necessary, why not just drop the "NOx" and just refer to "NO and NO_2". Also be careful, as elsewhere in the ES and IS, you sometimes use NOx when you mean NOy.

p. lxvii, line 14: Change "The particulates NH_4^+ …" to "Particulate NH_4^+ …". These ions themselves are not particles, but components of particles. Also (OK, this is really old-school here, but) I remember when "particulate" used to be an adjective…

p. lxvii, lines 21-27: Why use 3 different (and older) end dates - 2011, 2012, 2013 to describe "25-year" trends in emissions or concentrations since 1990? Why mention the decline in NO_2 concentrations but not the decline in SO_2 concentrations? You have the data and could/should describe actual 1990-2015 25-year trends in SO_2 & NOx emissions and SO_2 and NO_2 concentrations (or update all though 2016). In line 26, you could add "and NO_3^-" after "$SO_4^=$."

pp. lxvii-lxxiii: The clear, concise discussion and tabular (ES-1) and graphic (ES-2) summaries of ecological effects, causality framework and changes from last ISA are excellent! Would it be possible to add (under the "causal relationship" notation in the "Current Draft ISA" column of Table ES1), a few phrases like "under current conditions" or "under historical conditions", where appropriate. You could also use a different color in ES2 to distinguish between past vs. present effects.

p. lxxii, line 6: I think you mean NOy, not NOx.

p lxxiv, lines 7-8: This is the first and only reference, so far, to (absence of) effects from current concentration or deposition conditions. Is it to be implied that all other effects are occurring at current conditions? If so, why not say so? Maybe you could do this once here, for brevity, and say something like "For all other identified causal relationships identified in this ISA, the evidence indicates a causal association from current [or current and historical, or past and continuing] levels of S and/or N deposition."

p. lxxvi, lines 10-11: "… N enrichment effect starts with the accumulation of N in the soil." So this historical dep. predisposes the ecosystem to future damage, as N accumulates. However on other occasions you indicate that CLs are often higher as N deposition increases. At some point in the document, it would be useful to see a more detailed conceptual discussion of relationships between cumulative historical deposition, continuing current deposition, critical loads, and recovery. Given the legacy deposition, are current lower rate of deposition "sustainable" without incurring future damage or impeding future recovery?

p. lxxvi, generally: Somewhere in this section, a brief reference to important secondary effects of N deposition - including increases in wildfire fuel and frequency - could be mentioned.

p. lxviv, lines 1-3: "There is also evidence that decreasing sulfur deposition loads over time (observational studies of SOx deposition, experimental studies of simulated SOx wet deposition) result in lower concentrations of MeHg in water, invertebrates, and fish." This is suggestive of chemical and biological recovery. More up-front discussion of the concepts of chemical and biological recovery would be useful. These topics are nicely addressed at the end of the integrated synthesis and in appendices, but could at least be mentioned briefly in the Exec Sum. Wherever possible, I think evidence of chemical and biological recovery should be emphasized, as these provide strong confirmations of causality arguments, and also show that damage is (at least partially) "reversible" - at least in some locations within relatively short time scales.

p. lxxx, lines 29-30: "The gas-phase effects were not included in this diagram." it looks to me like phytotoxic effects from plant exposures to S and N gases are included in both Figures ES-2 and ES-3.

Chapter 1 Integrated Synthesis, line-by-line comments

p. 3, lines 1-3: It seems odd/unnecessary to refer to workshop questions that were considered, without stating them - or at least providing a few examples.

p. 3, line 28: Change "These are types of analyses, if pursued, require…" to something like "These are types of analyses which, if pursued, would require…" Or maybe just drop "are".

p. 4, line 17: …refers to papers published through December 2015, but Figure 1-1 refers to papers published through 7/31/2017.

p. 6, lines 8-21: This emphasis on causality is obviously important (and is generally very well done), but should identifying general causality be the only end goal? In addition to an affirmative indication that a causal relationship exists, it would require relatively little additional text (or space in a table) to indicate 1) whether the effect occurs under current conditions (or as a result of "historical and continuing deposition"), and maybe also 2) what's the approximate geographical extent or regional distribution of effected areas? For #1 above, I would think the oft repeated "confirmed or enhanced by studies published since the last 2008 ISA" would be sufficient to conclude that the effect is occurring under "current conditions".

p. 8, Figure 1-2: In the diagram, you refer to dry deposition of NOx. I think you mean NOy here (which you define below the figure, but otherwise don't use in the figure).

p. 11, line 4: "an essential factor" should be "essential factors".

p. 11, line 13: you could delete "more readily" or change it to "eventually" (more readily than what)? HNO_3 deposits more readily than NO or NO_2, but many transformation products deposit less readily than their precursors - for example NH_4NO_3 deposits less readily than HNO_3 or NH_3 and PM $(NH_4)_2SO_4$ deposits less readily than SO_2 or NH_3.

p. 11, lines 20, 21: Has "background N" (input to watersheds) been defined? Could it be explained here by something like "(without anthropogenic contributions)" - if that's what you mean?

p. 12, line 11: This sounds like a broad proclamation - pertaining to all lakes and all droughts? Should it be constrained/modified to reflect the results of this one study - "in the lake (or lakes) evaluated in this study…"? I would think that for any specific lake, the number would depend on lake (& drainage basin) characteristics - biogeochemistry, history, morphology, etc.

p. 14, lines 23-27: It would be informative to know whether empirical CL thresholds are determined spatially (similar ecosystems experiencing different current deposition) or temporally (a fixed ecosystem experiencing changes in deposition over time). For changes over time, can thresholds be different depending on whether deposition is increasing or decreasing?

p. 15, lines 16-20: Is it possible to comment on whether an empirical CL would be more similar to a dynamic or steady state modeled CL, and why?

p. 20, line 30: I think you probably mean NOy, not NOx.

p. 21, lines 15-17: This causality statement of plant injury from direct exposure to gaseous SOx and NOy is uniquely modified by the qualifying caution that there is little/no evidence this occurs at current levels of exposure. There seems to be an implication that for all the other causality statements the indicated effects are occurring at current levels of S and/or N deposition. If true, you should say so - or you should identify any other effects that may not be occurring under current conditions.

p. 27 (or anywhere in Section 1.2): It would be useful - in this discussion of "Connections, Concepts, and Changes" - to include discussion of something like "ecological responses to changes in deposition". This could include implications of the larger historical and cumulative deposition rates of SOx, NOy, S+N, and in some areas total N; evidence of chemical and/or biological "recovery"; time scales for and asymmetry of recovery; shifts in gradients of effects if specific target loads were met; remedial actions such as Ca additions, etc. An example question that might be posed is "what would be the (range of) expected ecological effect(s) if a CL which is currently being exceeded was attained by decreased emissions?"

p. 27, lines 6-12: This discussion is awkwardly worded and confusing. I'm not sure you mean that NH_3 (dry deposition of) accounts for 19 to 63% of total N deposition (although I suppose that could be true in agricultural areas). Perhaps you mean that NHx deposition accounts for 19 to 63% of total N? Also, this seems to imply that aerosol NH_4 is the only NH_4 in deposition. Wet deposition of NH_4 greatly exceeds aerosol NH_4 deposition everywhere and also exceeds dry deposition of gaseous NH_3 in most non-agricultural areas.

p. 27, line 15: Again, you seem to be ignoring wet deposition (although I suppose the S and N in wet deposition ultimately had either a gaseous and/or PM origin).

p. 27, lines 16-17: You could use the term "organic matter" or "particulate organic matter" rather than "organic carbon". Also, if you're describing total PM, rather than $PM_{2.5}$, then you should include crustal material (soil dust) and perhaps also sea salt. For PM_{10}, both of these may be more important than EC at many locations, even more so for TSP.

p. 28, line 2: NH_4 is also an important component of "acidifying precipitation".

p. 28, lines 11-13: You could mention NOx decreases here too. Something like: "a sharp decrease in SO_2 emissions and smaller but substantial decreases in NOx emissions in recent years have led to corresponding decreases in SO_4, NO_3, and $PM_{2.5}$ concentrations."

p. 28, lines 29-32: This is a bit too simplified. You might rephrase to something like IMPROVE and CSN "measure $PM_{2.5}$ and $PM_{2.5}$ components including NH_4^+ (CSN-only), NO_3^- , and SO_4^{2-}, although these data are typically not used to estimate deposition rates." (Aerosol N data have occasionally been used as surrogates for or indicators of N dep.

p. 29, lines 16-19: See previous comment on these older and different end dates. Update all 3 end dates to 2015 (or 2016) and use similar date ranges in exec summary p. lxvii. Also why not report the 25-yr trend in SO_2 concentrations as parallel to NO_2. See: https://www.epa.gov/air-emissions-inventories/air-pollutant-emissions-trends-data and https://www.epa.gov/air-trends

p.29, line 28, etc. A picky point, but you could update the TDEP maps & discussion to 2015 (2013-2015), which would be consistent with available SOx & NOx emissions, air quality and wet dep data - and give you a nice round 25 years since 1990…

p. 30, line 4: This link to Figure 2.4 leads to wrong figure.

p. 30, line 8: This link to Figure 2-32 leads to wrong figure.

p. 30, line 12: I wouldn't say figure 2-36 really supports this statement that wet S >> dry S, especially in areas of highest deposition. Also, the text mentions (relatively trivial) particulate S dep. but not SO_2 dry dep.

p. 34, lines 18-22: indicate that findings from 2008 ISA (and further supported by more recent studies) provide "several lines of evidence that past and current HNO_3 concentrations may be contributing to the decline in lichen species in the Los Angeles basin." Several comments here:

- This seems inconsistent with earlier claim (p. 31, lines 13-15) that "there is little evidence available to inform whether current monitored concentrations of gas-phase NOy and SOx are high enough to injure vegetation.
- The phrase "past and current" HNO_3 concentrations raises interesting questions about the combined influences of (higher) past and (lower) current exposures. Is it possible that past, cumulative exposures to long-lived species has caused injury from which current lower exposures (though perhaps not sufficient to have caused the injury in the first place) are nevertheless sufficient to prevent (or delay) recovery of the injured species?
- A third dumb question raised here relates to the distinction between damage caused or contributed to by "direct exposure" to a pollutant like HNO_3 vs. damage caused or contributed to by "dry deposition" of HNO_3 and other N to plant surfaces. There seems to be no discussion of this in the summary or appendices. Maybe this issue is relatively unique to lichens and other epiphytes where the concepts of exposure and dry deposition are more or less the same thing.

p. 35, lines 13-16, and elsewhere in this section. A bit more detail seems warranted in this initial description of nutrient enrichment beyond just "N additions generally stimulate plant growth and productivity (cumulative growth of all vegetation within a community)". It's also important that N-enhanced growth and productivity stimulation varies substantially among species, favoring faster-growing "N-loving" species at the expense of their slower-growing neighbors, leading to alterations in community composition and diversity.

p. 35, lines 25-26: OK, good! Here's one of the first indications of effects on <u>both acidification and N enrichment</u> occurring <u>at present levels of deposition</u> and <u>in ecosystems across the US</u> and This is important information that should be stated more clearly in the Executive Summary and earlier in the Integrative Synthesis, and perhaps added to the statements of casualty. Two key questions are: Are effects of S and/or N deposition being observed at current levels of S and/or N deposition (or should the phrase be "from current and historical levels of S and/or N deposition"? Are these effects limited to a few isolated mountaintops, or are they relatively widespread? Perhaps brief indications of yes/no effects at current exposures or deposition rates and geographical extent (limited areas or widespread) could be added under the "Current Draft ISA" column in Table 1-1.

p. 35, lines 27-29: The phrase "early signs of recovery" is a reminder that recovery hasn't yet been much discussed (aside from a brief mention on p. 15 discussion of steady state CL). The reader as of this point hasn't seen much discussion about chemical vs. biological recovery, timescales, etc. - such that "early" may not mean much. Maybe a bit more "recovery" discussion earlier on, or at least a pointer to the future section where its discussed in more detail.

p. 37: Table 1.2 is concise and informative!

p. 41, line 5: Would be informative to know if soils with C:N ratios below 25 are relatively common, rare, etc.

p. 41, lines 10-12: Very picky point, but the phrasing of "many types of ecosystems… except heathlands" is somewhat awkward. Depending on your intended meaning, I think it should either be "most types…except heathlands" or "many types… but not heathlands".

p. 41, lines 14-16: It is not clear what these 154%, 134% and 84% "changes in ecosystem N cycle" mean. N additions caused increased nitrification of 154% of what? (the amount of the N addition?)

p. 42, lines 25-28: This statement implies that CLs for N deposition are often developed without assessing whether - or the extent to which - the effects result from acidification or N eutrophication. Is this correct?

p. 43, lines 7 & 8: "N additions increased plant productivity". This broad statement seems inconsistent with previous discussions indicating variability among species in their responses to N addition, altering competitiveness and leading to decreases in biodiversity. For the losers in this N-modified competition, productivity is <u>not</u> increased, in the long term. Maybe you could say something like "N additions differentially increased [or altered] plant productivity…" This also makes me curious: what does it mean when the "productivity" of an ecosystem is "increased"? Could productivity be increased while biodiversity was being decreased, and could this continue indefinitely?

p. 44, lines 21,22: Could this "plants increase exudation as N availability decreases" be explained a bit more? What does this mean, and what are the implications for increasing or decreasing N deposition?

p. 44, line 34 to p. 45, line 2: The wording here could be clarified. Do you mean "locally rare" - such as isolated patches of arctic tundra on NE & NW mountaintops? I don't quite get what you mean by "organisms with specific traits will have either positive or negative responses". Could you add a parenthetical after traits (such as…). Also, I think #s 1 & 2 are observations, actions, phenomena or tendencies - but are not really "mechanisms".

p. 45, lines 30-33 or elsewhere: Are there any implications for long-term C sequestration if plants are (temporarily) storing more C in above-ground biomass? Increased litter depth, fire susceptibility, C storage duration, etc.?

p. 46, lines 20-21: Any implications that shifts to more shade-tolerant tree seedlings could lead to large-scale species shifts as over-story trees eventually die out?

p. 46, line 34: Not clear what you mean by "in <u>all three</u> N addition studies" (US studies, conducted - or reported on since the 2008 ISA or what?)

p. 47, lines 25-26: I believe there are also isolated communities of alpine tundra on Northeastern mountaintops (Mt Mansfield & Camels Hump, VT, Mt Washington, NH, Adirondacks, NY, etc.). See for example: Carlson et al. (2011) Distribution of Tundra in the Adirondack Mountains of New York, U.S.A., *Arctic, Antarctic, and Alpine Research*, 43:3,331-342, DOI: <u>10.1657/1938-4246-43.3.331</u>

p. 48, lines 8 & 9: Again, how can this be true, long-term, for all vascular plant species if N additions reduce biodiversity. Maybe you could say "most vascular species" or "short-term increases".

p. 49, lines 26-28: This "occurs at current rates of N deposition" is an important concept, and should be restated (or its converse) wherever confirming evidence exists. For many/most of the identified causal relationships, you present confirming or enhancing lit published since 2008): I think this concept: "research since 2008 has further documented these effects" could be taken to mean (and also formally used to confirm) that there is "evidence that these effects are occurring at current levels of N deposition".

p. 50, lines 31-33: This is one of the first (& few) references to fire (in arid regions - but not grasslands or forests?). Surely more above ground biomass (& reduced decomposition rates) in most areas increases fire fuel - and potential fire damage, BC emissions, temporarily-stored C releases, etc.

p. 51, lines 21-26: This observation - that CLs increase as N dep increases - explained by an argument that damage has already occurred in higher deposition areas - would seem to provide an important limiter to the CL definition. It suggests that a CL might never be exceeded at current conditions, if the current state of damage is always forgiven. I would like to see more discussion of this topic.

p. 52, Figure 1-7: Do the longer CL bars (ranges) for herbaceous plants, and to a lesser extent for trees, reflect differences among species? If so, maybe you could point this out, as it would be a very clear illustration of the potential loss of biodiversity.
p. 55, lines 26-29: It's not clear what the "15% to 98%" range refers to - or why there is a range. I would have expected a single # like the 53% you report for PA. Does the 15-98% range reflect differences among states in the Northeast?

p. 59, lines 23-25: The observation that denitrification can produce more N_2O "than was previously recognized" is not especially useful information, without context. Say why these new insights are significant, and what about a comment on the influence of atmospheric N_2O as both a greenhouse gas and a stratospheric ozone-depleter?

p. 59, line 26: Many areas have as much or more S deposited as (predominantly dry) SO_2 than as (predominantly wet) SO_4. See for example: ftp://ftp.epa.gov/castnet/tdep/images/s_dwpct/s_dwpct-2014.png

p. 64, lines 25-27: Again, this "larger role than was previously recognized" observation isn't useful information without some discussion of the implications of this revised understanding.

p. 64, lines 32-34: This observation of increasing P deposition is very interesting! However I don't see why increasing P (& decreasing N) would tend to cause a shift from N to P limitation. Au contraire… Also, I think for at least one of the (increasing P) studies cited later on in Appendix 9 (Stoddard et al. (2016)), the observation is for increasing P in lakes and streams, including in relatively remote areas. and distributed nationwide. Increased P deposition - from climate-related increases in windblown dust emissions - is hypothesized (but not really documented) as a possible causal/contributing factor. Other potential causes are also suggested - for example climate-related increases in forest fires or extreme precipitation events, leading to increased P from storm flows, or perhaps resuspension of P in sediments. Hand et al., 2017 (doi.org/10.1002/2016JD026290) have noted seasonal trends of increasing fine dust concentrations in the Southwest (Spring) and Southeast (Summer/Fall). Although trends were less distinct in other regions and seasons. If P deposition were increasing from increased dust emissions, we would also expect increases in crustal cations - Ca, Mg, K, etc. which would have other important implications for acid sensitive ecosystems. In some cases, atmospheric contributions of soil-derived base cations may equal or exceed cation replenishment from weathering (see for example: Derry &

Chadwick, 2007, (DOI: 10.2113/gselements.3.5.333). This is a potentially important topic that requires more discussion.

p. 67, lines 34-36: Not clear what you mean by this: you add NO_3, but blame DOC?

p. 68, lines 23-24: "A portion" is not very informative. Could you be more quantitative? You could also say "in Class I National Parks and Wilderness Areas" and might add "which are afforded special Clean Air Act protections." Otherwise, who cares about Class I?

p. 69, lines 22-23: You need some additional explanation of what "false negatives" are in this context.

p. 70, lines 10-12: There is an important concept here that might be discussed more thoroughly. Despite recent decreases in acidifying (and/or N-enriching) deposition, adverse biological effects persist in most cases. Once ecological damage occurs, it may be sustained even at lower levels of N+S deposition. Could the concepts of chemical and biological recovery be discussed in a bit more detail - beyond the oft-repeated "biological recovery has been limited"?

p. 75, lines 18-19: Does this "deposition exceeds CLs in 1/3 of lakes" apply only to Sierra Nevada Wilderness area lakes? Could you present similar exceedance statistics for other regions where you report CLs?

p. 75 in general: There's a lot of informative discussion about deposition limits needed to attain specific ANC thresholds - which generally range from 0 to 50 μeq/L, a range often taken to be minimally to moderately protective. What about some qualitative discussion of the shifts in various effects if/as ANC is changed from say 100 to 0 (or 0 to 100)? Future changes in S+N deposition will result in shifts in the distribution of ANC within a region, rather than the attainment of any specific ANC threshold in all surface waters - even though a specific ANC threshold may be the basis for setting a NAAQS.

Also, this is the first and only discussion that hints at the time scales that may be associated with (chemical) recovery. As indicated earlier, it might help to see some general "conceptual model" discussion of the separate and combined effects of historical and current S &/or N deposition, along with indications of what chemical and biological responses to decreased (or continuing or increased) S &/or N deposition might look like in the future.

p. 75, line 34: You could drop "Mountains" or the "s" in "Adirondacks".

p. 77, line 17: You could define "anammox" here - the first time we see it.

p. 88, line 10: I assume "state-listed" refers to an endangerment listing in one or more states?

p. 89, lines 9-16: You challenge the applicability of earlier studies by noting the use of much higher N enrichment rates, but then don't really close the deal. Could you add a phrase towards the end like "with effects evident from current levels of atmospheric N deposition".

p. 90, lines 22-28: Is there some reason these causality statements are not in bold type - while most others are?

p. 93, lines 26 & 27: "New research demonstrates that Hg methylation occurs at ambient sulfate concentrations within U.S. water bodies." This is one of many statements that indicates effects occurring at current "ambient" conditions. I think this is demonstrably true for most of the causal effects you identify - but is not always stated. (Again, I'm harping back to the statement that effects from direct SO_2 and NO_Y exposures are not likely at current ambient conditions.) I think continuing effects are more or less implied for almost all other indicated effects. Can you just say so?

p. 97, lines 26-29: This seems like an odd distinction between Al & MeHg vs. other indicators. Why should the metals decrease only to identified toxicity levels while other indicators need to move to preindustrial levels?

p. 98, lines 4-6: I think you could drop the second "at different rates" at the end of the sentence and the "declining" between "decreases in" and "deposition".

p. 101, line 12: I think you mean NOy, not NOx.

p. 104, lines 4-13: In discussing uncertainties of wet deposition estimates, you might also mention the PRISM model - used to enhance the spatial resolution of precipitation volumes in the NADP NTN TDep maps. PRISM uncertainties have been thoroughly evaluated, and the TDep maps will likely be used extensively in the REA and Policy Assessments (and could perhaps even form the basis for secondary NAAQS).

p. 107, lines 7-8: "They found that N deposition was negatively correlated with plant species richness at many locations, but positively correlated at others." I don't think this a very fair summary of the extensive Simkin et al. (2016) analysis. You might at least add ", with most of the positive correlations in areas with low N deposition averaging 3 kg $N \cdot ha^{-1} \cdot y^{-1}$ or less."

Post-meeting comments on the "soil" component of PM and associated ecological effects

The ISA includes an intended focus on ecological effects of particulate matter (PM), specifically including non-S, non-N components of PM (chapter 1.10). However, discussion of the "soil" or "crustal" component of PM is generally understated in the ISA. Soil isn't mentioned in chapter 1.10 or included as a component of PM in the Executive Summary (pp. lxvi, line 11 and lxvii, line 1) or in Chapter 1.3 Emissions and Atmospheric Chemistry (p. 27, lines 16 and 17). But even for fine particles, soil is typically a larger component of $PM_{2.5}$ than elemental carbon at most rural sites, larger than nitrate at many sites and even exceeds organic matter and/or sulfates at some sites and seasons (Hand et al. 2012). Soil is the largest component - roughly 50% - of springtime $PM_{2.5}$ in the Southwestern US (Hand et al. 2017). Airborne soil is primarily a coarse mode particle component, so it's contribution to PM in general (PM_{10} or larger) is <u>much</u> greater than its contribution to $PM_{2.5}$. Limited data on $PM_{10-2.5}$ composition indicate that soil is typically the <u>largest component of coarse particle mass</u> at most sites (Malm et al. 2007). The ISA PM discussion focuses primarily on fine particles ($PM_{2.5}$), but coarse particles ($PM_{10-2.5}$ and larger) are typically more important contributors to dry deposition than fine particles (Lin et al. 1994; Listari et al. 2004).

While most other components of PM (SO_4, NO_3, NH_4, POM, EC) are decreasing over time in most regions, soil has not recently decreased anywhere and so is becoming a proportionately larger contributor to PM in most US regions. Absolute concentrations of fine soil and coarse mass (presumably mostly soil) and wet deposition of soil-related elements are increasing in several regions in the western US, especially during the spring (Brahney et al. 2013; Clow et al. 2016; Hand et al. 2017; Achakulwisut et al. 2017). The onset of the springtime soil peak in the SW US is occurring earlier in the year, where - depositing on the snowpack - it may contribute to earlier snowmelt, adversely affecting hydrological cycles (Hand et al. 2016; Clow et al. 2016). Trends of increasing springtime SW soil concentrations and earlier onset of peak SW soil concentrations are influenced by combinations of natural cyclic and longer-term climatic changes, intercontinental transport and anthropogenic soil surface disturbances (Holmes and Miller 2004; Neff et al. 2005, 2008; Brahney et al. 2013; Hand et al. 2016; Achakulwisut et al. 2017; Webb and Pierre 2018).

The emissions, transport, chemical reactions with and deposition of soil have various important ecological implications (Field et al. 2010, etc.). Soil deposition provides an important source of base cations (BC), enhancing, equaling and often exceeding BC supplied by weathering (Miller et al. 1993; Draaijers et al. 1997; Kennedy et al. 1998; Ballantyne et al. 2011; Watmough et al. 2014). In eastern US areas with acidified soils and/or surface waters, atmospheric BC deposition has partially offset effects of acidifying deposition and may substantially increase the speed of recovery times, if SOx and NOy deposition continues to decrease. For remote oligotrophic surface waters in the western US, soil deposition is increasing primary productivity (Ballantyne et al. 2011). In chapter 7, you suggest that (increasing) atmospheric dust is the most likely cause of recent trends of increasing phosphorus in oligotrophic lakes and streams in all US regions (Stoddard et al. 2016), which has major ecological implications. Soil and sea salt (another occasionally important PM component neglected in the ISA) react readily with nitric acid, leading to formation of coarse mode $CaNO_3$ and $NaNO_3$, as evident in observations of significant concentrations of coarse particle nitrate at many sites (Lefer and Talbot 2001; Lee et al. 2008; Zhang et al. 2008; Allen et al. 2015: Bain et al. 2017). Globally, more than 40% of the total aerosol nitrate is associated with crustal dust (Usher et al. 2003). Thus, soil is an important, often dominant contributor to particulate nitrate deposition. Airborne soil is also an important source and transport host of bioaerosols, which cause or contribute to a variety of environmental effects (Griffin et al. 2001; Brown and Hovmøller 2002; Garrison et al. 2003; Kellogg and Griffin 2006; Haller et al. 2011; Fröhlich-Nowoisky et al. 2016), and which could be discussed in more detail in the ISA.

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