Preliminary Comments from Members of the 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 (First External Review Draft)

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Allen response to CASAC charge Chapters 3, 4, and 5

Review of Chapter 3 direct effects of gas phase on vegetation

Sulfur
There are few new studies of direct gas –phase effects of SO2 on vegetation since 2008 ISA. There is evidence of recovery of gas-phase effects on sensitive terrestrial species since the 1970 CAA, and a few new publications that document this since the 2008 ISA. A study on European silver fir (Elling et al. 2009) indicates 10 μg m^{-3} SO_2 as a critical level (= 4 ppb, p. 3-13), and silver fir growth is recovering because European SO2 levels are low. The U.S. 1982 secondary standard for SO2 is a 3-hour average of 0.50 ppm. However, the current 3-year average high is < ~4 ppb in most states (Fig. 2-15). Vegetation in the U.S. is recovering with this standard in place. The ISA concludes that there is a causal effect of gaseous SO2 on vegetation injury.

Nitrogen
The current EPA standard is 0.053 ppm NO2. Studies showing NO2 impacts on plants (reduced photosynthesis, leaf abscission or damage) have been done at considerably higher concentrations. While NO2 does have impacts, current standards protects against direct impacts (p. 3-8).
However, Vallano et al. (2012) show low levels of NO2 (0.03 ppm) have no direct phytotoxic effects but have a long-term fertilizer effect on invasive grass, so even low levels may cause ecosystem impacts (p. 3-10).

Recent studies have been done for HNO3 exposures. The ISA should explain how the EPA standard of 0.053 ppm NO2 relates to NHO3 atmospheric concentrations.

Low levels of HNO3 contribute to plant N nutrition. HNO3 exposures of 95 to 160 μg/m3 HNO3) cause cuticle damage in Abies and Quercus (Padgett et al 2009). The lichen *Ramalina menziesii* died exposed to 8-10 ppb HNO3 (Riddel et al. 2008), is locally extirpated from LA Basin (*3-12)*.

Reduced nitrogen is not considered in this analysis, only oxidized nitrogen. Working in Scotland, Sheppard et al. (2009) observed growth decline of a heathland shrub (Calluna vulgaris) and death of a lichen (Cladonia) at ammonia critical levels of 8 ug/m3 NH3.

Citation: Sheppard et al. 2009. Long-term cumulative exposure exacerbates the effects of atmospheric ammonia on an ombrotrophic bog: implications for critical loads. Pages 49-58 in Sutton, M. et al., eds. Atmospheric Ammonia: Detecting Emission Changes and Environmental Impacts. Springer Verlag.

The new evidence for direct phytotoxic effects of S suggests vegetation is recovering, and average concentrations of S are low enough in most of the US to protect sensitive tree species. The relationship between the 0.053 ppm NO2 standard and consequent HNO3 concentrations needs to be discussed. HNO3 fumigation studies on higher plants have been done at concentrations below the NO2 standard, but some lichens are sensitive to HNO3 at very low concentrations. Impacts of NH3 fumigation should be reviewed.

Review of chapter 4 biogeochemistry impacts

The biogeochemistry chapter summarizes the effects of elevated N and S on the N and S cycles based on an extensive literature search since the 2008 ISA. Some areas have received considerable attention, such as nitrification (Table 4-6) with many new studies confirming elevated nitrification with N deposition.

New models, and new applications of existing models for determining the fate of elevated N and S, are reviewed, with commentary about their usefulness.

p. 4-68 Recovery of soil acidity with reductions in S deposition has been limited (NAPAP 2011), although one new study is reviewed that shows increased pH over time.

Including the critical loads analysis for acidity of forest soils (Fig. 4-10) is very helpful. Would be nice to know CL for the "unknown" areas of the map. Fig. 4-11 for nitrate leaching fills in some gaps, but the scale is very coarse.

Several metanalyses are reviewed and main results presented—these are very helpful in understanding patterns of responses to elevated N (Fig. 4-7, Sec. 4.3.10)

Very important conclusion:
4-83: There is no single deposition level applicable to all ecosystems in the U.S. that will describe the onset of eutrophication or acidification, ecosystem sensitivity is heterogeneous.

A few corrections/additions/questions:

Fig. 4-1 shows a puzzling area in the California desert indicated as feedlot manure. There is scarce water in this region and little to no agriculture. The validity of some of these mapping efforts needs to be corroborated by comparing maps, models and data. (Similarly, for Fig. 2-24 what is the high N deposition (source and location) in Wyoming? This is not an urban area as indicated in the text.)

Sec. 4.3.1 and p. 4-9 both have discussions about N accumulation in organic matter and in mesic forest soils. However, N also accumulates in arid soils as inorganic NO3 and NH4 (Padgett et al. 1999, cited in Chpt 6).

p. 4-10 "deposition loads of 17 kg N/ha/yr led to the onset of NO3− leaching in the Sierra Nevada and San Bernardino mountains" (citation missing: Fenn et al. 2010).

p. 4-44 l. 18 "In grasslands, Rao et al. (2009) found N deposition may increase production and/or alter litter C:N ratios that increases soil C." Correction: These were not grasslands, but rather desert shrublands and woodlands with invasive annual grasses in interspaces.

Table 4-2: "New N gradient and meta-analysis studies confirm N addition increases nitrification" Does it matter what form of N is deposited? What if NO3 is deposited?

Table 4-12: Give citations in "Effect" column.

The following two studies are absent from the 2008 ISA and the 2017 ISA. They showed effects of elevated N on rates of N mineralization, nitrification, and microbial N in a type-converted exotic annual grassland that had previously been coastal sage scrub:

Sirulnik, A.G., E.B. Allen, T. Meixner, M.E. Fenn, M.F. Allen. 2007. Impacts of anthropogenic N additions on nitrogen mineralization from plant litter in exotic annual grasslands. Soil Biology and Biochemistry 39:24-32.

Sirulnik, A.G., E.B. Allen, T. Meixner, M.E. Fenn, M.F. Allen. 2007. Changes in N cycling and microbial N with elevated N in exotic grasslands of Southern California. Applied Soil Ecology 36:1-9.

Review of Chapter 5 acidification impacts

Chapter 5 reviews the acidification impacts of S and N deposition, and concludes that (together with the 2008 ISA) there is sufficient evidence to infer a causal relationship of this acidification on physiology and productivity, and plant, animal and microbial species richness/composition of terrestrial ecosystems.

Sec. 5.2 and Fig. 5-1 make a strong case that negative effects of acidification from elevated N and S occur because of reduced Ca and reduced based saturation in soil. This causes imbalances in plant Ca nutrition and other cations, elevated Al in some soils, and reduced growth of some tree species. These effects can be reversed by Ca addition as shown by multiple studies (Table 5-1). The Ca fertilization studies are relevant given the importance of Ca deficiency in acidified soils.

Much of the literature reviewed (Table 5-1, 5-2) concerns natural variability in soil pH, Ca concentrations and base saturation impacts on plant health and microbial composition, rather than effects of N or S deposition on acidification. Critical loads of S or N deposition on acidification cannot be deduced from such studies, although they are useful for understanding basic physiological principles of acidification effects on biota. The most useful studies are those that relate N or S deposition to Ca losses or acidification. These are the studies for which N and S deposition values are reported in Table 5-1 and further studies in Table 5-4.

Another difficulty of setting a CL for N or S deposition on acidification is illustrated in the modeling section 5.4. (p. 5-34). The PROFILE model (Phelan et al. 2014) estimated a range of CL from "4 to 10,503 eq/ha/yr using base cation weathering BCw. These values were 3X larger at the same sites using the clay correlation-substrate method and SMB models to estimate BCw rates and critical loads (McNulty et al. 2007)." Modeled values vary widely within and among studies.

The most useful studies for setting a CL are those that did fertilization at ambient levels of N and S deposition or that assessed biotic response across deposition gradients (Table 5-4). Results were highly variable by ecosystem type and taxa assessed. For instance, several dominant tree species are being affected (crown density, crown dieback) by current levels of N and S deposition, while others are less sensitive (Duarte et al. 2012). Lichens are highly sensitive to acidification by N and S deposition as are some taxa of soil microorganisms. A comparative analysis of responses by different species and taxonomic groups against deposition values using data from Table 5-4 might reveal patterns, or at least prioritization of sensitive taxa for setting CL.

To assist with reviewer comparisons among studies, give conversion for eq/ha/yr to kg/ha/yr (e.g., 1 eq N = 14 g N).

The body of studies synthesized for this chapter is extensive, and the conclusion that N and S acidifying deposition can be detrimental is well supported. The discussion is clear and supported by the publications reviewed. A synthesis of the data is needed to assess critical loads.

Dr. Praveen Amar

Preliminary Comments of Praveen K. Amar on February 2017 First Draft of "Integrated Science Assessment for Oxides of Nitrogen, Oxides of Sulfur, and Particulate Matter—Ecological Criteria"

May 13, 2017

Dr. Tom Armitage, Ph.D.
Designated Federal Officer
EPA Science Advisory Board Staff Office
U.S. EPA
1200 Pennsylvania Avenue, NW
Washington, DC 20460

Dear Tom:

This note provides my *very preliminary and brief* comments on EPA's February 2017 first external review draft "Integrated Science Assessment for Oxides of Nitrogen, Oxides of Sulfur and Particulate Matter- -Ecological Criteria." More complete comments will be provided at our CASAC meeting on May 24-25, 2017. As asked by our panel Chair, I also plan to submit formal written comments after the meeting to lead discussants on various chapters (Executive Summary, Chapters 1 and 2) as well as Appendices C and D, for which I am the lead discussant.

For Executive Summary and Chapter 1 (which is really an extended version of Executive Summary and titled "Integrated Synthesis"), CASAC is being asked to comment on the two-part question, presented below mostly in verbatim.

(a) "Please comment on the extent to which the Executive Summary and Chapter 1 meet their objectives. Note that Executive Summary is intended to be a concise *synopsis* of key findings targeted to a broad audience, whereas Chapter 1 is a more detailed *synthesis* of ISA's most policy-relevant findings.
(b) The causality determinations are summarized in Executive Summary and Chapter One. Please comment on the extent to which the causal framework is appropriately applied to evidence for each of the effects categories to form causal determinations.

For Chapter 2, titled "Sources to Deposition," that includes extensive background information on ambient concentrations and atmospheric deposition, the two-part question, asks the following (mostly presented below verbatim).

(a) Please comment on accuracy, clarity, level of detail, and relevance of the information presented regarding sources, chemistry, and measurement and modeling of *ambient concentrations*.
(b) Please comment on accuracy, clarity, level of detail, and relevance of information presented regarding measurements, modeling, *prediction* and trends of *atmospheric deposition* of nitrogen and sulfur including particulates and relevant processes.

The remaining charge questions (and not reproduced below) for Chapters 3 to 14 and Appendices relate to various ecological effects on types of ecosystems in the U.S. and considered in this ISA (terrestrial, wetland, freshwater, and estuarine/near-coastal ecosystems). Ecosystem effects of atmospheric

deposition of N, S, and PM considered in the ISA are grouped into three main categories: (a): Nitrogen-enrichment/Nitrogen-driven eutrophication (caused by NOy and particulate forms of nitrogen); (b) acidification (caused by NOy, SOx, and particulate forms of N and S); and (c) Sulfur enrichment (caused by SOx and particulate forms of Sulfur).

I have not yet reviewed these "effects chapters" in detail and I do not have relevant professional background in ecosystems and ecosystem effects caused by atmospheric deposition of N, S, and particulate matter to provide thoughtful comments. My professional background is in atmospheric processes including atmospheric fate and transport, chemical transport modeling, emission sources and control strategies, acid deposition, and application of scientific results to workable and cost-effective policy options for regional entities and state-level governments.

With this background information, I have some major comments and concerns that relate to Executive Summary and first two chapters. Before I note my concerns below, I note that the description of "causal determinations for relationships between criteria pollutants and ecological effects" (Table ES-1 and reproduced as Table 1-1 in Chapter 1) is well written and makes a good case that the causal framework is appropriately applied to the evidence for each of the effects categories to form causal determinations.

My first concern is on how the extensive and credible scientific information included in this ISA (including five case studies in Appendix C) on critical loads (including numerical values for CL of nitrogen, sulfur or both in kg/hectare/year) for various ecosystems effects (soils, forests/trees, lakes and streams) would be *actually applied* in a policy-relevant manner. It very well might be that this question will be answered more fully in time when this CASAC panel reviews the REA (Risk Exposure Assessment) document as well as the PAD (Policy Assessment Document) prepared by EPA staff. Nevertheless, CASAC panel should start the discussion on this important issue at our May 24-25, 2017, meeting.

Things to consider include how to make a "policy jump" from federal NAAQS ("ambient concentrations" of NO2 and SO2 and PM2.5 and PM10) to determination of critical loads for what actually "falls" on the soils, lakes, various tree species in the forests, wetlands, and estuaries. Also, CASAC panel should take into account how the previous CASAC panel on this effort (2008-2011) tried to address this CL issue for protection of aquatic ecosystems through dose-response of fish to ANC (acid neutralizing capacity) by introducing the concept of AAI (atmospheric acidity index). However, the generalized issue of "uncertainty" was raised about such a standard providing the necessary protection of lakes and streams with the level of confidence needed before such a standard could be promulgated. Same types of "uncertainty" issues need to be addressed in this ISA in the context of critical loads for various ecosystems in addition to freshwater systems considered in the previous ISA (2008).

Second general and important concern is on how the information on reactive reduced nitrogen compounds (ammonia gas, ammonium ion, ammonium nitrate, etc.) and their role in acidification and nitrogen-enrichment of ecosystems would be addressed in the determination of critical loads. It would also be important to get a better and more accurate national and regional- and state-level estimates of current ammonia emissions, and, equally if not more important, *projections of future ammonia emissions*. Unlike SOx and NOx emissions which are currently on a downward trajectory because of various federal and states regulations for stationary and mobile sources, ammonia emissions are projected to *increase* because of emissions from CAFO (concentrated animal feeding operations) and

other agricultural sources (see Ex. Summary, page lxii). Also, though it seems adequate, CASAC may wish to evaluate if the ISA accurately reflects the current scientific understanding of atmospheric chemistry and deposition of ammonia (ammonia, nitric acid, ammonium nitrate, the relative levels of nitric acid and ammonium nitrate as a function of ambient temperature and other atmospheric variables; bidirectional flux of ammonia at ground level, etc.), especially if future policies and standards are designed to control or at least "take into account" the ammonia emissions. For example, more NOx and SOx emissions reductions might be necessary to achieve a certain target load if ammonia emissions are *not* controlled, but simply are "taken into account".

Finally, third concern is on how to address the general topic of uncertainty. I do not have detailed comments to make at this time on the subject of uncertainty in estimating various levels of N and S (and, PM) deposition, and on how to address uncertainty in estimating various biogeochemical and ecological effects on various ecosystems. The subject of how to address uncertainty is critical in how the findings of this ISA would be utilized in future policy making and setting of secondary standards for protection of ecosystems in the U.S. at national and regional levels. The executive summary (page lxii) notes that "numerous factors cause uncertainty in estimating N and S deposition." It then goes on to outline various sources of uncertainty in emissions, atmospheric chemistry, and deposition of N and S and PM. Of course, there are additional uncertainties associated with inputs and outputs of various modeling systems of atmosphere (CMAQ) and ecosystem responses to acidification and eutrophication. There are also uncertainties associated with measurements of ambient concentrations, atmospheric wet and dry deposition, and ecological inputs and responses.

There is a whole field of scientific literature on quantitative and qualitative analysis of uncertainty in public policy and decision-making, exposure assessment, risk assessment and risk management as it relates to ecosystems. I am not sure if the current ISA includes sufficient data and science to addresses this important issue of addressing uncertainty in the future documents (REA and PAD).

Sincerely,

Praveen Amar, Ph.D., P.E.
Independent Consultant
Environment, Energy, and Climate Strategies

Dr. James Boyd

Boyd initial comments (focused on Charge Question 14: Ecosystem Services)

Charge Question

Chapter 14 is a summary of recent advances in ecosystem services frameworks, studies that evaluate the effects of anthropogenic nitrogen on ecosystem services and several "profiles" of threatened and endangered species for which nitrogen is listed as a stressor. Please comment on the accuracy, clarity, level of detail, and relevance of this summary of ecosystem services frameworks and the effects of nitrogen inputs on ecosystem services.

- Broad comment

The Ecosystem Services section does not clearly state what has really been concluded. Personally, I think that what can be concluded is something like (1) there is evidence that emissions/deposition may have a range of impacts to ecosystem services and their social value; (2) there are some economic studies that demonstrate such effects in broad terms; but (3) it remains methodologically difficult to derive economic costs and benefits associated with specific regulatory decisions/standards.

- Broad comments

The section leads with and devotes significant space to discussion of ecosystem services *frameworks*. Frameworks are useful and relevant (because they can help the agency identify causal pathways between specific ecosystem changes and potential social beneficiaries). But frameworks alone do not address the core issue, alluded to in the following kinds of terms:

> "They conclude that quantitative assessment is problematic due to a lack of units of measure to gauge changes in the quality and quantity of ecosystem services and a lack of dose-response relationships to indicate how quality and quantity may change as a function of changes in pollution exposures." p. 14-3
>
> "NAPAP (2011) concluded that the greatest challenge in developing specific data on the economic benefits of emission reductions lies with the availability of comprehensive scientific evidence that defines the extent and magnitude of the adverse effects that can be directly attributed to acid deposition from among multiple ecosystem stressors." p. 14-4
>
> "Although these assessments have varied considerably in their approaches, all have used simplified approaches that intentionally omitted much of the mechanistic and spatial complexity in how deposition affects ecosystem services." 14-8

By itself, having a consistent *framework* will not address these *causal gaps*. Causal gaps are the real issue, though frameworks can help identify those gaps.

- Comment

The section references studies that calculate human health-related benefits. Should those be more clearly identified as outside the scope of secondary standards evaluation?

- Comment

The report states that "Ecosystem services are often affected as a result of N or S deposition." (e.g. p. 1-85, 14-18) The word "often" is vague. Could/should language be changed to something like: "Since 2008 several studies have identified a range of ways in which N-S deposition affects socially valuable ecosystem services."

- Comment

A set of recent papers in Ecosphere relates critical loads to the ecological science of ecosystem production function relationships within a classification framework. EPA staff are included as co-authors. Is there a reason those papers are not cited?

> O'Dea et al, "Impacts to ecosystem services from aquatic acidification: using FEGS-CS to understand the impacts of air pollution" 2017
>
> Bell et al., "A framework to quantify the strength of ecological links between an environmental stressor and final ecosystem services" 2017
>
> Clark et al, "Nitrogen-induced terrestrial eutrophication: Cascading effects and impacts on ecosystem services," forthcoming.

- Small comment

Sentence is inaccurate and can probably just be deleted.

> "In economics literature, services are typically viewed as "flows" from the provider to the consumer that are measured over time." (14-2)

Dr. Douglas Burns

Charge Question #8. Review of Chapter 8 – Biological indicators of acidifying deposition and effects on biodiversity of freshwater biota. Lead discussants are: Drs. Douglas Burns, Robert Goldstein, William McDowell, and Stephen Schwartz.

Chapter 8 summarizes biological indicators of acidifying deposition and effects on biodiversity of freshwater biota. These effects have been well established for several decades. Please comment on the accuracy, clarity, level of detail, and relevance of information presented on the biological indicators of acidifying deposition and the effects of deposition on biodiversity in freshwater biota.

In general, I find the information presented in this chapter on biological indicators of acidifying deposition and the effects of acidification on biodiversity to be very good. Much of the science of acidification and its aquatic biological effects was well investigated in the 1980s and 1990s and was reflected in the previous EPA 2008 ISA. I would characterize updated research since the 2008 ISA as generally providing some incremental improvements in knowledge of biological effects and indicators, and the current assessment clearly and accurately reflects this new knowledge. An important point is that a causal relationship between acidifying deposition and effects on biota is still well supported as it was in the 2008 ISA.

Below are some comments and suggestions for this chapter. Most of these comments are generally minor to moderate in scope. I will begin with those that are moderate in scope and finish with the more minor comments.

<u>Moderate Comments</u>

- The chapter would benefit from a clearer and more definitive definition of biological recovery as this term is used in several places in the manuscript without the reader always knowing what exactly is meant. I would propose that the terms full or complete biological recovery and recovery trajectory be considered for use. Full or complete biological recovery would be a return to the same number of species of a given taxa as were believed to have been present prior to acidification. A recovery trajectory is one in which the number of species is increasing towards those believed to have been present prior to acidification, but has not yet reached this value. There are nuances to the recovery story such as density, which may not been well known prior to acidification. Also, the species numbers might return to pre-acidification values, but the mix of species may be different.
- The most important update from the 2008 ISA in terms of advancement in scientific understanding is probably the work on deposition effects on Atlantic salmon. The chapter does a good job on pages 8-16 to 8-20 of summarizing this work which suggests that acid deposition may be playing a role in limiting the recovery of this species, though this may not be stated quite as clearly and forcefully as it might have been.
- Section 8.4.6 on mitigation misses the fundamental point that there are strong differences in the intensity and duration of the response dependent on whether lime is introduced directly into the body of water or is spread onto watershed soils. This point and some supporting literature (1996 special issue of Biogeochemistry, edited by C.T. Driscoll) should be cited in this section. If

possible, I would suggest citing some of the conclusions from the recent liming review paper by Lawrence et al., 2016, Science of the Total Environment, 562: 35-46.

- On page 8-25, Lines 22-29, Hesthagen et al., 2008 make the point that the ANC value that indicates pertinent values of pH, inorganic Al, and Ca may be different and greater during chemical recovery than this ANC value was during acidification. In other words the value of ANC as a biological indicator may have shifted upwards (following a hysteretic path) during recovery from the indicator value during acidification. This is particularly relevant in evaluating ANC as a potential indicator of biological recovery. This point could be emphasized more clearly at the beginning of section 8.4 as it is relevant to the discussion of recovery.
- In section 8.5.1, it would be helpful to make a clearer distinction between controls in glaciated vs. unglaciated regions. For example, till thickness has been shown to be a key control on the pH and ANC of Adirondack lakes (work of Driscoll, Newton, and others from the 1980s). Thickness seems to act as a control on subsurface residence time and the role of deeper groundwater discharge, regardless of till content. Also, in unglaciated regions, sulfate adsorption can be substantial due to the age and extensive weathering of soil profiles, whereas glaciated soils show much less sulfate adsorption. These factors have affected acidification and will likely affect the recovery trajectory.

<u>Minor Comments</u>

- Page 8-33, Line 33 – Although much of episodic acidification can be driven by simple dilution, an important point is that even dilution of base cation concentrations may be affected in part by historic base cation depletion of watershed soils, which limits the rapid release of Ca and Mg to rapid runoff during storms.
- Page 8-5, Line 24 – I'm not sure that we can say with confidence that acidification effects on primary productivity are negligible. I would say that there have been few studies on this topic and the results have been difficult to interpret. For example, net ecosystem production is an important quantity in this regard, and there is evidence that acidification can affect the rate of ecosystem respiration (through suppression of mineralization rates of organic matter), which would decrease net production. I would be more comfortable saying the effects on production are uncertain.
- Page 8-9, Line 9 – The acid BAP was actually first introduced in Burns et al., 2008b
- Page 8-9, Line 22 – should use mayfly here as above rather than Ephemeroptera
- Page 8-56, Line 22 – No Catskill reference here. Could use: Baldigo, B.P. and Lawrence, G.B., 2001. Effects of stream acidification and habitat on fish populations of a North American river. *Aquatic Sciences-Research Across Boundaries*, *63*(2), pp.196-222.
- Page 8-57, Line 6 – It can help to state that the target load can be less than, equal to, or greater than the critical load.
- Section 8.5.4 to 8.5.6 – It would be helpful to use consistent critical loads units throughout these sections to facilitate easy comparisons.
- Page 8-66, Line 2 – Adirondack is misspelled
- Section 8.5.6.2 – The important point regarding an inability for full recovery is that in acid-sensitive regions, some of the water bodies likely never had an ANC value that may be targeted

for recovery in modeling applications. For example, a stream that has a pre-industrial ANC that is estimated at 25 ueq/L could not be expected to recover to an ANC value of 50 or greater. This points out the limitation of defining a fixed value for recovery and targeting recovery to that value in model simulations. This approach may overstate critical load exceedance.

- Page 8-67, Line 32 – The paragraph that begins here looks a little anomalous. It seems an attempt to discuss the role of TOC on acidification and recovery. There is a large body of literature on this topic, much of it published since the 2008 ISA, and this paragraph does not really adequately discuss all of the pertinent issues related to TOC/DOC. For example, Al binding and the likely role in minimizing Al toxicity. I assume that the role of TOC/DOC is well covered in Chapter 7. I think either this topic needs to be discussed in greater detail here or this paragraph could otherwise be eliminated.
- Page 8-75, Line 28 – An important point regarding Baron et al., 2011b is that these are critical loads for episodic acidification and most other critical loads investigations are focused on chronic acidification.

Ms. Lauraine Chestnut

Lauraine Chestnut
Preliminary Comments for NOx/SOx Secondary NAAQS Review of ISA
May 17, 2017

1. Executive Summary and Chapter 1

The Executive Summary and Chapter 1 provide overviews of the ISA. The Executive Summary is intended to be a concise synopsis of key findings targeted to a broad audience, whereas Chapter 1 is a more detailed synthesis of the ISA's most policy-relevant findings.

a. *Please comment on the extent to which the Executive Summary and Chapter 1 meet their objectives.*

b. *The causality determinations are summarized in the Executive Summary and Chapter 1, please comment on the extent to which the causal framework is appropriately applied to evidence for each of the effect categories in chapters 3-12 to form causal determinations.*

A great deal of information is presented in the ISA and summarized in these two sections regarding how N and S deposition changes soil and water chemistry and how these changes affect terrestrial and aquatic ecosystems. There is emphasis on what is established as causal relationships and on what is known about critical loads, levels at which specified changes are observed to begin to occur. What is missing in these summary/overview sections is a full picture of the geographic extent and degree to which these changes have happened or are continuing to happen at current conditions.

In addition, a more complete story is needed about why the changes that have occurred, or are still occurring, matter. This does not need to be tied to direct human uses, but connections need to be made between the changes and things like sustainability of the ecosystem or viable habitat for species that are native to the area. This may be addressed more fully in the risk and exposure assessment, but this kind of synthesis of the scientific evidence would make the ISA a more useful assessment for policy.

Sections 1.3.1 and 1.3.2 present important basic concepts about what an ecosystem is and about why biodiversity is important. It would be helpful for policy considerations if the results in the literature reported in subsequent sections were more fully connected back to these concepts. For example, is any change in species composition or biodiversity necessarily harmful or detrimental? Certainly, a complete loss of a certain species that had been previously present seems like a significant change, but is a 10% or 20% loss, for example, a detrimental change? How do the changes associated with N and S deposition affect the functioning or sustainability of the ecosystem?

Pages 1-21 and 1-22 present important concepts regarding critical loads. It seems clear that exceeding a critical load does not necessarily mean that an impactful change has occurred. It depends on what the indicator of change is for which the critical load is defined. Therefore, the usefulness of critical load findings from the literature for policy assessments depends on the indicator for which the critical load has been estimated. The case needs to be made that the indicator represents a change that is significant. Just defining a critical load is not enough.

Page 1-73: This is a good summary of where harmful effects of N-related eutrophication are present at current conditions in the US.

Page 1-84: The apparently long time frame for recovery needs to be addressed in the risk and exposure assessment. Would further reductions in deposition speed the recovery at all?

14. Ecosystem Services

Chapter 14 is a summary of recent advances in ecosystem services frameworks, studies that evaluate the effects of anthropogenic nitrogen on ecosystem services and several "profiles" of threatened and endangered species for which nitrogen is listed as a stressor. Please comment on the accuracy, clarity, level of detail, and relevance of this summary of ecosystem services frameworks and the effects of nitrogen inputs on ecosystem services.

This chapter is fine as far as it goes, but more consideration about how the understanding of ecosystem services could inform the overall science assessment would be useful. Perhaps this comes in the risk and exposure assessment, but here this chapter seems just stuck into the document, not connected to the other parts.

It is clear that the study of ecosystem services is not to a point where a comprehensive quantitation assessment of the effects of N and S on ecosystem services is feasible. However, the concepts of ecosystem services and how they are impaired by effects of N and S deposition could help tell the story of why these effects matter.

It is very clear from studies cited here, and from previous S-related ecosystem services literature (e.g. regarding acidification in the Adirondacks), that the value/significance/importance of ecosystem services is not primarily the result of effects on direct human uses. Protection and sustainability of the natural environment is highly valued and when this has been quantified it greatly exceeds the value of direct human use.

The list of species for which N is a stressor is lacking context. How widespread and how significant are the stresses for each of these species under current conditions?

15. Appendices

Case studies at five locations in the U.S. (Southern California, Northeastern U.S., Rocky Mountain National Park, Southeastern Appalachia, Tampa Bay) are included in Appendix C to support potential place-based risk and exposure assessment options to be conducted by the Office of Air Quality Planning and Standards.

Please comment on the adequacy of the information for the case studies and identify additional considerations, if any, relevant to evaluation of effects in these locations.

The case studies provide a nice opportunity to get more specific about N and S effects, which vary so much across different types of ecosystems. The inclusion of Class I areas is also useful because these

areas have been designated as important to protect and preserve in their natural condition. However, this does not mean that it is not important to protect the health of ecosystems in other locations.

Listing the threatened and endangered species that are present in each of the case study areas is also useful, but the reviews do not make an explicit connection between the changes caused by N and S deposition and health or habitat for these species.

It seems the ISA is focusing on research findings since the last ISA. However, it is not clear why the Adirondacks area was dropped as a case study. This area has been significantly impacted by N and S deposition in the past and has been extensively studied. It would seem to be an appropriate choice as an ongoing case study area for this review. Changing to the northeast adds a Class I area with Acadia National Park, but it appears that the effects of N and S deposition in this area are not as significant as they have been in the Adirondacks.

It makes sense to include PM in this ISA to the extent that sources of N and S deposition are also sources of PM, such that these co-occur in the environment and may be reduced by the same potential control measures. The evidence summarized in Appendix D describes the mechanisms by which PM can affect vegetation and biota, but it is not clear to what extent any such effects are occurring in the US under current conditions.

Dr. Charles Driscoll

page	line	comment
lxvi	Table ES-1	typo second line in first category "alteration"
lxxiii	28	here and throughout the document inorganic Al is probably not the best term, because soil/sediment Al is largely in an inorganic form and is non-toxic. Probably should use dissolved inorganic Al
1-47	15	Need to clarify the uniits of Bc:Al. eq/mol?
1-56	5	Should be PnET-BGC
1-57	10	Statement is not correct. Some models, such as PnET-BGC predict all acid base species, including pH, species of Al, in addition to ANC
1-57	11	Should provide some additional clarification to statement. ANC is a human chemical contruct to enhance understanding of acid-base processes and does not have any direct relevance to biological impacts.
1-62	14	Seems to be a mistake. "maintain an ANC of 74 eq/ha-yr…" 74 eq/ha-yr is a loading not an ANC value. Clarify.
1-73	3	Susceptible to eutrophication or experiencing eutrophication?
1-73	8	Water quality has diminished or deteriorated, rather than decreased
1-74		Would it make more sense to place the order of the wetland section before the marine section to follow the flow from the atmoshere to uplands to surfacewaters to wetlands to marine ecosystems?
2-1		Give an example of a species represented in NOz
2-1	29	Is the first phrase in the sentence begining "Excess NH3" linked to the phrase that follows? By nitrification is the author referring to oxidation of NH3? Nitrification of or nitrification in? A strangely phrased sentence.
2-4	Table 2-1	These are U.S. emissions correct? This should be clarified in the table title.
2-16	13	transported from , correct?
2-66		Would it be possible to show a map of % dry nitrogen deposition for the CONUS similar to 2-26. Even better if this could be shown for both oxidized and reduced nitrogen.
2-66	26	parks (lower case)
2-77	6	Most surface waters are net sources of atmospheric carbon dioxide, so I don't think this statement is correct or relevant
2-82	Figure 2-34	In Figure 2-34 it would be helpful to clarify what the data points for transference ratios represent
2-18		Background values for ambient air concentrations are not that helpful for an effects document. It would be better to provide specific concentrations in precipitation and total deposition of backgound values to place current deposition values in perspective. The % contribution as background deposition is also not particularly useful and

		this % changes with the period of observation. Absolute background deposition values would be most helpfu.
4-19	Sulfate	I am concerned that this section does not address the pH-dependent adsorption of sulfate. In sites where the pH is changing this can influence the retention/ release of soil sulfate. See Fakhraei, H., C. T. Driscoll, J. R. Renfro, M. A. Kulp, T. Blett, E. F. Brewer, and J. Schwartz. 2016. Critical Loads and Exceedances for Nitrogen and Sulfur Atmospheric Deposition in Great Smoky Mountains National Park, USA. Ecosphere, 7(10). doi:e01466. 10.1002/ecs2.1466.
4-4	17	The most important process of soil acidification is the leaching of cations with drainage waters. The quantity of precipitation and runoff is an important determinant. van Breemen, N., J. Mulder, and C. T. Driscoll. 1983. Acidification and alkalization of soils. Plant and Soil 75:283-308.
4-22	Base cations	I am not sure what science should be included in a section on base cations. There is no mention of the calcium silicate experiment conducted at Hubbard Brook. There has been quite a bit of relevant literature from this experiment. Shao, S., C. T. Driscoll, C. E. Johnson, T. J. Fahey, J. J. Battles, and J. D. Blum. 2016. Long-term responses in soil solution and stream-water chemistry at Hubbard Brook after experimental addition of wollastonite. Environmental Chemistry, 13(3), 528-540. doi:10.1071/EN15113.; Battles, J. J., T. J. Fahey, C. T. Driscoll, J. D. Blum, and C. E. Johnson. 2014. Restoring soil calcium reverses forest decline. Environmental Science & Technology Letters 1:15-19.; Johnson, C. E., C. T. Driscoll, J. D. Blum, T. J. Fahey, and J. J. Battles. 2014. Soil chemical dynamics after calcium silicate addition to a northern hardwood forest. Soil Science Society of America Journal 78:1458-1468.; Green, M. B., A. S. Bailey, S. W. Bailey, J. J. Battles, J. L. Campbell, C. T. Driscoll, C. Eagar, L. Lepine, G. E. Likens, S. V. Ollinger, and P. G. Schaberg. 2013. Decreased water flowing from a forest amended with calcium silicate. Proceedings of the National Academy of Sciences 110:5999-6003.; Nezat, C. A., J. D. Blum, and C. T. Driscoll. 2010. Patterns of Ca/Sr and 87Sr/86Sr variation before and after a whole watershed CaSiO3 addition at the Hubbard Brook Experimental Forest, USA. Geochimica et Cosmochimica Acta 74:3129-3142 DOI: 3110.1016/j.gca.2010.3103.3013.
4-26	Aluminum	I don't know how the ISA can say there have been no papers on aluminum leaching. Above I provide studies from the wollastonite addition experiment. Also see Fakhraei, H., and C. T. Driscoll. 2015. Proton and aluminum binding properties of organic acids in surface waters of the Northeastern, USA. Environmental Science & Technology 49:2939-2947.; Fuss, C. B., C. T. Driscoll, and J. L. Campbell. 2015. Recovery from chronic and snowmelt acidification: Long-term trends in

		stream and soil water chemistry at the Hubbard Brook Experimental Forest, New Hampshire, USA. Journal of Geophysical Research Biogeosciences 120:2360-2374.; Driscoll, C. T., K. M. Driscoll, H. Fakhraei, and K. Civerolo. 2016. Long-term temporal trends and spatial patterns in the acid-base chemistry of lakes in the Adirondack region of New York in response to decreases in acidic deposition. Atmospheric Environment, 146, 5-14. doi:10.1016/j.atmosenv.2016.08.034.
4-48	1-2	Do we know that decreases in pH supress DOC production? It is difficult to separate this effect from abiotic effects. There should be some text added on changes in partitioning of DOC with changes in soil pH which could drive changes in DOC mobility.
"4-48	Dissolved organic carbon	It would be good to add in some text on the acid base chamistry of organic acids and potential changes in dissolved organic matter quality. Fakhraei, H., and C. T. Driscoll. 2015. Proton and aluminum binding properties of organic acids in surface waters of the Northeastern, USA. Environmental Science & Technology 49:2939-2947.
4-55	Soil monitoring databases	I mentioned the long-term wollastonite addition study at Hubbard Brook above. There is also a long-term soil solution data base; see Fuss, C. B., C. T. Driscoll, and J. L. Campbell. 2015. Recovery from chronic and snowmelt acidification: Long-term trends in stream and soil water chemistry at the Hubbard Brook Experimental Forest, New Hampshire, USA. Journal of Geophysical Research Biogeosciences 120:2360-2374.
4-56	4	I do not understand the sentence on the Lehman et al. 2008 study. This should be rewritten so it is clearer.
4-64	Models	I don't understand how you could say there have been no applications on PnET-BGC since 2008. See the following references. Fakhraei, H., C. T. Driscoll, J. R. Renfro, M. A. Kulp, T. Blett, E. F. Brewer, and J. Schwartz. 2016. Critical Loads and Exceedances for Nitrogen and Sulfur Atmospheric Deposition in Great Smoky Mountains National Park, USA. Ecosphere, 7(10). doi:e01466. 10.1002/ecs2.1466.; Pourmokhtarian, A., C. T. Driscoll, J. Campbell, K. Hayhoe, A. M. K.Stoner, M. B. Adams, D. Burns, I. Fernandez, M. J. Mitchell and J. B. Shanley. 2016. Modeled Ecohydrological responses to climate change at seven small watersheds in the northeastern U.S. Global Change Biology 23: 840-856. doi:10.1111/gcb.13444.; Zhou, Q., C. T. Driscoll, and T. J. Sullivan. 2015. Responses of 20 lake-watersheds in the Adirondack region of New York to historical and potential future acidic deposition. Science of the Total Environment 511:186-194.; Zhou, Q., C. T. Driscoll, T. J. Sullivan, and A. Pourmokhtarian. 2015. Factors influencing critical loads and target loads for the acidification of lake-watersheds in the Adirondack region of New York. Biogeochemistry 124:353-369.; Zhou, Q., C. T. Driscoll, S. E. Moore, M. A. Kulp, J. R. Renfro, J. S. Schwartz, M. J. Cai, and J. A. Lynch. 2015. Developing critical loads of nitrate and sulfate in the Great Smoky

		Mountains National Park, United States. Water Air and Soil Pollution 226:1-16.; Fakhraei, H., C. T. Driscoll, P. Selvendiran, J. V. DePinto, J. Bloomfield, S. Quinn, and C. Rowell. 2014. Development of a total maximum daily load (TMDL) for acid-impaired lakes in the Adirondack region of New York. Atmospheric Environment 95:277-287.; Bytnerowicz, A., M. Fenn, S. McNulty, F. Yuan, A. Pourmokhtarian, C. T. Driscoll, and T. Meixner. 2013. Interactive effects of air pollution and climate change on forest ecosystems in the United States: Current understanding and future scenarios. Pages 333-369 in R. Matyssek, N. Clarke, P. Cudlin, T. N. Mikkelsen, J.-P. Tuovinen, G. Wieser, and E. Paoletti, editors. Developments in Environmental Science. Climate Change, Air Pollution and Global Challenges. Elsevier Physical Sciences Series.; Pourmokhtarian, A., C. T. Driscoll, J. L. Campbell, and K. Hayhoe. 2012. Modeling potential hydrochemical responses to climate change and rising CO2 at the Hubbard Brook Experimental Forest using a dynamic biogeochemical model (PnET-BGC). Water Resources Research 48, W07514:13pp.; Wu, W., and C. T. Driscoll. 2010. Impact of climate change on three-dimensional dynamic critical load functions. Environmental Science & Technology 44:720-726.; Fenn, M. E., C. T. Driscoll, Q. Zhou, L. E. Rao, T. Meixner, E. B. Allen, F. Yuan, and T. J. Sullivan. 2015. Use of combined biogeochemical model approaches and empirical data to assess critical loads of nitrogen. Chapter 10. In W. De Vries, J.-P. Hettelingh, and M. Posch, editors. Critical Loads and Dynamic Risk Assessments: Nitrogen, Acidity and Metals for Terrestrial and Aquatic Ecosystems. Springer, Dordrecht, The Netherlands.
4-65	16	The sentence starting with Zaehle (2013) is not clear and should be rewritten.
5-10	Table 5-2	In Battles et al. 2014 sugar maple response to changes in Ca/Ali in soil solutions are reported.
5-13	1	Is there any evidence for episodic acidification in soil?
5-26	17	Clarify the species abundance increase with increasing pH.
5-30	11	Are the units here inappropriate for pH? Should be unitless, correct?
5-32	17	driven?
5-32	18	Is this statement that the highest atmospheric fate factor occur on the west coasts globally relevant or only for the temperate zone in the Northern Hemisphere?
5-34	Acidification models	There must be a bias against PnET-BGC. As mentioned in chapter 4 PnET-BGC has been used to evaluate acid rain effects on soil acidification.
5-43	Impacts of ambient deposition	It would be helpful to indicate the year/ period over which these assessments were made, as deposition has changed markedly in recent years.

Another paper		There is a new paper on the recent response of red spruce to decreases in acid deposition that may be of interest. Wason, J. W., Dovciak, M., Beier, C. M. and Battles, J. J. (2017), Tree growth is more sensitive than species distributions to recent changes in climate and acidic deposition in the northeastern United States. J Appl Ecol. doi:10.1111/1365-2664.12899
12-4	9	Sulfate in not particularly mobile in soils will a high clay content and high in amorphous iron and aluminum oxides.
12-5	28	Soil content of amorphous iron and aluminum oxides is also an important controller of sulfate adsorption
12-6	12	Flocculant layer?
12-6	28	This statement is not correct. The quantity of sulfate retained in lake sediments is highly variable and depends on the mean depth and hydraulic residence time of the lake. Kelly, C. A., J. W. M. Rudd, R. H. Hesslein, D. W. Schindler, P. J. Dillon, C. T. Driscoll, S. A. Gherini, and R. E. Hecky. 1987. Prediction of biological acid neutralization in acid-sensitive lakes. Biogeochemistry 3:129-140.
12-23	Zones of high methylmercury fractions across the landscape	Yu et al. found no variation in concentrations of total mercury, methylmercury and %MeHg with pH in Adirondack lakes. Yu, X., C. T. Driscoll, M. Montesdeoca, D. Evers, M. Duron, K. Williams, N. Schoch, and N. C. Kamman. 2011. Spatial patterns of mercury in biota of Adirondack, New York lakes. Ecotoxicology 20 1543-1554 Selvendiran et al. 2008 found methylmercury and %MeHg greatly increase in waters draining wetlands compared with drainage from uplakes in the Adirondack landscape. Selvendiran, P., C. T. Driscoll, J. T. Bushey, and M. R. Montesdeoca. 2008. Wetland influence on mercury fate and transport in a temperate forested watershed. Environmental Pollution doi:10.1016/j.envpol.2007.12.005: 154:46-55.
12-24	24	base of the foodchain
12-27	26,33	Onondaga Lake
12-27	Seasonality and temperature	Selvendiran et al (2008) and Gerson and Driscoll (2016) observed marked increases in concentrations of methylmercury and %MeHg in Adirodack streams during the summer low flow period. Selvendiran, P., C. T. Driscoll, J. T. Bushey, and M. R. Montesdeoca. 2008. Wetland influence on mercury fate and transport in a temperate forested watershed. Environmental Pollution doi:10.1016/j.envpol.2007.12.005: 154:46-55. Gerson, J. R., C. T. Driscoll. 2016. Is mercury in remote forested watershed of the Adirondack Mountains responding to recent decreases in emissions? Environmental Science and Technology, 50, 10943-10950. doi:10.1021/acs.est.6b02127

12-28	Total mercury concentrations	Gerson and Driscoll (2016) recently examined factors driving long-term decreases in total mercury, methyl mercury, but not %MeHg in an Adriondack stream and lake. They found that these changes were consistent with decreases in atmospheric mercury deposition, but methylmercury concentrations and %MeHg were not affected by long term decreases in sulfate or changes in hydrology. Gerson, J. R., C. T. Driscoll. 2016. Is mercury in remote forested watershed of the Adirondack Mountains responding to recent decreases in emissions? Environmental Science and Technology, 50, 10943-10950. doi:10.1021/acs.est.6b02127
"12-29	pH	Yu et al. found no variation in concentrations of total mercury, methylmercury and %MeHg with pH in Adirondack lakes, but bioaccumulation of MeHg at the base and through the food chan was enhanced with decreases in lake pH. Yu, X., C. T. Driscoll, M. Montesdeoca, D. Evers, M. Duron, K. Williams, N. Schoch, and N. C. Kamman. 2011. Spatial patterns of mercury
12-31	5	Dittman et al. 2010 also found increases in MeHg and total Hg with increases in the hydrophobic and high molecular weight fraction of dissolved organic carbon. Dittman, J. A., J. B. Shanley, C. T. Driscoll, G. R. Aiken, A. T. Chalmers, J. E. Towse, and P. Selvendiran. 2010. Mercury dynamics in relation to dissolved organic carbon concentration and quality during high flow events in three northeastern U.S. streams. Water Resources Research 46, W07522, doi:10.1029/2009WR008351.
12-32	14	Iron oxides can form <u>surface</u> complexes with inorganic mercury.
12-32	Nitrate	Matthews et al. (2013) document the application of calcium nitrate as a whole -lake sediment treatment to limit the production of methyl mercury in a mercury contaminated lake. Matthews, D. A., D. B. Babcock, J. G. Nolan, A. R. Prestigiacomo, S. W. Effler, C. T. Driscoll, S. Todorova, and K. M. Kuhr. 2013. Whole-lake nitrate addition for control of methylmercury in mercury-contaminated Onondaga Lake, NY. Environmental Research 125:52-60.
12-44	Sulfur oxide effects on methylmercury	This recent article was mentioned above but it is also relevant here. Gerson and Driscoll (2016) recently examined factors driving long-term decreases in total mercury, methyl mercury, but not %MeHg in an Adriondack stream and lake. They found that these changes were consistent with decreases in atmospheric mercury deposition, but methylmercury concentrations and %MeHg were not affected by long term decreases in sulfate or changes in hydrology. Gerson, J. R., C. T. Driscoll. 2016. Is mercury in remote forested watershed of the Adirondack Mountains responding to recent decreases in emissions? Environmental Science and Technology, 50, 10943-10950. doi:10.1021/acs.est.6b02127

12-76	Sensitive ecosystems	Evers et al. (2011) was not peer reviewed but includes relevant information on ecosystem mercury sensitivity. This analysis was done for the Great Lakes drainage area. The authors found that lands with forest cover had greater mercury concentrations in game fish than lands in agricultural cover. This was attributed to the enhanced deposition of mercury by forests, and abundance of wetlands and low productivity. Evers, D. C., J. G. Wiener, C. T. Driscoll, D. A. Gay, N. Basu, B. A. Monson, K. F. Lambert, H. A. Morrison, J. T. Morgan, K. A. Williams, and A. G. Soehl. 2011. Great Lakes Mercury Connections: The Extent and Effects of Mercury Pollution in the Great Lakes Region. Biodiversity Research Institute. Gorham, Maine. Report BRI 2011-18. 44 pages.
12-77	7 and elsewhere throughout the chapter	The authors indicate that mercury associates with thiosulfate groups in organic molecules. Mercury also associated with reduced sulfur groups, sulfhydryl groups.
13-1	15	data are
13-1	12	Earth systems
13-2	Figure 13.1	Should clarify the significance/meaning of "+" and "-".
13-5	25	Need to be clear by what is meant by units, molar or mass units presumably.
13-9	18	I am not clear on the statement that acidification driven changes in nitrogen occur at higher levels of nitrogen addition than for initial changes to the carbon cycle. Is there a reference for this? Is this statement true?
13-10	31	Recently, Warren et al. (2016) suggested that mobilization of dissolved organic matter may help mitigate the effects of temperature increases on brook trout survival. Warren, D. R., C. E. Kraft, D. C. Josephson, and C. T. Driscoll. 2016. Acid rain recovery may help to mitigate the impacts of climate change on thermally sensitive fish in lakes across eastern North America. Global Change Biology. doi:10.1111/gcb.13568.

Dr. Mark Fenn

GENERAL COMMENTS OF MARK FENN:

In some of the chapters, changes in various soil microbial communities in response to atmospheric deposition are described. To the extent that the current knowledge base allows, it would be very helpful to address the 'so what' question. If there are changes to microbial communities in response to deposition, what is the significance of these changes? For example, many studies show N deposition effects on ectomycorrhizal communities. What do studies say this means for things like forest sustainability and resilience? In many cases do the N-sensitive species just get replaced with functionally-equivalent species that are more N-tolerant?

---In Table 5-1: The descriptions (right hand column) are much more detailed than the tables in chapter 6. Is there a consistency issue here?

---Chapter 6 includes summary statements saying for what effects sufficient evidence exists to conclude these effects are caused by atmospheric deposition---either in the 2008 ISA or based on newer evidence. I don't see such statements in chapter 5.

MORE SPECIFIC COMMENTS OF MARK FENN:

EXECUTIVE SUMMARY (pp 59-82 of the pdf):

--p lxii (p 62 of the pdf), l 29: Southern California has considerable areas with N deposition greater than 15 kg/ha/yr.

--Table ES-1, description of section 8.6, I suggest using a different word for "lateration" (lateration of species richness). What does this mean? I couldn't find it in any dictionary.

--p lxxix (p 79 of the pdf), line 25: N loading rates of 50 or 100 kg N/ha/yr are mentioned. Presumably this refers to hectares of the total watershed (drainage basin) feeding into the estuary? This should be clarified within the text.

CHAPTER 1 (Integrated Synthesis):

--p 1-8, l 27-28: I question this statement that high elevation systems such as tundra tend to have high N deposition---they are just highly sensitive to modestly-elevated N deposition. Actually ionic N and S in wet deposition such as snow is highly dilute at these high-elevation sites in the Rockies or Sierra Nevada, although precipitation volumes are higher; and dry deposition fluxes are low compared to low and mid elevation sites. Cloudwater can lead to elevated N deposition, but this is mainly observed in the montane eastern sites (but these aren't alpine sites) and in mid-elevation in the West.

--p 1-18, l 17-21: This sentence could use some minor editing for clarification. It refers to the effects of deposition and then again to deposition, then mentions other sources of ambient N and S deposition. It

would be helpful to insert a word like 'non-atmospheric' in referring to other sources of ambient N. Then it is more readily apparent to the reader the contrast between N inputs driving the CL and those additional input sources that must also be considered.

--p 1-39, l 8-9: I would include central California in this statement, or more specifically mention the Sierra Nevada range where epiphytic lichen community changes resulting from N deposition are widespread.

CHAPTER 2:

-- p 2-6, l 15-24: On-road emissions of NH3 needs to be discussed with recent emissions information. The paper by Sun et al. (2017; see citation below) can be cited in this regard. The relative importance of ag vs vehicular emissions of NH3 varies across the landscape.
On-road NH3 emissions in the US determined by Sun et al. (2017) is twice that of the NEI. This increases on-road sources from 3% to 7% of the total U.S. NH3 emissions. On-road NH3 emissions are greater than ag emissions in counties containing near half of the U.S. population. The authors estimate that vehicles account for 13% of total U.S. emissions in winter and 53% of the U.S. population live in counties where vehicle emissions outweigh ag in winter; however ag emissions are also highly uncertain and could be underestimated. Mobile lab CO2 and NH3 emissions data are used by Sun et al. to determine NH3:CO2 emission ratios in six cities in the U.S. and China. Using this ratio and reliable on-road CO2 emissions estimates, the authors calculate on-road NH3 emissions in the US.

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.

Several studies have demonstrated that emissions of NH3, particularly from urban areas are underestimated (Kean et al. 2009).
Kean, A.J., Littlejohn, D., Ban-Weiss, G.A., Harley, R.A., Kirchstetter, T.W. and Lunden, M.M. 2009. Trends in on-road vehicle emissions of ammonia. Atmos. Environ. 43: 1565-1570.

<u>In regards to NH3 emissions from heavy-duty vehicles:</u>
As a result of the introduction of more stringent standards in California and by the USEPA for emissions of NO_x and particulate matter, heavy-duty vehicles now include a mix of natural gas engines with three-way catalytic converters and diesel engines equipped with selective catalytic reduction (SCR). For the latter device, aqueous urea is injected as a reductant for NO_x control (Thiruvengadam et al. 2016). Both of these engine/NOx control methodologies for heavy-duty vehicles result in NH_3 production. Light-duty diesel powered cars and fleet vehicles also use SCR for NO_x emissions controls and emit NH_3 (Bishop and Stedman 2015). Emission rates of NH_3 for heavy-duty vehicles using natural gas engines equipped with three-way catalytic converters had NH_3 emission rates that were 5-9 (Bishop et al. 2011) and 10 (Thiruvengadam et al. 2016) times greater than from light-duty vehicles.

Bishop, G.A. and Stedman, D.H. 2015. Reactive nitrogen species emission trends in three light-/medium-duty United States fleets. Environ. Sci. Technol. 49, 11234-11240.

Bishop, G.A., Schuchmann, B.G. and Stedman, D.H. 2011. Emission changes resulting from the San Pedro Bay, California ports truck retirement program. Environ. Sci. Technol. 46: 551-558.
Thiruvengadam, A., Besch, M., Carder, D., Oshinuga, A., Pasek, R., Hogo, H. and Gautam, M. 2016. Unregulated greenhouse gas and ammonia emissions from current technology heavy-duty vehicles. J. Air & Waste Manage. Assoc. 66: 1045-1060.

--p 2-43, l 5-6: This statement is problematic, because there is an AMON sites with NH3 data near Salt Lake City, but not a CASTNET site and thus no analogous pNH4 data fof SLC. The other site mentioned in this sentence seems to actually be in northern IL, bordering on southern WI.

--p 2-58, l 20-23: Uptake of N and possibly S by the canopy is another prominent mechanism by which throughfall fluxes are lower than wet deposition fluxes in forests where dry deposition or occult deposition is not elevated. This should be mentioned.

--p 2-60, l 3: This study was done in the Sierra Nevada mountains, not the Sierra Madre.

--p 2-67, caption to Fig 2-24: The figure caption begins: "Three-year average percentage of total nitrogen deposition by species". This could be edited to state up front that what is shown in the figure are the N species not measured in the monitoring networks. Likewise the text in lines 1-3 on this page could also be improved to make this clear.

CHAPTER 3:

Little of major significance has changed in recent years, and thus, I have no comments on this chapter.

CHAPTER 4:

--p 4-4, l 27-29: As mentioned in my comments for Appendix C, soil acidification has been well documented in the LA Basin, thus would be good to acknowledge a citation to that effect here. Here is my comment from Appendix C:
"--p C-136, l 16-17: Although the semi-arid ecosystems of southern California are not highly sensitive to acidification, soils in high N deposition areas of chaparral and forested areas in the LA Basin have acidified significantly; this is briefly discussed for chaparral on p. 149 of Pardo et al. 2011C (or more specifically the chapter citation, Fenn et al 2011a). Soil pH and base saturation changes in forests impacted by N deposition in S. California are discussed on pp. 154 of Pardo et al. 2011C (also see references therein)."

--p 4-6, l 11: Another point that could be briefly mentioned here is that tree species often differ in whether they preferentially exhibit canopy uptake of NO3 or NH4. For example, we found strong preferential canopy uptake of NO3 in the Pacific Northwest and cite many studies reporting the same (Fenn et al., 2013). Many studies show other conditions in which NH4 uptake by canopies is greater.
Fenn, M.E., Ross, C.S., Schilling, S.L., Baccus, W.D., Larrabee, M.A., and Lofgren, R.A. 2013. Atmospheric deposition of nitrogen and sulfur and preferential canopy consumption of nitrate in forests of the Pacific Northwest, USA. For. Ecol. Manage. 302, 240-253.

--p 4-10, l 11-12: If a reference is needed here, the original work establishing this CL for NO3 leaching is Fenn et al. 2008.

--p 4-10 l 36-37: To my understanding, this sentence isn't accurate as written. With increasing N deposition high N loss can and usually does occur, but most studies I'm familiar with show that high percentages of N are still retained in the ecosystem or watershed with high N deposition inputs. It's not unusual for 80% or so of N to be retained, while high leaching losses still occur. Losses downstream or from the riparian system as trace gaseous N losses is another issue, however.

--p 4-20, Table 4-20. In this entry about the publication by Jung et al. I would just note that subsequent work has shown that soils are not acidifying in the Athabasca Oil Sands Region---actually, likely the opposite because of high base cation deposition in the region. As evidence see the following publication:

Watmough, S.A., Whitfield, C.J., and Fenn, M.E. 2014. The importance of atmospheric base cation deposition for preventing soil acidification in the Athabasca Oil Sands Region of Canada. Science of the Total Environment 493: 1-11.

--p 4-24, Table 4-5: In the study by Lieb et al at Niwot Ridge, ambient deposition is 8 kg/ha/yr not 8-15 kg/ha/yr.

--beginning on p 4-43: Section 4.3.8 (Nitrogen Mineralization): One index that has been used is percent of the N mineralized that is nitrified---termed as relative nitrification.

--Daycent-Chem are mentioned in passing on p. 4-58, l 9 and p. 4-80, l 18, but are not discussed as the other models listed are discussed.

CHAPTER 5:

---p 5-12 to 5-13: Should more discussion be added regarding the uncertainty in the Bc:Al ratio which is protective for various species or conditions?

--p 5-30, l 10-11: The units for pH are given as meq/100 g. Does this actually refer to units of H^+? If so, this sentence should be edited to reflect this.

--p 5-31 to 5-32: The global scale analyses by Roy et al. (2012 and 2014) entails large uncertainties and doesn't seem to provide useful definitive information for the ISA. Seems these 2 paragraphs can be deleted.

--p 5-32 to 5-33: Likewise I question the usefulness of including the studies by Whitfield et al. in the ISA. These are from the Athabasca Oil Sands Region (AOSR) in northern Alberta where I have also worked on deposition and effects issues for several years. In a sense, the industrial emissions in the AOSR is more of a point source situation, at least in comparison to the acidic deposition scenarios in the eastern US. In the AOSR, the affected zone is primarily within 25 km of the industrial core (see Fenn et al. 2015). Along with Whitfield, Shaun Watmough and I published a more recent paper (2014) further documenting that BC deposition, much of it from fugitive dust apparently, that counteracts acidic deposition and may actually be causing alkanization of soils there. So this is a special case and a very

unusual situation in a dry boreal forest---so how applicable is this to the U.S. situation and how pertinent to the ISA? Not very in my opinion.

Here are the 2014 and 2015 papers:

Watmough, S.A., Whitfield, C.J., and Fenn, M.E. 2014. The importance of atmospheric base cation deposition for preventing soil acidification in the Athabasca Oil Sands Region of Canada. Science of the Total Environment 493: 1-11.

Fenn, M.E., Bytnerowicz, A., Schilling, S.L., and Ross, C.S. 2015. Atmospheric deposition of nitrogen, sulfur and base cations in jack pine stands in the Athabasca Oil Sands Region, Alberta, Canada. Environ. Pollut. 196: 497-510.

---Follow up comment for the AOSR: on p 5-48, lines 11-13, this conclusion is not the current thinking---again I emphasize that more current research (see Watmough et al 2014) concluded that because of unusually high base cation deposition in the AOSR, there is evidence that the soils are actually becoming more alkaline.

--p 5-40, Table 5-4: Jensen et al. 2014 is the first entry in the table on this page. The N addition rate is given only in terms of ammonium sulfate. Would be good to include what the addition rate was as N and as S for easy comparison to other studies in these more standard units.

--p 5-51, l 31-32: Would be good to say which nutrients were added in the fertilization treatments. I'm assuming base cations were not the main nutrients, but this should be specified.

--p 5-52, l 22-26: It is good to discuss the uncertainty issue here. However, I assume this applies to all of the similar studies reviewed in this section. Should this be emphasized in a more general way in reference to the other studies as well?

CHAPTER 6:

--p. 6-1 (pdf p. 414): In line 3 add 'productivity' to the name of section 6.1?

--p. 6-9: In the paragraph on mycorrhiza, what about the question of N-induced changes in mycorrhizal community changes----does this affect ecosystem function and stability and plant vitality? That is, do such mycorrhizal community shifts matter functionally?

--p 6-162 (pdf p 575): In the table, entry for Mediterranean California, the old N CL values are for NO3 leaching in streams from montane forested watersheds (17 kg/ha/yr) and tree mortality in mixed conifer forests (39 kg/ha/yr), but the new CL given is for coastal sage scrub vegetation type. So the old and new CLs are for very different veg types that occur in different elevations/habitats.

--p 6-17: In the section on European forest responses to N deposition, the following study shows a strong growth response of forests across a clear throughfall N deposition gradient in Italy:

Ferretti, M., Marchetto, A., Arisci, S., Bussotti, F., Calderisi, M., Carnicelli, S., Cecchini, G., Fabbio, G., Bertini, G., Matteucci, G., De Cinti, B., Salvati, L., and Pompei, E. 2014. On the tracks of nitrogen deposition effects on temperate forests at their southern European range - an observational study from Italy. Global Change Biology 20, 3423-3438.

--p 6-20: Many studies, including a review by Fog (1988; cited herein) and a slew of studies since then have shown that added N slows long-term litter decomposition and leads to OM accumulation.

--In Table 6-2 should the study by Suz et al. 2014 be included in the table? This novel study separates mycorrhizal growth responses out by their soil exploration abilities (ie, short or more extensive soil exploration).

--p 6-69, l 8-11: This is a good point, but aren't similar challenges also true for several of the other vegetation types discussed in chapter 6?
--p 6-74, l 6: The N addition value of 12.4 is incorrect. This is the throughfall deposition value at the most polluted site. N addition levels in this study were 5 and 30 kg/ha/yr.
--p 6-105, l 11-13: I suggest modifying the statement that N addition suppressed bark beetle activity at the high N deposition site. As mentioned in the paper, the larger point is that the high N deposition site was already so N-enriched that tree mortality and bark beetle activity was already high across all N addition treatments. Subseqent surveys (unpublished) have born this out with much greater mortality at the high N deposition site as predicted by Jones et al 2004. In Table 2 there does appear to be fewer living trees with bark beetle activity in the N addition treatments at the polluted site, but still there were high percentages of dead trees and living trees with beetle activity across all treatments.

--p 6-109, l 6-7: I question the statement that high elevation sites are hot spots for N deposition within the context of Western regions. High elevation sites likely have higher precipitation, but most is as snow with very dilute ionic concentrations. The statement from the report is true for some montane sites in the eastern US due to increased cloudwater N deposition, but these sites are relatively low elevation compared to alpine and subalpine Sierras and Rocky Mts. In reality, in the Rocky Mts and more especially in the Sierra Nevada, high elevation sites are rarely high deposition----usually the highest deposition may be ca. 6 kg/ha/yr, although measurements are uncertain. Hotspots in the West are typically low- or mid-elevation sites downwind of large urban regions, CAFOs or when both ag and urban emissions are influencing an area.

--Table 6-23: The N addition rate column gives the units in the column heading as kg N/ha/yr, but in many of the studies the unit of N pollution exposure is for gaseous atmospheric concentrations. Seems the column heading needs modification.

--p 6-134, l 1-4: I think it would be important to note here that in the Johansson et al (2012) study the N treatments were applied directly to the tree canopy as N solutions----thus, the lichen is being treated directly with the treatment sprays. Otherwise, as the text reads it seems that the typical ground application of N fertilizer is the method of application.

--p 6-146, l 9: Actually as can be seen from Table 2 of Fenn et al. 2010, the CL for both chaparral and oak woodlands is 5.5 kg N/ha/yr, just as given in Pardo et al 2011C. So the value for the CL on line 9 needs to be corrected accordingly and the following sentence stating a different CL given in Pardo et al 2011c isn't needed.

--p 6-157, l 3-13: In this discussion of the study by McLauchlan et al 2014, seems worth mentioning briefly the two potential explanations the authors also mention as to why no signs of N excess are evident----the annual burning and release of N and high capacity for N storage in soil.

--p 6-161, Table 6-28: In the second entry in this table (coastal sage scrub), the correct values for the low and high CLs for conversion to exotic grasslands are 7.8 and 10 kg N/ha/yr (see Table 13.3 in Pardo et al. 2011c). The current low value of 6 is actually for serpentive grasslands (see Table 13.4) and the current high value of 33 kg N/ha/yr given in Table 6-28 is for biodiverstiy of forest understory in the San Bernardino Mountains (see Table 13.5 in Pardo et al. 2011c).
--p 6-162, Table 6-28: In the entry for Mediterranean California (3rd from the last for this table), the older CL values from Pardo et al 2011c are incorrect. The correct values are 7.8 and 9.2 kg N/ha/yr (see Table 13.3 in Pardo et al. 2011c). This is for "decrease in arbuscular-mycorrhizal spore density, richness and percent root infection".

CHAPTER 7:

--p 7-4, l 15-17: Actually N is deposited in myriad forms, so this sentence should be modified in some manner. One possibility is just to say N is deposited in various reduced and oxidized forms, including organic N, and in wet or dry forms as well.

--p 7-18, l 9-11: I know the dogma in most all of the forestry textbooks is that net nitrification is not the norm. But, in many semiarid ecosystems, such as throughout California for example, net nitrification is the norm even in low deposition chaparral and forest sites. But relative nitrification (% net N mineralized that is net nitrified) is increased with chronic N deposition. See for example, Fenn et al. 2005:

Fenn, M.E., Poth, M.A., Terry, J.D., and Blubaugh, T.J. 2005. Nitrogen mineralization and nitrification in a mixed conifer forest in southern California: Controlling factors, fluxes, and nitrogen fertilization response at a high and low nitrogen deposition site. Can. J. For. Res. 35:1464-1486.

CHAPTER 8

--p 8-26, Section 8.3.6.4: In some of the studies cited, I presume that the possible direct biological effects of pH are not necessarily separated from Al effects perse? This same question can apply to other biological effects (i.e., organisms other than fish) as well. Controlled studies such as those of Kroglund et al. (2008) seem to look at these factors separately, but in many of the field studies, aren't Al toxicity effects potentially confounded with pH or ANC, even though effects aren't mutually exclusive?

CHAPTER 9:

--p 9-9, section 9.1.5: I think it is important to include this section on uncertainty and inconsistent findings in regards to putative N responses in high elevation lakes of the Western US. My understanding is that the indicator diatom species are not always fool-proof indicators of N enrichment and it is unclear at this point what drives community composition in some lakes of the Sierra Nevada and possibly the Rockies. One likely factor is climate change as discussed on p 9-24, l 27-31 and elsewhere in the chapter.

--p 9-20: Should the Heard and Sickman 2016 paper be cited in Table 9-2? This paper isn't yet cited anywhere in the document:

Heard, A.M. and Sickman, J.O. 2016. Nitrogen assessment points: Development and application to high-elevation lakes in the Sierra Nevada, California. Ecosphere 7: 1586.

--p 9-35, l 31-32: Should NH_3 be NH_4^+ in this sentence?

--p 9-38, l 7-8: I would add citation of the Fenn et al. 1999 paper and Riggan et al. 1985 here to cover the two mountain ranges mentioned.

--p 9-38, l 30 to p 9-39, l 3: The caveat should be mentioned here, however, that N deposition estimates at these high elevation sites in the Rockies and Sierra Nevada generally entail considerable uncertainty, particularly for dry deposition. Niwot Ridge probably has the best dry + wet deposition data because of more intensive measurements there. Current simulation deposition models certainly aren't reliable for deposition at these sites. I now see these points are briefly discussed on p. 9-41.

CHAPTER 10:

--p 10-49, l 8-11: I'm wondering why such high numbers of estuaries in California were listed as impaired in the survey by McLaughlin et al (2014) when in Fig 10-9 (national map) seems to show relatively fewer impaired estuaries in California. Seems somewhat inconsistent.

CHAPTER 11:

--p 11-30, l 11-13: I've worked with Kelman Wieder a bog/fen specialist in the Athabasca Oil Sands Region in northern Alberta where many bogs and fens are common. He has noted that responses to N of bogs and/or fens in Europe are different than in the US. Wieder et al. (2016) discusses possible explanations for these differing responses in the following paper:

Wieder, R.K., Vile, M.A., Scott, K.D., Albright, C.M., McMillen, K., Vitt, D.H., and Fenn, M.E. Differential effects of high atmospheric N and S deposition on bog plant/lichen tissue and porewater chemistry across the Athabasca Oil Sands Region. Environ. Sci. Technol. 50: 12630-12640.

CHAPTERS 12-14:

I have no comments/suggestions for these chapters.

APPENDIX C:

--Figs C-36A & C-37: Fig. C-37 based on empirical wet deposition data illustrates that TDEP (Fig. C-36A) greatly underestimates the relative proportion of N deposition that occurs in reduced forms in JOTR. We have bulk and throughfall data from JOTR that also indicates that NH4-N and NO3-N deposition are approximately equal.

--p C-136, l 16-17: Although the semi-arid ecosystems of southern California are not highly sensitive to acidification, soils in high N deposition areas of chaparral and forested areas in the LA Basin have

acidified significantly; this is briefly discussed for chaparral on p. 149 of Pardo et al. 2011C (or more specifically, Fenn et al 2011a). Soil pH and base saturation changes in forests impacted by N deposition in S. California are discussed on pp. 154 of Pardo et al. 2011C (also see references therein).

--p C-140, l 7-10): The incorrect literature reference is cited for this threshold; the correct literature reference is the Fenn et al. 2011b, the ESA monograph on air pollution thresholds.

--p C-149, Fig. C-42: The third study area, endpoint, reference shown from the top of Fig C-42 (Sierra Nevada Mts, NO3 leaching, Fenn et al. 2011a), has two errors: First of all this CL is for Rocky Mountain Western Lakes and the correct literature reference is Fenn et al. 2011b, the ESA monograph on air pollution thresholds (see Table 3 therein).

--p C-149, Fig. C-42: Likewise for the fourth entry from the top of this figure (JOTR-creosote bush scrub) the correct literature references are Fenn et al. 2010, 2015 (not Fenn et al. 2008).
And for the 12 entry from the top (JOTR, pinyon-juniper, the correct refs are Fenn et al 2010 and 2015.
The "California lichen protection" entry in the Fig. is missing the literature reference.
For the "Western Sierra Nevada lichen elmination" entry, Fenn et al 2008 is the original more detailed CL report.

MINOR EDITORIAL COMMENTS:

--Acronmyms: NHy is listed but not NOy (used on page ix, line 7 of Exec Summ)
--p 1-8, line 14: What is the year of the Rockstrom citation?
--p 1-21, l 27: Need to complete the sentence by adding words such as "is observed".

--p 2-9, equation 2-1: Seems that M should be defined in this equation.
--p 2-37, l 14: The value of 100 ppt is given. Why not call this 0.1 ppb, thus using the same units as in the figure referred to (Fig. 2-10).
--p 2-41: In the caption to Fig. 2-13 would be good to state that these are annual average NH3 concentrations.
--p 2-84: In the caption to Fig. 2-35 the phrasing oxidized and reduced nitrogen deposition suggests that oxidized and reduced N are each shown separately. Better to say in the caption something like total N deposition, oxidized + reduced, or the sum of oxidized and reduced N.

--p 4-29, l 6: Change "next" to "net".

--p 5-19, l 37: "American beach" is misspelled (should be beech).
--p 5-20, l 21-22: Seedling density increased from 16 to 32 seedlings/m^2, a doubling; yet this statement says the response was 'nearly doubling'. Why is the word 'nearly' used if it was a doubling?
--p 5-27, l 20: The units for soil CEC should be included.
--p 5-34, l 7: There is an extraneous "the" in this line. Likewise on p 5-48, l 18.

--p 5-34, l 12-17: The point of the Profile estimates being three times larger than the clay-correlation substrate method is repeated twice---seems redundant.
--p 5-34, l 29: Regarding "loss on ignition", shouldn't it be clarified what element or compound is being lost on ignition?
--p 5-36, l 22: It is unclear what the actual treatment amounts were for the combined N and S treatment. Was it 60 kg N/ha/yr + 60 kg S/ha/yr?

--p 6-21, l 30: After 'Much' insert the word 'of'.
--p 6-22, l 21-22: This sentence need to be edited, the wording "N additions increased" doesn't seem to be what is meant here. Sentence should be giving gene expression results in response to N additions.
--p 6-40, l 9-10: Sentence has typos, needs editing.
--p 6-68, l 11: The word 'on' seems to be extraneous and can be deleted.
--p 6-75, l 31: The words 'decreased lower' is redundant.
--p 6-76, Table 6-11: The word 'productivity' is misspelled in the Table caption.
--p 6-84, l 5: The word 'than' is missing from this sentence.
--p 6-84, l 5-6: Usage of 'also' and 'as well' in this sentence sounds redundant.
--p 6-135, l 5: "Ecto-mycorrhizae" doesn't require a hyphen.
--p 6-155, l 2: Sweden is misspelled.

--p C-132, l 17-20: Correct this sentence to read: "The CL was derived using linear regression of stream water NO_3^- concentrations during the winter high flow period and annual throughfall N deposition at 11 locations in the southern Sierra Nevada and San Bernardino Mountains." The correction specifies that throughfall deposition was annual, not only during winter; the original sentence was vaguely written in the Fenn et al. 2015 book chapter.

--p C-140: For the Fenn et al. 2008 entry (add stream NO3 leaching as an additional focus)

--p C-142: Insert the word 'on' after 'focuses'.
--p R-43: Reference by de Vries et al 2014b: The journal title needs to be added.
--p R-71 (and Table on p C-141): The reference by Grulke et al. should be 2009 not 2008 (I have the book and am a coauthor on the chapter).

Dr. Ivan Fernandez

Comments on selected sections as of 5.16.2017
Ivan J. Fernandez
University of Maine

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

(First External Review Draft)

PREFACE

1Xiii line 11 "agricultural activities" here may include changes in the extent and composition of forests, but that could be explicitly stated for clarity.

1xix line 37 It would seem more consistent with the previous discussion to refer to this as lower S deposition, rather than using emissions.

1xx lines 10-11 As stated, this suggests that from trees to bacteria and fungi, organisms behave the same to NH4 vs NO3 and other species. Is that valid and reflected in Section 6.2.1?

1xx line 20 Is the implication that one or both conditions (N deposition and acidifying deposition) are required for this sensitivity?

1xxii lines 22-26 Is this statement based on empirical data, CL calculations, or other? This suggests ongoing soil acidification at current levels of deposition, with no evidence for recovery. That is probably not true.

1xxiv line 21 'have' should be 'has'

1 xxiv line 23 Should 'adsorption' really be 'desorption' as used here?

1xxvi line 9 As worded, seems to suggest ANC is the one with concern. Revise.

1xxvii line 24 should be 'biogeochemical'

1xxvii line 30 altered growth of what?

1xxx line 17 should be 'causal'

1xxxii line 6-7 This sentence refers to a gradient study with apparently increasing fish Hg with increasing S deposition then mentions a single value (11/9 kg/ha/yr)? What is the comparison to here?

1xxxii line 10 'sulfur' is spelled out and throughout there is inconsistency with the use of symbols vs spelling chemical species out. This should be consistent throughout.

CHAPTER 1

1-3 line 31 “to” ecological effects?

1-8 line 18 The largest increase in what?

1-12 line 31 Omit the comma after NOx.

1-19 line 12 Need a space in the same.

1-25 lines 31-32 This statement seems to offer only correlation in support of a linkage between increasing tree growth and declines in SO2. Since many things may have changed at this location over this time period, did the evidence in support of this statement show a causal linkage, and if so, that should be included in this statement.

1-27 line 8 Delete the ‘all’ at the end of the line.

1-29 line 7 Change to, Base cations counterbalance acid anions in soil solution.

1-35 line 7 consequence(s)?

1-36 line 32 Might read better if ‘showing’ was inserted between ISA and ‘that’.

1-37 line 22 The sentence ending in ‘…ecosystem C storage” should continue on to say attributable to N (if that is the intent of the statement). There are many factors that could cause these changes, and the linkage to N deposition should be clarified here.

1-37 line 31 delete ‘both’

1-37 Can the second ‘analyses’ be replaced by ‘data’ in this sentence?

1-37 line 34 to 1-38 line 3 There is a need throughout this chapter to be clear on the intent of the statements related to recent trends and responses of ecosystems. This paragraph suggests that new evidence since 2008 suggests that community composition is occurring, but does not explicitly say as a result of N. The last sentence then suggests that the impact of N is now clear. Given the rates of change in temperature, precipitation, S deposition and other factors in the last few decades in the US, it is critical to be explicit in the linkages implied in these statements. The details may be in the supporting chapter, but this chapter also needs that clarity of intent.

1-38 line 15 Insert ‘community’ before composition.

1-42 line 31 to 1-43 line 4 In this opening paragraph it is unclear what contrast is being made. The first sentence talks about ‘sensitive’ ecosystems and gives three characteristics. The contrast seems to be that during this review new insight is available governing “vulnerability” and then a list of six factors are listed, but some are the same as the first. Is there meaning in the choice of sensitive vs vulnerable here? Is there intent to highlight new factors in the listing? Needs clarification.

1-44 line 8 What kind of ‘Al concentrations’? Exchangeable? Soil solution? Total?

1-49 line 22 It is probably best to say “evidence for biological recovery” to be consistent with the chemical recovery statement, and to clarify the difference between what is known through research and what may be true but unknown if the research has not been done.

1-51 line 13 I would insert “relatively” in front of mobile anion. SO4 is almost always partially retained, as is even Cl, and those not a strictly conservative anion in these acid-sensitive watersheds.

1-54 line 32 No comma needed after ISA.

1-54 line 34 Comma needed after ‘peak’.

1-55 lines 4-6 It would probably be better to frame this as sources of base cations, and sinks of base cations, rather than increases and decreases. You can have weathering and atmospheric deposition providing base cations while labile base cation supply in the soils is going down.

1-55 line 5 Missing an end bracket after PnET/BGC.

1-59 line 32 thresholds ‘for’ instead of ‘to’

1-62 lines 8-10 Some would question then the definition of biological recovery, which may be dealt with later in the chapter. Throughout this section and here, the term biological recovery sounds like it means full recovery, which some would argue is not the case if you have new species. In addition, the use of recovery can often be interpreted in this section as full recovery, yet I think the use of the term typically means some evidence of recovery, but not necessarily full recovery, however it might be operationally defined. Perhaps earlier in the introduction of the concepts of biological and chemical recovery the distinction between full and partial recovery could be articulated. The chapters may deal with this but that should be clarified in the Integrated Synthesis as well.

1-62 line 14 Is eq/ha/yr the intended units for ANC here? Is that correct?

1-63 line 7-8 ‘In another...’ is not a complete sentence.

1-63 line 9 change ‘have’ to ‘has’

1-66 line 4 should be kg N/ha/yr, not kg/N/yr

1-66 line 32 Its not quite clear what the intent of this sentence is for the contribution of nutrient-enhanced coastal acidification. Is this intent of this sentence for coastal acidification’s contribution to ocean acidification, or to the ability of organisms to build shells?

1-67 line 27 add a comma after end bracket

1-67 line 32 change ‘receiving’ to ‘receive’

1-68 line 5 eliminate period after ecosystems

1-68 lines 18-20 The first two sentences are not well integrated. After the first sentence makes a statement about coastal acidification process, the second sentence jumps to the 'ocean', but it is not clear if that ocean concept here is inclusive of coastal systems or this is making a statement about another process linked, but elsewhere, having to do with open ocean acidification.

1-73 line 3 I was expecting a map for Fig 10-8 given this sentence, but it is something else entirely. Is that the intent here?

1-80 line 6 omit the comma

1-84 line 34 Change to "This first draft ISA…"

1-85 lines 3 It could be useful to follow the sentence about the N reference to state that not such similar review of climate modifications of response to S has been done.

Chapter 4

4-4 line 29 This statement omits the mechanism of denitrification just mentioned about. Since it is stated without qualification, all mechanisms should be included in the list.

4-4 line 11 Odd use of tense here with 'was'. If this study is the current state of knowledge, then it 'is' unclear.

4-9 line 10 The Campbell reference here links to a reference by Bates on S. Please verify ALL linked references in the document.

4-9 line For clarity, state '…increases soil N concentrations.'

4-10 lines 5-12 This paragraph discusses NO3 leaching as an absolute. That is, conditions when it occurs and when it does not. This should be clarified to indicate whether and when 'chronic' NO3 leaching is intended, versus ANY NO3 leaching, which occurs regularly at N deposition levels below the thresholds discussed. For example, NO3 leaching during spring snowmelt is common in even low deposition scenarios. This is discussed further down in dealing with the kinetics of NO3 leaching.

4-11 line 24 insert 'the' before microbial community.

4-19 line 9 I would use the word 'cations' rather than 'base cations' here, as SO4 will leach with any positively charge ion, not just the bases. If the bases are abundant, then they will dominate and if the supply is low, they will be depleted. The cation could all be Al if it starts out acidic.

4-19 line 20 delete 'deposition' as it is the S that is accumulating.

4-20 Table 4-4 Is it by intent that the section is called S accumulation, adsorption and leaching but the table is only about the last two of these? If so, why?

4-22 lines 3-4 Not true, all base cations are not essential plant nutrients. Replace 'are' with 'includes'

4-22 line 7 replace ‘causes’ with ‘can cause’, since if base cation supply is high it does not necessarily have to cause acidification of the soil.

4-22 line 25 The tense sometimes used in this writing is present tense, when the statements, findings, or conditions are all now past. It seems more prudent to use the past tense, particularly if one result is later followed by a new finding with a different conclusion. Thus, here, an acidification threshold ‘was’ calculated makes more sense.

4-22 line 27 located ‘in’ one of the…

4-22 line 31 N plus S addition

4-23 Table 4-5 Both table and section are consistent as to heading, but while the S and N sections alluded to processes, the base cation sections are just labeled base cations. It would be better to have a parallel structure to these discussions, starting with the headings and table titles.

4-26 line 23 It does not make sense to state that there is no risk <10 when the statement just proclaimed adverse effects at 1.0 or 0.2, both meeting the criteria of <10?

4-26 line 31 I would revise this to include parenthetically something like (often as well-drained soils with <60% soil moisture) after the aeration factor since both aeration and soil moisture content are about the available O2.

4-27 line 5 ‘are’ associated

4-27 line 10 ratio ‘is’ a…

4-27 line 24 Change to ‘terrestrial ecosystem soils’ if that is the intent. Many would consider groundwater and riparian zones all parts of a terrestrial ecosystem so I believe the contrast intended here is specifically with soils.

4-37 line 11 End the first sentence with the phrase ‘as a function of N addition’ to constrain the focus of the Eisenlord study.

4-40 Table 4-7 Freeman and Zak entry, Effect of Deposition, second sentence needs rewording, suggest ‘These results represent a plausible…’

4-43 line 1 change ‘of’ to ‘than’

4-44 line 3 I am not sure what this intends to say as worded?

4-44 line 5 That seems an odd statistic to highlight, since the range of available N increases across all levels of N addition could be quite large, particularly in instances when the reference condition is near detection li\mits.

4-44 line 18 increases production of what?
4-48 line 19 change ‘decreased’ to ‘decreasing’

4-48 line

Figure 4-9 title seems a bit ambiguous by stating “under nitrogen”? What about under elevated nitrogen inputs or deposition?

4-53 line 2 I question the value of using “…with increasing depth.” Here as the key factor. While technically perhaps correct, the rest of the paragraph that explains that statement does not talk about depth but about the contrast between organic soil materials and mineral soil materials. Yes, the organic soil materials are at the surface, but the phenomena described are influenced by the different type of soil materials more than a simple depth factor.

4-55 lines 12-16 This is a long sentence following the introductory sentence on long-term monitoring. The Hubbard Brook reference reads well, but the Niwot Ridge portion seems oddly structured and is pointing to a 10 year experiment and not long-term monitoring as worded.

4-55 lines 17-21 For the three causes of the shift to a net N sink, (1) does not say what about gaseous fluxes caused the shift, (2) seems straight forward, and (3) seems to define sink with accumulating which mean the same thing.

4-55 line 31 threshold for what?

4-60 line 34 What are the units for these measures of variability?
4-63 line 18 drop ‘required’

4-64 line 9 change to ‘…for updates of this model.’

4-69 line 17 omit the comma

4-71 lines 10-14 This starts off talking about “since” the publication of Pardo which is 2011, and then discusses older publications (2009 and 2010) as the new work?

4-73 lines 20-22 I am not sure I agree with this sentence. There are naturally occurring anions in soils that range from Cl from marine aerosols to organic anions of varied mobility. The impact of mobile anions from S and N is partly attributed to their various mobility characteristics as well as their concentrations, which are elevated due to deposition. That is oversimplified in this statement.

4-73 line 18 and line 21 Perhaps say “..dominant naturally derived acid…’ You can have sulfuric acid that is ‘natural’ in weathering minerals.

4-78 line 6 to be consistent with the first two items listed, the last should probably be ‘…and acidity (pH).’

4-79 lines 16-17 The tense is plural so it should be ‘analyses’ and sulfur is not capitalized.

4-79 line 29 If the text uses trivalent Al as the Al variable, then Mn should be divalent.

4-80 line 22 ‘…can cause accelerated base cation leaching…’

4-80 lines 23-26 Something is wrong with this sentence.

Overall Chapter 4 appears to be a thorough update on the core topics. There is significant repetition of the same material in several places. While there is mention of N and S deposition effects on Ca, Al and other base cations, there is no mention in this chapter of the influence of particularly N deposition on phosphorus dynamics in soils, although this discussion may be preserved for linkages to the biotic implications in later chapters. Nevertheless, since the importance of many of the N dynamics in soil biogeochemistry discussed in this chapter are biotic in nature, research on the impacts of atmospheric N deposition on N-P dynamics should be mentioned.

Chapter 5

Table 5-1 In the description for the Beier work both ‘correlated’ and ‘related’ are used. If they are both the same type of correlative evidence, use correlated for both. In the Horsley entry, the term ‘base cation-acid cation nutrient gradient’ is unclear. Does the addition of the word nutrient imply this includes N and/or P? Does the word add anything to the meaning? Pitel and Yanai entry description in the middle should be ‘Site(s)’. Note: Overall, this table is a good summary of key studies on soil linkages to effects dealing mostly with acid-base dynamics. It could be useful to know more about the soils. This is particularly critical when quantitative findings are reported as it is essential for both context and comparison among studies to know if these statements are about O horizons, mineral subsoils, or A/Ap horizon systems. I would try to include that in the text or by adding a column.

5-12 line 29 comma should go after Spranger ref, not before

5-13 line 23 this is not the first time the Mg symbol is used

5-13 line 30 Since the form of the analyte is specified for base cations or H and Al, what form does ‘Al concentrations’ at the end of the sentence represent?

5-15 line 3 I am still looking for consistency in the use of related versus correlated. If it was a correlation, then best to use that term. If there was more evidence than the statistical correlation that the authors reported, then related works.

5-15 line 9 Is this an editing issue for the whole document or is there a convention for how often chemical symbols need to be redefined? Seems inconsistent. K has already been used.

5-18 line 5 Since this says “the” Ca:Al ratio it is unclear whether there was a correlation with ANY value of this ratio, or this is just about the 0.03 threshold. Please clarify.

5-29 line 23 Previously noted, but this sentence uses both correlated and related, and I suspect the same meaning is intended for both. Thus, the same term should be used if this is the case.

5-30 lines 10-12 Similarly, previously noted but here is another example of where the strength of this ISA as a resource can be enhanced by being clear on the soil materials involved. The Pabian study sampled only O horizons. The the meaning and value of the ranges reported would be dramatically different if this included both O, A, and mineral subsurface B horizons. If

the data are reported it suggests that the reader will benefit from the quantitative information, but the type of substrate is critical. We likely would not lump peat bogs and upland forest B horizons in set of characterization data. This is valid for all similar discussion of particularly forest soil properties.

5-30 line 21 change 'if' to 'of'

5-31 line 12 Ideally, it would be good to point to the section(s) of Chapter 4 that support evidence of ongoing acidification, rather than point to the whole chapter.

5-31 line Too many plurals, change to 'mountain top and ridge forest ecosystems'

5-34 lines 17-18 As written, these results do not differentiate between differences in how the models perform versus actual differences in weathering rates. Thus, it seems to be missing a step to state that Pennsylvania therefore has a higher weathering rate as stated here.

5-34 line 31 change 'were' to 'was'

5-35 line 28 insert 'the' before nontreated

5-35 line 29 '…high(er)…'

5-48 line 16 These units are incorrect. The discussion should be standardized to one format of units to avoid confusion for the reader, and these types of errors. These should be mmolc/m2/yr

5-49 line 18 Should be 'kg N/ha for 3 years'

5-55 line 18 delete the second 'responses'

5-56 line 18 insert 'base' before the word cations

5-56 line 22-24 There is something wrong with the sentence structure here.

Chapter 6

6-5 line 6 It appears the beginning of the sentence is missing.

6-5 line 18 change 'changes' to 'change'

6-5 line 30 I think this line is where the issue is, but this sentence does not read correctly.

6-7 line 36 I think these should be 'meta-analyses' and 'syntheses'.

6-12 Table 6-1 In the Effect of …. column, entries that state 'Increase with NO3, NH4, and NH4NO3 not significant' are hard to interpret. The table footnote says only significant differences are listed. That would suggest here that there is a significant increase in NO3 and NH4, but then it is confused by saying AND NH4NO3, suggesting that too was significant, but then it is followed by not significant? What is not significant? Why show it? Clarify.

6-15 line 23 change 'have' to 'has'

6-15 line 33 Delete 'Notably' and capitalize 'Although..

6-21 line 30 '…much (of) this…'

6-32 line delete one of 'must be'

6-34 line '….photosynthesis, (and) dark respiration…'

6-40 line 9-10 '…in previous to N cycle…' does not make sense as written.

6-43 line 28 '…concentration (in) three of four…'

6-54 line 10 Change 'changed' to 'change'.

6-55 line 17-18 It would be useful here to state what the lowest treatment rate was as the highest rate is noted earlier, and this result on the lowest rate is being highlighted.

6-84 line …'smaller (than) the response…'

6-86 line 9 Change comma to period and start new sentence with 'Changes in an individual…'

6-86 line 15 '…the presence (of) non-native…'

6-89 line 11 Add 'was reached' to the end of the sentence?

6-91 lines 8-14 As noted earlier, it would be far more valuable if information about what soil horizons were targeted in these studies was included given the significant differences in organic and mineral soil systems.

6-95 line 8 '…(were) a large number…'

6-95 line 12 Change 'forests' to 'forest'.

6-100 line 18 Change 'to' to 'with'.

6-103 line 19 '…affect(ed)…' Also, nice example in this sentence of identifying soil material type in defining the response end points.

6-109 line 12 '…(in) alpine…'

6-110 line 30 '…both (of) the two dominant…'

6-117 line 16 '…ecosystems(.)'

6-118 line 2 Change 'explaining' to 'explained'.

6-139 line 20 Delete 'Temporally" and start the sentence with Whereas…

6-139 line 24 Change "or" to "at the"

6-143 line replace 'of' with a comma

6-149 line 7 …ecosystem(s)…

6-149 line 27 …parks (in) the…

6-149 line 35 Given the complete focus on N, it could be useful to insert 'biomass' after 1000 kg/ha to assure no confusion.

6-157 line 14 Change 'documented efforts' to 'documented research', 'CL' efforts to 'CLs'

Note: This discussion of CLs is described as work in North America but seems to lack any insight from international research on this topic, particularly from countries with comparable ecological conditions as in Europe. Some recognition of that science seems warranted to put in context the US work.

Chapter 13

13-1 line 15 change to 'pool(s)' and data 'are', not 'is'

13-1 line 17 I would reword the sentence to state that there remains 'significant uncertainty' relative to climate effects on ecosystem response to N. Using 'certainty' as the criteria is too absolute for most of the science in this ISA.

13-5 lin 14 '…supply (can) alter…'

13-5 line 17 Technically I think photo-oxidation would be an additional mechanism of C oxidation not listed here.

13-5 line 19 '…until biotic N demand is satisfied or another factor becomes limiting…'

13-5 line 29 So the 'we' here makes me realize that the excerpt is verbatim. As such, no changes suggested should be made, unless ISA commentary on this single publication is included in this narrative.

13-8 line 23-25 This statement excludes the potential effect on soil respiratory losses. Should it read 'while not increasing respiratory losses' to include both plant and soil respiration? The authors do not appear to have measured soil respiration separately.

13-9 lline 8 delete 'effects'

13-9 line 8-9

13-12 line 16 I know this is all a direct quote, and I checked the reference and this is what they wrote. However, it seems like this was intended to read '…that did (not) follow…'

13-12 lines 23-24 As with the above, accurate to the publication, but this seems to have a sentence structure issue.

13-12 line 32 'is' should be 'are'

Chapter 13 on climate uses a direct quote of a single published paper to discuss the implications of climate change on the subject of this ISA. The Greaver et al. 2016 reference is an excellent synthesis on this rapidly emerging subject and is justifiably a focus of this chapter. The chapter title is 'Climate Modification', whereas the Greaver et al. 2016 manuscript is

focused on ecological responses to N as influenced by climate. As such, a broader conceptual framework for how climate change can influence the effects of N and S deposition is lacking, although many factors are included in the key reference, and many relevant references are scattered throughout the ISA. However, the beginning of this chapter does not lay out that larger framework for how climate change is relevant to the topic of this ISA, before focusing on a recent synthesis paper to address the topic. For example, climate change topics related to N and acidification and recovery such as the changing vernal transition and snow cover (e.g., Groffman et al. Biogeochemistry 56:135-150, Sorensen et al. Ecology 97:3359-3368), hydrology and phenology (e.g., Groffman et al. BioScience 62:1056-1066, Fuss et al. Biogeochemistry 131:35-47), altered hydrological implications for base cations (e.g., Kopacek et al. EST 51:159-166), extreme weather and lake recovery (e.g., Strock et al. 127:353-365) or altered glacial meltwater impacts on N delivery to alpine lakes (e.g., Daggett et al. Aquat Sci 77:511-521) are not included in this chapter. Given the rapid rate of new publications on this topic, it should be possible in the revision of this ISA to offer addition relevant references and a framework for the broader topic in introducing the topic.

Dr. James Galloway

General Comments

1. The ISA is an excellent compilation of critical research on the impacts of N, S and PM on ecosystems.
2. Within each chapter, at the end of each major section, it would be helpful to have a summary statement about what is new with respect to Causal Determination.
3. At the end of each chapter, it would be helpful to have a summary of what is new since the 2008 ISA with respect to Causal Determination (such as done in Chapter 9).

Executive Summary

Page lx, lines 17-18: "This ISA determines whether oxides of nitrogen, oxides of sulfur, and particulate matter concentrations in the air or depositing from the air cause ecological effects". Although NH_3 is mentioned (or implied, i.e., NHx) in many places in the document, it is not in this key sentence in the ES. Can it be?

General: Table ES-1 is excellent, as is the entire ES. What I found missing was a section at the end that pulled everything together with respect to addressing (or not) the issues that led to the the Administrator's actions with respect to the 2008 ISA.

Chapter 1: Integrated Synthesis

General: This chapter provides an excellent introduction to the ISA. To make it more effective, I suggest that the key findings be placed in bold font, as was done in some places (e.g., Section 1.8.; end of Chapter 3.

Chapter 2: Source to Deposition

Page 2-15, Section 2.3.2: Organic N & S. This section contains a nice concise summary of the topic. However, at the beginning it says the organic N deposition 'must be considered', but then does not say the impact on our knowledge about the effects of N deposition if they are not considered due to limited data.

Page 2-29, Section 2.5.4.1: Remote Sensing of Ammonia. There is an excellent recent paper on this topic: Warner, J. X., R. R. Dickerson, Z. Wei, L. L. Strow, Y. Wang, and Q. Liang (2017), Increased atmospheric ammonia over the world's major agricultural areas detected from space, *Geophys. Res. Lett*., 44, doi:10.1002/2016GL072305.

Chapter 4. Soil Biogeochemistry

Page 4-72, Section 4.7: Summary: This section is well done but appears to be more than a 'Summary'. At ~12 pages long it is difficult to find the key summary messages. The authors might consider a more targeted approach in presenting the key messages from this chapter.

Chapter 5: Biological Effects of Terrestrial Acidification

Page 5-55, Section 5.61: Physiology and Growth: In this section is the statement "Consistent with the findings of the 2008 ISA, the body of evidence is sufficient to infer a causal relationship between acidifying N and S deposition and the alteration of the physiology and growth of terrestrial organisms

and the productivity of terrestrial ecosystems". The statement implies to me that the 2008 ISA found the same thing. However, in Table ES-1, it says that the 2008 ISA did not include this finding.

Chapter 9: Biological Effects of Freshwater Enrichment

General: While the summaries at the end of each section are helpful, as noted earlier it would be helpful to have a summary at the end of the chapter.

Chapter 13: Climate Modification

General: This chapter was excerpted from Gleaver et al. (2016), *Key ecological responses to nitrogen are altered by climate change*. As the title indicates, the paper focuses on nitrogen, and in that regard, it is an excellent foundation for this chapter.

General: With respect to sulfur, the chapter authors state that "relatively little work is conducted on how climate modifies ecosystem response to S (deposition)". While this may be correct, there are some commonalities between N and S that should be explored. For example, there are several statements in Figure 1 of Gleaver et al. (2016) that might also apply to S. We recommend that the authors use the 2106 paper as a tool to determining where there might be link between climate change and ecosystem response to sulfur deposition.

General: In addition, there are other resources that may be of assistance to addressing the chapter's charge. A notable one is the *Third National Climate Assessment* (Melillo et al., 2014), especially the chapters on Ecosystems (Chapter 8) and Biogeochemical Cycles (Chapter 15).

Chapter 14: Ecosystem Services

General: This is a valuable addition to the ISA, but since it is a new approach since 2008, it would be helpful for the chapter to lay out suggestions for future work to make the findings more useful to future assessments.

13. Climate Modification of Ecosystem Response

Introduction

Chapter 13 describes how climate, specifically temperature and precipitation, modify ecosystem response to nitrogen and sulfur deposition. CASAC made the suggestion to include this topic in their comments on the draft Integrated Review Plan in April 2016.

Charge

Please comment on the accuracy, clarity, level of detail, and relevance of information presented on modification of ecosystem response due to changes in temperature and precipitation

Comments

This chapter was excerpted from Gleaver et al. (2016), *Key ecological responses to nitrogen are altered by climate change*. As the title indicates, the paper focuses on nitrogen, and in that regard, it is an excellent foundation for this chapter.

With respect to sulfur, the chapter authors state that "relatively little work is conducted on how climate modifies ecosystem response to S (deposition)". While this may be correct, there are some commonalities between N and S that should be explored. For example, there are several statements in Figure 1 of Gleaver et al. (2016) that might also apply to S. We recommend that the authors use the 2106 paper as a tool to determining where there might be link between climate change and ecosystem response to sulfur deposition.

In addition, there are other resources that may be of assistance to addressing the chapter's charge. A notable one is the *Third National Climate Assessment* (Melillo et al., 2014), especially the chapters on Ecosystems (Chapter 8) and Biogeochemical Cycles (Chapter 15).

Melillo, Jerry M., Terese (T.C.) Richmond, and Gary W. Yohe, Eds., 2014: *Climate Change Impacts in the United States: The Third National Climate Assessment*. U.S. Global Change Research Program, 841 pp. doi:10.7930/J0Z31WJ2.

Dr. Frank Gilliam

Summary/Comments on ISA for Oxides of Nitrogen, Oxides of Sulfur, and Particulate Matter—Ecological Criteria, Chapter 11: Nitrogen Eutrophication Effects in Wetlands

Frank S. Gilliam

Hans Paerl

Chapter 11—Nitrogen Eutrophication Effects in Wetlands—reviews recent literature on the effects of excess nitrogen (N) on a wide variety of wetland ecosystems. Not only do the authors of this chapter use wetland classification as established in relevant literature, but they also provide a useful, informative summary table to distinguish among numerous wetland types based on several classification criteria, including soil-based, hydrology-based, and soil/hydrology/vegetation-based classifications. This is a rather exhaustive list that includes more wetland types than is covered in the review. Those emphasized in the chapter are salt and freshwater marsh, mangrove, riparian wetland, and bog/fen. This is an extensive handling of literature, primarily since the 2008 ISA, and the findings regarding N-mediated effects on the biogeochemistry of wetlands is largely confirmatory of the results reported in the 2008 ISA, i.e., there is a causal relationship between N deposition and the alteration of biogeochemical cycling in wetlands. This new ISA, however, adds to the body of knowledge regarding responses of wetlands to excess N by including other response criteria, e.g., plant physiology and plant architecture. The body of evidence is sufficient to infer a causal relationship between excess N and alteration ecophysiology, species richness, community composition, and biodiversity in wetlands.

As an aside, it is easy for an ecologist, such as myself, with an east coast bias to think of coastal wetlands as predominantly an east coast phenomenon, and that would be incorrect. Thus, the authors of this chapter are to be commended for their even handling of the general topic of wetlands and their biogeochemical and plant community responses to excess N, wherein west coast references from California to Washington State are reviewed, along with east coast references from Florida to Maine, and along the Gulf Coast from Florida to Texas.

Studies reviewed regarding biogeochemical responses of widely contrasting wetland ecosystems to excess N generally involved experimental additions of a wide variety of forms of N, from NH_4NO_3 to urea and even sewage sludge, and at widely varying amounts, in many cases well in excess of 1000 kg N/ha/yr. Most response variables were N mineralization, nitrification, and denitrification, but also soils

microbial communities, especially the specialized bacterial groups associated with nitrification and denitrification. Effects of N on soil carbon (C) cycling in wetlands were minimally addressed, primarily because a meta-analysis (which included wetlands, among other non-forested ecosystems) failed to find any effects of N on net ecosystem exchange of carbon. However, in support of findings of the 2008 ISA, a separate meta-analysis revealed that experimental additions of N can increase methane (CH_4)—an important greenhouse gas—by nearly 100%

Regarding plant response variables, quite numerous studies were reviewed that examined the effects of N (often combined in several studies with effects of CO_2) on plant production and biomass. Results of these studies were understandably varied, considering the widely contrasting wetland types and forms/amounts of added N, along with whether experimental treatments included CO_2. New criteria in this ISA not included in the 2008 ISA included plant stoichiometry and ecophysiology. Because stoichiometry examines the balance of nutrients in organisms, and because excess N can alter greatly the availability and uptake of essential plant nutrients, this is an important characteristic for consideration. As before, there were widely varying results among the numerous studies reviewed.

Other plant response variables include plant architecture, demography (including reproduction and mortality of individual wetland species), and biodiversity. The latter is especially relevant given that high biodiversity is characteristic of most wetland ecosystems. Once again, given the numerous wetland types represented, along with contrasting treatments, results varied greatly among studies. Finally, critical loads of N for wetlands were considered. At the time of the 2008 ISA, such critical loads had either not been determined or not been published for wetlands. Since that time, however, critical loads have been published, generally varying between 3 and 13 kg N/ha/yr.

As has already been suggested, the authors of this chapter are to be commended for their handling of this broad, challenging topic. The chapter seems well-written and comprehensive with regard to references and background information on these various ecosystem types. Indeed, the number of studies reviewed ranges from extensive to nearly exhaustive. We see no need for further changes.

Dr. Daven Henze

1. Executive Summary and Chapter 1

xi,11: seems unnecessary to list NH4+ here, given that NHx is listed on the first line of the following page.

xii,25: Would be easier to say increases in deposition of NH3, or reduced N, rather than NH4+, whose dry deposition has decreased throughout much of the eastern US and increased only slightly elsewhere (Fig A-17).

xiii,33 and 1-8,11-13: These few lines about the Anthropocene is an interesting yet tangential side-point that could be removed.

xix,30, and 1-14, 8-9: This seems to overlook significant trends in different regions of the country, where total N deposition rates have increased owing to increased NH3 dry dep (stemming from increased fertilizer application), or decreased owing to decreased oxidized N deposition (stemming from reduced NOx emissions). Maybe just reword to say that the "national" rather than "overall" deposition of N has been relatively unchanged? Otherwise, the word "overall" might just be taken to mean oxidized + reduced, or wet + dry.

xxii,10: Alpine ecosystems also receive N deposition owing to sources in non-adjacent regions (e.g., Lee et al., 2016).

1-12, line 28: Fig 1-2: Does not account for NH3 from vehicles, despite recent studies showing contributions of vehicles to NH3 emissions being the dominant source in many urban counties (e.g., Sun et al., ES&T, doi:10.1021/acs.est.6b02805, 2017).

1-12, 8: And mineral dust, particularly given this refers to PM (not PM2.5).

1-13, 2: Soils/fertilizer can also be a source of NOx emissions (e.g., Hudman et al., ACP, 2012). This is further discussed in Chap 2 (page 2-6).

1-13, section 1.2.2: Doesn't explicitly say how wet deposition is measured or modeled.

1-27, line 8: remove "all"

Fig A-11: why such high nitrate dry dep in Florida in 2000-2002? We don't see this peak in particulate nitrate concentration estimates in Florida in Fig 2-12 (page 2-40) or nitric acid (Fig 2-11, page 2-39).

2. Atmospheric Chemistry

Table 2-1: This table strikes me as overly precise; as mentioned in the supporting discussion, there are considerable uncertainties associated with many of these values. At the very least, it should specifically state the year and version of the NEI being used. At best, some estimates or uncertainties, ranges, or

notes on levels of certainty (high, medium, low) could be included.

Another question about Table 2-1 is the apportionment of soil NOx – about 10 % of soil NOx emissions are owing to fertilizer applications (e.g., Hudman et al., 2012). Why are these then not classified as agricultural emissions, as is the emissions of NH3 owing to fertilizer?

2-5, line 4-18: Evidence of NOx emissions overestimates also in Texas during the GoMACCS campaign (Yu, S. C., et al. (2012), Comparative evaluation of the impact of WRF-NMM and WRF-ARW meteorology on CMAQ simulations for O_3 and related species during the 2006 TexAQS/GoMACCS campaign, *Atmospheric Pollution Research, 3*(2), 149-162.) And note the paper by Travis is now published.

2-6, 15-24: NH3 emisisons are quite uncertain. This should be discussed, including references to the relevant literature. NH3 emissions from transportation may also be underestimated in the NEI by x2 (e.g., Sun et al., ES&T, 2017).

2-6, 35: Some top-down studies don't use CTMs, such as the constraints on NOx and SOx emissions derived from OMI (e.g., Fioletov et al., GRL, 2011).

2-14: What about uptake of HNO3 by coarse-mode dust?

2-17, 15: The lifetime of NOx is typically a few hrs, owing to rapid dry deposition. It seems odd to pit this against the lifetime of HNO3, which is also short, but I'm not sure that the latter is significantly shorter.

2-17, 19-26: Perhaps this is just a writing style issue, but the text seems to make a point about NOx lifetime vs that of HNO3, and then backs this up (for example) by reference to the lifetime of NH3 and SO2, which doesn't flow (logically). Perhaps all of these are just examples of the broader topic of this paragraph (different species have different lifetimes), which should be clarified.

2-20 16: Strangely worded, as emissions cannot be transported (emitted species can be though).

2-17, 27 through 2-21, 14: This very generic section on atmospheric transport feels a bit dated, and somewhat out of place. While some review of basic terminology relevant to transport discussions seems warranted, the content here seems a step or two removed from the aspects of transport critical for discussion of N and S. It seems like boilerplate description of transport associated with the Bermuda high copied from another report. This is but one mode of transport of interested to S and N deposition. Boundary layer dynamics and mixing, mountain up-slope effects (e.g., transport into Rocky Mt National Park from ag and urban areas to the east), continental-scale (e.g., transport to Rocky Mt. NP from CA) and transcontinental-scale long-range transport all come to mind.

2-23, 33: sometimes models are used here as well

2-24, 26: which has shown → have shown

Section 2.5: This section focuses on (some) ground network measurement techniques and remote

sensing techniques. What about measurements from aircraft (e.g., work from John Nowak on NH3), mobile platforms (Mark Zondlo, also NH3), or other research-grade techniques? Or does the title of the section just need to be renamed?

2-29, Section 2.5.4.1: This section unfortunately does not include the best remote sensing measurements available for NH3 – those from CrIS. These measurements essentially combine the accuracy of TES with the spatial coverage of IASI. See details in Shephard and Cady-Pereira, AMT, 2015, and Dammers et al., AMTD, 2017.

2-30, 1-4: This is a strangely incomplete and out of place overview of the use of remote sensing to constrain NH3 emissions. It doesn't even mention the most relevant works targeting US emissions. Regardless, discussion of top-down emissions estimates should be the subject of another sections (e.g., end of Section 2.2) and should be kept separate from a description of the observations here.

3-33, 3: Shouldn't discussion of NO2 have been in the earlier section on OMI NO2?

Section 2.5.4: What about the techniques used in the SEARCH network?

Section 2.5.5.1: Should probably also include discussion of SO2 work from Fioletov et al., GRL, 2013, and others from NASA Goddard. There should also be a distinction between generating and using retrievals of SO2 columns, which aren't intrinsically tied to GEOS-Chem, and extrapolating these column concentrations to estimates of surface concentrations, as done in Lee et al. (2011) and Nowlan et al. (2014).

2-39, Fig 2-11: Later, in Section 3 (3-12, 37; 3-13, 8-10), it is noted that exposure to HNO3 may have been a driver of declines in lichen in the LA basin, and may be continuing to have an impact. I believe this is the only direct gas-phase impact of S or N species referenced to believed to be occurring in ambient conditions in recent times in all of section 3. The cited studies of HNO3 impacts on lichen were referenced to daily peak exposure. Would it be useful then to present estimates of daily peak HNO3 values, rather than / in addition to annual averages?

2-40, 7-12: I believe that paper only identifies a 5% /yr trend in December. Another possible reason discussed was increasing domestic NH3.

Section 2, figures: Scanning across the figures of species distributions in this section leads to a misleading first impression as to the spatial variability of different compounds owing to some seemingly arbitrary choices of color-bar scales. For example, the distribution of SO2 concentrations looks quite flat, as the max value plotted is ~6 ug/m3 on a scale up to 20. In contrast, nitogen oxide concentrations are plotted on a much better scale (Fig 2-9), such that gradients associated with particular sources (cities, power plants, and roads) are visible. Can all of the figures in this section be adjusted so that the max color-bar scale matches the max value on the map?

2-43, Section 2.6.6: Would it be worth providing information on current levels of 3hr max SO2 concentrations, as that is what the NAAQS are based on, instead of / in addition to the 3 year annual mean?

2-52, 19: remove comma: models, typically

2-54, 35: I wasn't sure about the claim that aerosol nitrate is found "mainly" in the coase mode in the Eastern US. Could this be clarified, and referenced? Are they referring to nitrate uptake on dust, or sea salt?

2-55, Fig 2-18: The axis labels are nearly illegible – could these be remade?

2-57, 4-6: Well, yes, that is bound to happen since the unidirectional model doesn't include the upward flux.

2-59, 31: Is there any large-scale evidence of this type of NO2 emission from e.g. SOAS/SEACRS? As found in the BEARPEX study mentioned on the next page, much of this may be converted to other species before it exists the canopy.

2-66, 3: The statement that N dep is overall in reduced form – is that based on the total budget of reduced N dep to oxidized N dep? Or does that mean there is a greater area where the former dominates? Is organic N accounted for in this statement? I thought (e.g., Zhang et al., ACP, 2012) that oxidized N dep was larger than reduced N dep in the US under current conditions, although that is expected to change in the near future as NOx emissions decrease and NH3 remain the same or increase (e.g., Ellis et al., ACP, 2013; Paulot et al., ES&T, 2013).

2-70, 9: This result from Krotkov 2016 was just over the Eastern US (see their Fig. 1). But the decreases in NO2 are generally supported by evidence from remote sensing, reported in Krotkov as well as several other studies cited therein (Duncan et al., 2013; Lamsal et al., 2015; Lu et al., 2015; Russell et al., 2012; Tong et al., 2015).

2-70, 16: This is also owing to reduced levels of sulfate, and hence less NHx present as ammonium sulfate.

2-72, 27-32: Didn't they also suggest increasing NH3 may play a role?

2-75, 1-2: A little redundant with the text at the end of page 2-72.

2-74: Fig 2-29, 2-30 and 2-31 -- any comment on the changes at the tip of Florida?

Section 2.9, 2-80…: This section delves into the details of a study by Koo et al. (2012) documenting variability across two models for transference ratios. Transference ratios are the inverse of atmospheric lifetimes, weighted by the ratio of the volume over which the concentration is calculated to the area over which the flux is calculated, ie the height of the system. Presumably since the latter isn't a variable quantity across studies, variability in transference ratios is just variability in the lifetime of NOy, SOx, or NHx. And then I tend to think that variability in lifetime has been more widely studied across models.

Section 2.10: I think there should be an additional paragraph or two in Section 2.10 (or an entire section) devoted to source-receptor estimates for N dep within the US. Lee et al. (ACP, 2016) evaluated the sources driving deposition of reactive N in several Federal Class I areas in the US. This study quantified

the natural contribution, as well as provided maps of the footprints for each location. Locations such as Big Bend had substantial contributions from soil NOx (~20%) as well as foreign emissions. Another important point of this work was the domestic, yet long-range influence of some anthropogenic emissions on N deposition in remote parks such as Rocky Mountain, which was impacted by NOx and NH3 emissions from CA. This type of long-range influence (on deposition in RMNP in particular) was also previously identified in works such as Paulot et al. (ES&T, 2013), Benedict et al. (JGR, 2013) Malm et al. (JAWM, 2013), and Thompson et al. (JGR, 2015).

2-42, Section 2.6.4: Increasing trends in NH3 are mentioned in several other locations throughout the report (e.g., 2-87, 1) – have these trends been detected with measurements from AMoN or SEARCH?

General: It struck me as a bit odd that there is a lot of discussion of CLs, but for the section on the distributions and trends in actual deposition values, the latter were presented with little reference to their magnitudes in relationship to CLs. Works such as Ellis et al. (ACP, 2013) and Lee et al. (ACP, 2016) broadly examine drivers of deposition above CLs, as well as others. Should this be discussed?

General: There were some pretty strong statements about the importance of bidirectional exchange, in a few locations. However, no results were shown for how bidirectional exchange impacts model estimates of concentrations or deposition. Could/should these be included?

2-90, 5-17: It may be good to also reiterate the results of Kim (2015) here as well, in terms of interactions between organics and inorganics impacting aerosol neutralization.

2-87, 18-26: As with the main body, the summary should be updated to reflect uncertainties in NH3 emissions.

<u>3. Gas phase effects</u>

3-4, 9-15: My sense is that this is (rarely) exceeded – can this be concluded based on CMAQ estimates and measurements discussed in Section 2? Figure 2-15 could be referenced, although it only shows the 3 year mean.

3-8, 6: Extra period.

3-8, 5-8: Could also refer to Fig 2-10 of this assessment.

3-11, 33: Could also refer to Fig 2-11 for estimates of typical annual ambient HNO3 concentrations

<u>4. Soil Biogeochemistry</u>

4-2, 12-20: There are many additional studies on the sources giving rise to N deposition beyond the one work cited here, at the regional (Benedict et al. (JGR, 2013; Malm et al., JAWM, 2013; Thompson et al., JGR, 2015), national (Zhang et al., ACP, 2012; Lee et al., ACP, 2016), and international (Paulot et al., ES&T, 2013) scales.

5. Terrestrial Acidification

6. Terrestrial Nitrogen Enrichment

7. Aquatic Biogeochemistry

8. Freshwater Acidification

9. Freshwater Nitrogen Enrichment

10. Estuarine and Near Coastal Nitrogen Enrichment

Fig 10-4: Can obtain higher resolution version? This one is blurry.

11. Wetland Nitrogen Enrichment

12. Sulfur Enrichment

13. Climate Modification of Ecosystem Response

13-2, Fig 13-1: It wasn't readily obvious to me from the figure and caption what the significance of the + and – symbols are.

13-8, 20: Placement of the citation is a bit odd.

I didn't have much to comment on this section, other than to wonder if there shouldn't be a counterpart in e.g. Ch 2 describing the impacts of climate change on the distributions of N and S inputs to ecosystems. This is only briefly touched on in Ch 13 (13-3, 2-7).

14. Ecosystem Services

15. Appendices

Dr. William McDowell

Initial comments on Ch. 12.

This chapter presents a detailed overview of the effects of sulfur enrichment. The accuracy, level of detail, and relevance of information presented on the biological effects of sulfide phytotoxicity and the mechanisms linking sulfur and mercury methylation. What is lacking, however, is an overview that links the cause-effect chains that are embedded in the detailed analysis.

There is some redundancy in the sections, and the topics addressed therein.
I suggest the following reorganization:
12.1 Introduction and causal statements
12.2 Effects of sulfur deposition on the sulfur cycle as written
12.3 Interactions between S deposition and Hg
- 12.3.1 Hg cycle and the importance of methylation
- 12.3.2 Sulfate-reducing prokaryote biology and genetics
- 12.3.3 Drivers of Hg methylation potential: physiological response of SRPs and rates of methylation to various drivers such as DOC, pH, etc
- 12.3.4 Deducing the strength of drivers under field conditions from whole-system field experiments
- 12.3.5 Inferring the strength of drivers from assessment of patterns in drivers and MeHg under ambient conditions

Dr. Erik Nelson

May 15, 2017

Erik Nelson's comments on *Chapter 13: Climate Modification*

1. For nonagricultural terrestrial ecosystems (pages 13-3 – 13-4):

    - What is the expected impact of N addition on soil C sequestration in areas that become wetter? Will soil C sequestration rates increase or decrease as N increases?
    - What is the expected impact of N addition on soil C sequestration in areas that become drier? Will soil C sequestration rates increase or decrease as N increases?
    - What is the expected impact of N addition on plant net primary productivity (NPP) in areas that become wetter?
    - What is the expected impact of N addition on plant net primary productivity (NPP) in areas that become drier?
    - In what regions of the country could we expect to see "bursts" of nitrate concentration in rivers, streams, and lakes? What will community water systems need to do to deal with these bursts?

2. Why no discussion on "Nitrogen Cycling: Transport and Transformation" <u>in ecosystems dominated by monoculture agriculture</u> (e.g., Iowa)? It appears that ecosystems dominated by agriculture is a system that Greaver et al. (2016) do not cover.
However, these effects need to quantified if we are to get a fuller picture of the welfare impacts of N and S addition to the US landscape. How will N and S addition in conjunction climate change affect crop growth and the non-point water pollution issues that are bedeviling the Midwest?

3. On page 13-5 we have the following: "This additional growth increases the overall amount of C stored in plant biomass; one unit of N input may cause an additional 24.5 to 177 units of forest C uptake." I am trying to get a handle on this magnitude of this effect. It would be great if the rate of additional forest C uptake was placed in some sort of context. For example, how much additional N is deposited across an acre of US forest every year? Is this effect large enough that if it were not explicitly included in terrestrial carbon models then model output would be significantly off?

4. Similarly, is the finding that "Belowground, initial findings are that N addition tends to increase the C stored in the soil organic layer and in root biomass…" (13-7) something that terrestrial C sequestration models need to explicitly incorporate or are the changes in soil C due to additional N so small that they will get lost in the noise of the sequestration models?

5. On page 13-6 we have the following: "Our synthesis of existing meta-analyses indicates that aboveground NPP is highly responsive to N addition and enhanced precipitation, <u>while</u>

temperature rise does not increase aboveground NPP." I am surprised by the underlined passage.

6. A little more background on "the ameliorative effect of enhanced weathering" (line 21, page 13-10) would be helpful.

7. *Nitrogen, Climate, and Biodiversity*. This sentence does not make sense to me: "In terrestrial systems that get warmer and wetter, eutrophication may be amplified if endogenous N sources are low or dampened if endogenous sources are high/more liberated to meet community demand." (lines 24 -26, page 13-11). How can eutrophication be dampened if more N is added to the system?

8. A table or map that summarizes species / assemblages that are likely to be negatively impacted by added N and climate change would be helpful.

May 16, 2017

Erik Nelson's comments on *Chapter 14: Ecosystem Services*

1. The cataloging of the various ecosystem service frameworks is unnecessary (pages 14-1 – 14-2). Just choose a framework and go from there (you can cite the alternative frameworks).

2. For the literature review on page 14-3, lines 8 – 29 also see the following papers:

    a. "Damages and Expected Deaths Due to Excess NOx Emissions from 2009-2015 Volkswagen Diesel Vehicles." S.P. Holland, E.T. Mansur, N.Z. Muller, A.J. Yates. Environmental Science and Technology. 50(3): 1111-1117. DOI 10.1021/acs.est.5b05190.

    "We use the AP2 air pollution integrated assessment model to connect excess ground-level NOx emissions to changes in ambient concentration of pollutants and associated deaths and monetary damages. The main component of damages is human mortality, but AP2 also accounts for morbidity, crop and timber yields, degradation of buildings and material, and reduced visibility and recreation. For human mortality, AP2 uses results from Pope et al. to specify the effect of PM 2.5 exposure on adult mortality rates and results from Bell et al. to specify the effect of O3 exposure on all-age mortality rates."

    "We find 46 excess deaths and $430 million in excess damages from driving all the vehicles with defeat devices from initial sale through 2015. These numbers

are approximately 25 times their allowed counterparts. Approximately 88% of damages are due to PM 2.5.The remaining damages result from exposure to ambient O3."

"Next, two additional scenarios assess assumptions in the AP2 model. The alternative dose response scenario uses the dose response for PM 2.5 given in Lepeule et al. This plausible alternative assumption doubles our estimates. The alternative VSL scenario uses a $2 million Value of a Statistical Life (VSL) rather than the standard $6 million VSL employed by the EPA (both expressed in year 2000 dollars). The $2 million VSL is based on a meta-analysis of Mrozek and Taylor that focused entirely on revealed preference studies. Using it lowers the damage estimate considerably."

b. 2016 "Air pollution emissions and damages from energy production in the U.S.: 2002-2011." P. Jaramillo and N.Z. Muller. Energy Policy 90: 202 - 211.

This paper uses the AP2 air pollution integrated assessment model as well.

c. Chan, H. Ron, et al. The Market for Sulfur Dioxide Allowances: What Have We Learned from the Grand Policy Experiment? No. w21383. National Bureau of Economic Research, 2015.

3. In Section 14.1.1 a table that summarizes the ecosystem service damage estimates from additional N would be more helpful then the two figures. This table could break down damage estimates by region and service. Further, it is not clear to me which studies separately quantify ecosystem service damages from the atmospheric portion of additional N loading. The proposed summary table could also separate ecosystem service damages due to the atmospheric portion of additional N loading from the damages due to the terrestrial portion of N loading.

4. There are a slew of papers on the projected impact of ocean acidification on fisheries. A Google Scholar search on "the economic impact of ocean acidification US" will turn them up. A table that summarizes these estimates for US fisheries would be great. (Examples: Mabardy, Rebecca A., et al. "Perception and response of the US West Coast shellfish industry to ocean acidification: the voice of the canaries in the coal mine." Journal of Shellfish Research 34.2 (2015): 565-572).

5. Overall this chapter needs to do a better job of delineating estimated ecosystem service damages due to additional atmospheric N versus estimated ecosystem service damages due to additional terrestrial N. It aloes has to do a better job of displaying the range in damage estimated from additional N.

6. It would also be nice to know how much less additional N would be added to ecosystems if the more stringent Secondary NAAQS standards for Oxides of Nitrogen and Sulfur were put into place.

May 17, 2017

Erik Nelson's comments on *Case Studies*

Northeast case study

1. There is so much data here. It is hard to digest it all. The tables in this section are especially opaque. The tables give me no insight into what is happening in the Northeast; they are hard to interpret. The Tables have too much data and dat that is not organized in a user-friendly manner.
1. My read of Figure C-5.B (p. C-14) is that is that wet deposition of the various chemicals has fallen over time in HBEF. Why this downward trend? Can we extrapolate the trends in wet deposition with and without stricter regulations?
2. "In general, wet deposition typically exceeds dry deposition of N and S in this case study area." (p. C-15). Why? Why does this matter?
3. West Bear Brook is a controlled experiment on the ecological impact of increased N deposition. Does that suggest that it should be featured in an expanded case study? This experiment would seem to represent one of our greatest opportunities for isolating the impact of increased N deposition on ecological functions.
4. Our role is to give a recommendation on whether a stricter ambient air quality standard is needed. Of all of the data presented in Appendix C.1 section C.1.5 would seem to be the most relevant to our decision.

Southeastern Appalachia case study

1. This passage seems important: "The capacity of these compartments for storing N and S are not infinite, and the capacity to retain N has been exceeded in some areas (ecosystems have reached N saturation), as evidenced by increased nitrate leaching and acidification of some surface waters. This suggests that ecosystems are impaired by N deposition, and that there may be a lag time in ecosystem recovery if deposition is reduced (Fakhraei et al., 2016; Cai et al., 2011a)." (C-46, lines 15-20).
2. Figures C-9 and C-10. Can we extrapolate the trends in wet deposition with and without stricter regs?
3. Note this passage: "This quantity is in agreement with an earlier recommendation for Class I wildernesses in the Forest Service's eastern region, that N deposition should not exceed 5−8 kg N/ha/yr to protect terrestrial biota (Adams et al., 1991). That report also recommended an annual load for total sulfur deposition not to exceed 5−7 kg S/ha/yr to protect terrestrial biota (Adams et al., 1991). The critical loads set by Adams et al. (1991) were based on observations across class I wildernesses in the Forest Service eastern region; the deposition limits were based on the deposition loads of three Class I areas where eutrophication and acidification were not observed." (C-56, lines 1-12). Where are we on this recommendation? Would stricter regulations get us closer to this goal?

Tampa Bay case study

1. Again we see that the wet deposition of the various chemicals has fallen over time (Figure C-15). Can we extrapolate the trends in wet deposition with and without stricter regulations?
2. Lines 9 -13, page C-69: "According to BRACE, reductions in N emissions from mobile sources must be a part of the strategy to reduce N loading to the Bay, and that control of atmospheric N emissions both within and outside the Tampa Bay watershed is important to restore and maintain good water quality and a healthy ecosystem within the bay." How would a change in NAAQS regulations change the N emissions from mobile sources?
3. So this case study highlights a great success. How can we use the success of Tampa to inform our decision?

Rocky Mountain and California case studies

1. Unlike the eastern case studies wet deposition has not fallen in the west. Why?
2. What as a committee are we supposed to do with these case studies? How do they help us make a decision?

Dr. Hans Paerl

Hans Paerl comments on EPA/600/R-16/372

Chapter 7

Fig. 7.1 on P. 588: There's no mention of denitrification in this figure, even though it depicts the N cycle.

P. 595, lines 10-12, add reference to support statement: Paerl, H. W., J.T. Scott, M.J. McCarthy, S.E. Newell, W.S. Gardner, K.E. Havens, D.K. Hoffman, S.W. Wilhelm and W.A. Wurtsbaugh. 2016. It takes two to tango: When and where dual nutrient (N & P) reductions are needed to protect lakes and downstream ecosystems. Environmental Science & Technology 50: 10805−10813.

Chapter 9

P. 733, lines 24-26, it is stated: "N limitation appears to be increasingly common in freshwater systems, probably because their nutrient dynamics are being altered significantly by growing agricultural and urban P inputs" Please add the following recent reference:
Paerl, H. W., J.T. Scott, M.J. McCarthy, S.E. Newell, W.S. Gardner, K.E. Havens, D.K. Hoffman, S.W. Wilhelm and W.A. Wurtsbaugh. 2016. It takes two to tango: When and where dual nutrient (N & P) reductions are needed to protect lakes and downstream ecosystems. Environmental Science & Technology 50: 10805−10813.

P. 734, Figure 9.2: Denitrification is missing in this figure of the N cycle.

P. 735, lines 7-9: It is stated that "In highly productive freshwaters, nutrient enrichment from N deposition usually does not stimulate primary productivity or community change because P is more commonly the limiting nutrient."
This is not necessarily true. N limitation tends to be more prevalent in highly eutrophic lakes:
See Elser 2007 (cited in the report).
Also: Paerl, H. W., J.T. Scott, M.J. McCarthy, S.E. Newell, W.S. Gardner, K.E. Havens, D.K. Hoffman, S.W. Wilhelm and W.A. Wurtsbaugh. 2016. It takes two to tango: When and where dual nutrient (N & P) reductions are needed to protect lakes and downstream ecosystems. Environmental Science & Technology 50: 10805−10813.

P. 735, lines 28-29: Add: Paerl, H. W., J.T. Scott, M.J. McCarthy, S.E. Newell, W.S. Gardner, K.E. Havens, D.K. Hoffman, S.W. Wilhelm and W.A. Wurtsbaugh. 2016. It takes two to tango: When and where dual nutrient (N & P) reductions are needed to protect lakes and downstream ecosystems. Environmental Science & Technology 50: 10805−10813.

P. 738, lines 10-14: It is stated that "This pattern of increasing P limitation with increasing N deposition in the Adirondack sub-region was also evident in foliar N:P ratio from seven tree species examined in the same study. Tree and lake data did not support a transition from N toward P limitation for the entire

five-state region, but that shift was supported for the Adirondack sub-region." We need to be careful here. Inferring nutrient limitation from N:P ratios can be tricky, especially if one or both nutrients are saturating.

This comment also applies to lines 19-21

Chapter 10

P. 776, lines 3-6: It is stated that, "Increased production of CO2 from degradation of organic matter associated with eutrophication along with atmospheric anthropogenic CO2 inputs can result in formation of carbonic acid and make the ocean water more acidic."
This is still quite speculative, especially the degradation of organic matter part. Eutrophication will also lead to enhanced uptake of CO2 due to higher rates of primary production. How can you have more CO2 released by mineralization than is produced by what is fixed due to eutrophication?

P. 777, Figure 10-1: Delete "concentration of available nutrients" and substitute "availability of nutrients".

P. 778, line 4: Insert "excessive" before "N".

P. 778, lines 26-28: It is stated that "atmospheric deposition of reduced N has increased relative to oxidized N and this trend is expected to continue in the future under existing emission controls (Pinder et al., 2008; U.S. EPA, 2008a)." The increase in atmospheric reduced N is a troubling trend, as this form of N is highly bioreactive and often a preferred source of N for phytoplankton, including harmful species (Glibert et al., 2016).

Glibert, P., F. Wilkerson, R. Dugdale, J. Raven, C. Dupont, P. Leavitt, A. Parker, J. Burkholder and T. Kana. 2016. Pluses and minuses of ammonium and nitrate uptake and assimilation by phytoplankton and implications for productivity and community composition, with emphasis on nitrogen-enriched conditions. *Limnology and Oceanography* 61:165-197.

P. 779, line 4: Substitute "a significant fraction" for "much of".
line 5: Insert "biologically-available" before "N".

p. 780, Line 5: Insert "the" before "nutrient balance".
Line 10: Substitute "in the lower bay and sound regions" for "on the outer bay".
Line 18: Substitute "input" for "management".
Line 24: Insert "downstream" before "distances".
Line 26: Substitute "estuarine" for "estuaries".

P. 782, line 31: Substitute "overriding role in controlling" for "more important role".

Table 10-1: In the indicator "dissolved oxygen", It should be noted that oxygen depletion mainly occurs in bottom waters under stratified conditions. In surface waters, oxygen levels can actually be quite high because algal blooms are photosynthetically producing it (often at supersaturating levels).

In the SAV part of the table, it is stated that "distribution of SAV in Chesapeake Bay is used as an indicator in the *EPA Report on the Environment* (U.S. EPA, 2016f)." "indicator" of what?

P. 785, figure 10-3: Note that oxygen depletion occurs in subsurface waters (mainly if they are stratified), not surface waters, as mentioned above.

P. 785, line 3: Insert "often" before "directly".
Line 10: Insert "(i.e. freshwater inputs and tidal flushing)" before "in".

P. 786, line 9: Substitute "elevated" for "high".

P. 787, lines 30-31: It is stated that "The authors suggested that this results from uptake of ambient NO3− by phytoplankton enabled by increased NH4+ loads." This sentence doesn't make sense. Overall, SFO Bay is not a very good example of N-driven eutrophication because of its short residence time (high flushing rates) as well as high turbidity (interactive effects of light limitation). Paerl and Piehler (2008) (Fig. 11) show definitively the effects of N enrichment on Chl a stimulation in the Neuse River Estuary, which is a microtidal (long residence time) system in which nutrient limitation/stimulation can be readily detected. This would be a better example.
Paerl, H.W. and M.F. Piehler. 2008. Nitrogen and Marine Eutrophication. pp 529-567, In, D.G, Capone, M. Mulholland and E. Carpenter (Eds.), Nitrogen in the Marine Environment, Vol. 2. Academic Press, Orlando.

P. 789, line 2: Substitute "increasingly" for "increasing".
Lines 9-10: Substitute "at maximum levels" for "the highest".
Line 15: Change to "nutrient-impaired lakes and rivers discharging to the bay".
Line 19: Insert "dinoflagellate" before "HAB"
Line 21: Insert "cyanobacterial" before "HAB"
Line 23: Delete "not NO3"

Table 10-3: Under Maryland and Virginia Coastal Bays category, it is stated that "Virtually all of the N in the water column is now in the chemically reduced form, NH4+ or DON" I don't think this is true. There is also NO3 present.

(Table 10-3 Continued)

In the Ten Mile Creek/Indian River Lagoon..It is stated that "Chlorophyll *a* was negatively correlated with N concentrations. This result is thought to be due to the strong influence exerted by hydrologic factors (such as freshwater inflow, salinity, pH, and temperature), which were all positively correlated with Chl *a* concentrations during this study." The second sentence isn't clear. It seems to contradict the previous sentence.

As for the Nueces Estuary, Corpus Christi, just making conclusions based on N:P ratios as to Chl a conditions is too simplistic. There is probably light limitation interactions going on along with nutrient limitation in this highly turbid estuary.

In San Francisco Bay, the results of the N isotope study were not very definitive as to the actual source of the NH4 supporting growth of M*icrocystis* blooms. The NH4 could have come from regeneration of previously loaded N sources or from externally-supplied sources, such as wastewater effluent from the upstream wastewater treatment plants.
Also, in SF Bay, there is strong evidence that there are light-nutrient limitation interactions going on, which would control phytoplankton growth and bloom dynamics.

P. 795, lines 9-10, it is stated that "In other places such as lagoons with limited oceanic exchange, macroalgae may be a more sensitive biological indicator than phytoplankton." That's not true in other lagoonal estuarine systems such as the Albemarle-Pamlico and Laguna Madre, where phytoplankton are more sensitive indicators. it depends on clarity as well, because light limitation can interact with nutrient limitation to determine planktonic vs. benthic production.

P. 796, lines 9-10. NO3 toxicity in macrophytes??? This is highly speculative and I suggest omitting it unless there's direct, irrefutable evidence for this.
Lines 18-20, it is stated that "The decomposition of organic matter associated with increased algal abundance consumes DOand can reduce DOconcentrations in eutrophic waters to levels that cannot support aquatic life." It should be noted that DO depletion largely occurs only in bottom waters, under stratified conditions, not throughout the entire water column.

P. 797, lines 25-31, it is stated that "Effects of low DO appear to be exacerbated by presence of multiple stressors. For example, Gobler et al. (2014) examined concurrent effects of low DO and acidification on early lifestages of bay scallops (*Argopecten irradians*) and hard clams (*Mercenaria mercenaria*). Observations in later lifestages of clams indicated that growth rates decreased by 40% in combined exposures to hypoxia and acidification. Additional studies with earlier lifestages indicated effects were more severe with costressors than with either hypoxia or acidification alone." In low tidal, or semi-lagoonal and lagoonal systems, freshwater inflow runoff can also exacerbate hypoxia by enhancing vertical stratification. This is an important driver of hypoxia in some coastal systems as well, like the N. Gulf of Mexico receiving Mississippi R. inflow.

P. 806, lines 11-12, it is stated that "DIN was the best predictor and effects were observed at approximately 5 to 10 mg/km/yr inorganic N." Was DIN correlated at the same time with Chl a? If N was limiting, one would expect DIN to be inversely correlated with productivity and biomass. This may be a time-lagged correlation
Line 19, insert "supplied" before "can".

P. 808, line 2, Insert "at least" before "one".

P. 818, **10.5 Nutrient Enhanced Coastal Acidification**
The effects of CO2 enrichment are a two-way street. Primary productivity is enhanced as is the formation of organic matter. There can't be any more organic matter decomposed than is formed by nutrient-enhanced primary production (eutrophication), so productivity is limiting the exchange of CO2. Assuming there will also be some burial of organic matter, one can make the assumption that there actually net CO2 removal from the atmosphere from coastal eutrophication processes, rather than net CO2 release back to the atmosphere.

P. 819, Figure 10-8: There should be an arrow from atmospheric CO2 to "Algal blooms from nutrient inputs".

P. 820, lines 3-13. The same comment as given above (on P. 818) applies to this paragraph.

P. 826, lines 4-6: It is stated that "**the body of evidence is sufficient to infer a causal relationship between N deposition and changes in biota including altered growth, species richness, community composition, and biodiversity due to N enrichment in estuarine environments. "** The causal relationship should also include the effects on total primary production and total algal community biomass.
Lines 15-20: It is stated that "The ratio of reduced-to-oxidized N deposition has shifted toward increased NH4+ relative to NO3− in coastal areas especially in the eastern U.S. Large diatoms are more efficient in using NO3− than NH4+, and the increased NH4+ relative to NO3− in the eastern U.S. favors small diatoms (Paerl et al., 2000; Stolte et al., 1994). This alters the foundation of the food web. Some newer studies support these observations of NO3− and NH4+ and diatom species distribution (Heil et al., 2007)." I don't agree with this statement and it is a misquote of the Paerl et al. (2000) reference. That reference never concluded that large diatoms are more efficient in using NO3 than NH4. What's more important to mention here is that there is preference for NH4 over NO3 and this could lead to selective stimulation of primary production, especially in light-limited estuarine and coastal waters.

Summary/Comments on ISA for Oxides of Nitrogen, Oxides of Sulfur, and Particulate Matter—Ecological Criteria, Chapter 11: Nitrogen Eutrophication Effects in Wetlands

Frank S. Gilliam

Hans Paerl

Chapter 11—Nitrogen Eutrophication Effects in Wetlands—reviews recent literature on the effects of excess nitrogen (N) on a wide variety of wetland ecosystems. Not only do the authors of this chapter use wetland classification as established in relevant literature, but they also provide a useful, informative summary table to distinguish among numerous wetland types based on several classification criteria, including soil-based, hydrology-based, and soil/hydrology/vegetation-based classifications. This is a rather exhaustive list that includes more wetland types than is covered in the review. Those emphasized in the chapter are salt and freshwater marsh, mangrove, riparian wetland, and bog/fen. This is an extensive handling of literature, primarily since the 2008 ISA, and the findings regarding N-mediated effects on the biogeochemistry of wetlands is largely confirmatory of the results reported in the 2008 ISA, i.e., there is a causal relationship between N deposition and the alteration of

biogeochemical cycling in wetlands. This new ISA, however, adds to the body of knowledge regarding responses of wetlands to excess N by including other response criteria, e.g., plant physiology and plant architecture. The body of evidence is sufficient to infer a causal relationship between excess N and alteration ecophysiology, species richness, community composition, and biodiversity in wetlands.

As an aside, it is easy for an ecologist, such as myself, with an east coast bias to think of coastal wetlands as predominantly an east coast phenomenon, and that would be incorrect. Thus, the authors of this chapter are to be commended for their even handling of the general topic of wetlands and their biogeochemical and plant community responses to excess N, wherein west coast references from California to Washington State are reviewed, along with east coast references from Florida to Maine, and along the Gulf Coast from Florida to Texas.

Studies reviewed regarding biogeochemical responses of widely contrasting wetland ecosystems to excess N generally involved experimental additions of a wide variety of forms of N, from NH_4NO_3 to urea and even sewage sludge, and at widely varying amounts, in many cases well in excess of 1000 kg N/ha/yr. Most response variables were N mineralization, nitrification, and denitrification, but also soils microbial communities, especially the specialized bacterial groups associated with nitrification and denitrification. Effects of N on soil carbon (C) cycling in wetlands were minimally addressed, primarily because a meta-analysis (which included wetlands, among other non-forested ecosystems) failed to find any effects of N on net ecosystem exchange of carbon. However, in support of findings of the 2008 ISA, a separate meta-analysis revealed that experimental additions of N can increase methane (CH_4)—an important greenhouse gas—by nearly 100%

Regarding plant response variables, quite numerous studies were reviewed that examined the effects of N (often combined in several studies with effects of CO_2) on plant production and biomass. Results of these studies were understandably varied, considering the widely contrasting wetland types and forms/amounts of added N, along with whether experimental treatments included CO_2. New criteria in this ISA not included in the 2008 ISA included plant stoichiometry and ecophysiology. Because stoichiometry examines the balance of nutrients in organisms, and because excess N can alter greatly the availability and uptake of essential plant nutrients, this is an important characteristic for consideration. As before, there were widely varying results among the numerous studies reviewed.

Other plant response variables include plant architecture, demography (including reproduction and mortality of individual wetland species), and biodiversity. The latter is especially relevant given that high biodiversity is characteristic of most wetland ecosystems. Once again, given the numerous

wetland types represented, along with contrasting treatments, results varied greatly among studies. Finally, critical loads of N for wetlands were considered. At the time of the 2008 ISA, such critical loads had either not been determined or not been published for wetlands. Since that time, however, critical loads have been published, generally varying between 3 and 13 kg N/ha/yr.

As has already been suggested, the authors of this chapter are to be commended for their handling of this broad, challenging topic. The chapter seems well-written and comprehensive with regard to references and background information on these various ecosystem types. Indeed, the number of studies reviewed ranges from extensive to nearly exhaustive. We see no need for further changes.

Mr. Richard Poirot

Comments on 1st Draft ISA on SOx, NOx & PM – Eco. Criteria, R. Poirot

Executive Summary and Chapter 1

a. Please comment on the extent to which the executive Summary and Chapter 1 meet their objectives.

The Executive Summary and Integrated Synthesis both do a good job of meeting their intended objectives. While both are lengthy, the subject area is complex, and I can't think of suggested sections to shorten or leave out. The writing is clear and consistent in style and detail (does not appear to have been drafted by too many different authors as is sometimes the case with such complex reviews). An objective of such summaries is to leave out as many caveats and qualifiers as possible and still tell the truth. Again, I think these sections do this very well.

I think the Integrated Synthesis does a very good job of identifying and explaining the concept of critical loads. However, these CL are often derived for very specific biological or chemical endpoints. But ecosystems don't often respond to anything with a high degree of numerical precision. Rather, there is likely to be a broad gradient of effects that result from widespread reductions of SOx, NOx (or NHx) emissions that might be employed to achieve specific CL endpoints for individual species. A bit more description of this kind of shifts in gradients of effects, could be a useful complement to the CL discussions. A related point would be to add some discussion of how/if various CLs might relate to chemical or biological recovery, or how recovery times might be influenced by the magnitudes of emissions reductions.

b. Please comment on the extent to which the causality framework is appropriately applied to evidence for each of the effects categories in chapters 3-12 to form causal determinations.

I think the causality framework is appropriately applied and clearly communicated throughout these sections. The use of summary tables that show if and how causality determinations have changed since the last review cycle – along with links to the more detailed technical chapters – is very helpful.

One minor point is that while many/most of the causality statements relate to effects associated with current concentrations/ depositions, a causal association is identified for phytotoxic effects from exposures to gas-phase SOx & NOx compounds, but additional statements indicate that this does likely not occur at current ambient concentrations. Then some examples are provided that seems to indicate there are current effects. It would be helpful to clarify this – especially since the basis for current secondary standards is from direct exposures to gas phase compounds

p. lxii, lines 13-20: This summary of SOx and NOx emissions trends seems either out of place here or incomplete. Here or elsewhere it would be useful to add associated points such as:

- Ambient air concentrations and deposition of SOx & NOx have also declined proportionately, while emissions, concentrations & deposition of NHx have increased.
- Ecological effects of SOx and NOx deposition represent responses to both current and (much higher) historical levels of deposition.
- Much higher historical levels of SOx and NOx deposition have altered the manner in which and degree to which ecosystems may respond to continued current or changed future levels of SOx & NOx (& NHx) deposition. Etc.

p. 1-9, Table 1-1: Here and elsewhere, these 'causal determination' tables are a very effective, concise way of summarizing current state of scientific understanding, with emphasis on causal relationships which have been recently identified or for which the confidence of causality has substantially increased since the last ISA. (combined with useful pointers to the appropriate section of the ISA)!

p. 1-18, lines 12-17: While an "emphasis on identifying thresholds" may be true for health effects, it could also be noted that health effects have historically been observed at progressively lower concentrations as knowledge has advanced and as ambient concentrations have declined. Thresholds have not been identified for effects of PM and O_3 on mortality or neurological effects of Pb.

Here or elsewhere, it might be useful to include some cautionary comments on use of CLs. For example, CLs are generally estimated for specific sensitive elements of the environment, at assumed steady-state conditions, and are dated as "according to present knowledge". A gradient of ecological effects are likely to occur at deposition levels above and below CLs estimated for specific ecological endpoints.

p. 1-21, lines 26-27: This sentence needs a verb & object (has been observed, is anticipated, etc.). Also, I think it would be better to employ a less casual CL definition. For example, an oft-cited definition from the (1988) UNECE Workgroup on Nitrogen Oxides is "a quantitative estimate of an exposure to one or more pollutants below which significant harmful effects on specified sensitive elements of the environment do not occur according to present knowledge". (Nilsson, J. and P. Grennfelt. 1988. Critical loads for sulphur and nitrogen. UNECE/Nordic Council workshop report, Skokloster, Sweden. March 1988). The definition you offer here implies there are no harmful ecological effects on any ecosystem components if a critical load is attained.

p. 1-22, table 1-5: The "critical biological responses" listed here are rather severe. It seems likely that there are a range of adverse effects which can occur at deposition levels below which those that result in seedling death, reproductive failure and species loss. Perhaps this would be a good place to mention some limitations of CL.

p. 1-24, line 32, p 1-25, line 1 and lines 7-12: This causality statement differs from other similar causality statements in that it says "yes there is a causal relationship between exposure to these pollutants" but "No, there is no evidence of effects at current ambient concentrations". Most of the other causality statements include "under current conditions". Then there seems to be a bit of

conflicting evidence reported on **p. 1-26, lines 17-20** indicating that there are effects on lichens in LA resulting from current HNO_3 exposures.

p. 1-43, lines 20-23: This is an interesting observation and theoretical explanation: that areas with higher N deposition tend to exhibit higher N CLs – because historical N deposition has altered these ecosystems from their prior pre-deposition conditions. In this case, I suppose a given CL would be expected to assure that no additional damage would occur beyond that which has already occurred. Perhaps then there would be a second lower CL that would return the ecosystem to (or closer to) its original, healthier condition? A related question is – to what extent (if any) is ecological "recovery" expected to occur if a given CL is attained? The above example – where past deposition has resulted in a higher critical load seems to apply that there will not be recovery to or toward a past healthier condition, but only prevention of additional future damage.

p. 1-48, figure 1-7: Here or elsewhere (perhaps in glossary) it would be useful to show how S+N is expressed in eq/ha/yr – maybe with some conversion factors to other commonly employed deposition units. For example, here you use units of $meq/m^2/yr$ and elsewhere use eq/ha/yr. Could consistent units generally be employed throughout, or include a handy conversion table in glossary or sidebar.

p 1-50, line 1: I don't think chl *a* has previously been defined in the text.

p. 1-55, lines12-13 and 17-19: This "greater role than was previously understood" is a pretty vague statement. Can you be a bit more specific?

p. 1-61, figure 1-8 caption: Could you explain somewhere what "10th percentile aggregation" means.

p. 1-61, line 6: This is a pretty pessimistic blanket statement of total ignorance. Might change to something like "the timing and extent of biological recovery are highly variable among different species and ecosystems, and some systems may not ever return to pre-acidification conditions."

p. 162, lines 9-10: Here or elsewhere it would be useful to discuss how chemical and/or biological "recovery" (in whatever forms and with whatever lag times recovery might take) might relate to critical loads. As per previous comment on p. 143, is any degree of recovery expected if a CL is met, or is the CL only protective against additional future damage?

p. 1-72, lines 21-22: This definition "…retain <u>or flush</u> nutrients" doesn't sound right.

p. 1-83, lines 12-15: Is there any indication of whether this increasing DOC represents a return to pre-anthropogenic-acidification conditions – or is this is a new state? If increased DOC constrains ANC & pH recovery but reduces Al toxicity, what does that say about the chemical indicators as a basis for setting CLs?

p. 1-85, line 8: It would be helpful here to add a few lines explaining what ecosystem services are.

2. Source to Deposition

p. 211, lines 7-9: 25 to 50% of what?

p. 2-34, lines 20-23: IMPROVE also measures $pSO4^{2-}$ from the Module B nylon filter by IC. I think that in the past, IMPROVE used 3xS (from XRF on the Teflon filter) rather than directly measured SO_4^{2-} to calculate $(NH_4)_2SO_4$ in the IMPROVE algorithm. I think this may still be the case – to avoid having to change all the baseline and tracking progress data for the Regional Haze Rule – but I think the directly measured SO_4 is currently considered a better indicator of fine particle sulfate.

p. 2-35, lines 25-28: As Indicated in previous comment, I think S data were historically employed in the IMPROVE algorithm to calculate (assumed) ammonium sulfate. Since the Regional Haze Rule is trend-based, this use of S rather than SO_4 in this algorithm may still be employed to maintain continuity with the 2000-2004 "baseline" data. However, I think additional issues have been identified with current and historical S data, and that directly measured SO_4 is currently considered a better measure than S. See IMPROVE Data Advisory DA00023 (http://vista.cira.colostate.edu/improve/Data/QA_QC/Advisory/da0023/da0023_DA_SSO4_update.pdf).

p. 2-39, figure 2-11 caption (and in several subsequent figure captions): "Clear" should be "Clean".

p. 2-41, figure 2-13: Clean, not Clear. Are all AMON sites also CASTNet? There's a more recent 2014 version of this at: https://www3.epa.gov/castnet/docs/annual_report_2014.pdf

p. 2-44, figure 2-15 and other similar figures: Several of these figures use a scale apparently intended to show change from much higher previous levels. They would convey more useful information if plotted at lower scales that would show magnitude and spatial patterns of current concentrations.

p. 2-80, lines 26-28: Dry deposition fluxes aren't "measured" at CASTNET sites, but are estimated from concentrations and micro-met data. A separate issue is the representativeness of a point measurement to the grid cell represented in the models.

4. Soil Biogeochemistry

p 342, line 3: Here and elsewhere throughout the report when you refer to CLs from the Pardo et al., 2011 document the references to ecoregions are always to Omernick Level 1. It might be useful to add a comment somewhere on how these relate to the level III ecoregions which were featured in the AAI index employed in the previous NAAQS review.

14. Ecosystem Services

Please comment on the accuracy, clarity, level of detail, and relevance of ecosystem services frameworks and the effects of nitrogen inputs on ecosystem services.

Overall, this chapter provides a good summary of recent literature within the context of the 2008 state of the science. It is difficult, however, to get a sense of specifically how this new knowledge might be employed in the current 2ndary SOx/NOx review. Possibly a descriptive assessment of an expected shift in various ecosystem services from proportionate decreases (or increases) in SOx, NOx or NHx would make a good accompaniment to the more "bright line" threshold approach for meeting specific CL limits.

The ES profiles for select species are well done and informative. Although they reinforce concerns that a more comprehensive evaluation of total ecosystem services benefits from SOx/NOx reductions may not be currently feasible.

p. 998, line 9: It seems an omission to leave out the importance of the distinct balsam fir scent to the enjoyment of hiking & camping in northern forest parks & wilderness. Many hiking trail descriptions include reference to "the sweet scent of balsam fir..."

15. Appendices

a. Please comment on the adequacy of the information for the case studies and identify additional considerations, if any, relevant to evaluation of effects in these locations.

Overall, the information in the case studies is clearly presented, relevant and helpful. Exclusion of the (data dense) Adirondack area is reasonable, given the thorough coverage of that region in preceding chapters.

One minor point is that the Cleavitt et al., 2015 Lichen study mentioned in the Southeastern C2 case study included measurements from Acadia NP and Gt Gulf Wilderness (near Hubbard Brook) and is more relevant to and could be discussed in the C1 Northeastern case study. I also note that while the Northeast case study includes a section C.1.5 on "Recovery", there is no parallel "recovery" section in the Southeast case study. Granted recovery has been much less (or slower) in the SE, it could still be informative to show the relatively small or nonexistent chemical improvements for contrast. Maybe you could contrast with NE and illustrate with some regional TIME and LTM stream & lake chemistry trends.

I wonder if any of the case study areas have nearby CASTNET and/or low level SO2 & NOy monitors. If so, it would be interesting to see how well the TDEP deposition is reproduced by the combination of measured gas + aerosol species and Transfer functions – as proposed in the previous NAAQS review. If none of these case study areas have sufficient measurements, it would be informative to see those kind of calculations at locations with available data. Maybe an approximation of this could be done at all CASTNet sites using the sum of HNO_3 and pNO_3 instead of NOy.

b. Please comment on the adequacy of the characterization of non-nitrogen and non-sulfur particle associated components and their ecological effects.

This information seems like an adequate update on this narrowly focused topic – which includes only ecological effects (but excludes other welfare effects such as soiling, materials damage, visibility impairment and climate-forcing) from PM (including primarily non-SOx & non-NOx PM components). It also seems awkward to include since there's no associated "Source (to formation) to Deposition" discussion in Chapter 2 – except for S & N-containing particles. While awkward, it seems harmless to include it here – and would likewise seem harmless to include when the PM ISA is revised.

p 1142, line 22: Sea salt particles can also react with anthropogenic pollutants like HNO_3 - forming $NaNO_3$ and releasing Cl gas. Relatively large particle $NaNO_3$ can be an important contributor to N deposition in polluted coastal locations. Soil particles – of natural origin (well, sort of) - also react with anthropogenic precursors like SOx, NOx & NHx.

p 1183, lines 30-33: These 2 statements seem contradictory. Diffuse radiation is also diminished in intensity, as some light is scattered back to space.

p 1186, lines 15-16: And formation rates of secondary organic aerosols from biogenic VOC can be substantially enhanced by anthropogenic pollutants including acidic aerosols, metals, NOx, etc.

Dr. Armistead (Ted) Russell

NOx-SOx Secondary ISA Review

Armistead Russell

The Integrated Science Assessment (ISA) for Oxides of Nitrogen, Oxides of Sulfur and Particulate Matter – Ecological Criteria, is an extensive review of the science that is important to assessing how the subject pollutants impact ecological endpoints of concern. While the ISA is extensive, and the EPA staff and others who contributed to the document are congratulated for putting it together, it does have some flaws and gaps as discussed below. Also, given the length of the document, I read it in chunks and may miss-state things in the review that got lost from picking the document up and putting it down so many times, so sorry in advance. There is a whole lot of information here. Given my assignment, I have focused more on the Executive Summary and Chapters 1 and 2, as well as the appendices, but the comments reflect other parts of the ISA as well.

The document is large, and there are areas where it is unnecessarily so. One that comes to mind is the sections on meteorology and depositions. While both are important topics, the level of detail should fit needs of the document. Oh, I noticed the ISA no longer leads off as being concise, which is fine given its size. On the other hand, the idea is that the ISA is supposed to be as concise as appropriate. I would suggest an edited version strive to be more concise.

Most of the document is written in relatively neutral, scientifically-appropriate, style. However, there were a few cases when the document strayed. For example, in the Executive Summary, page lxiii, starting at line 30, presents an overly alarming paragraph, much of it unneeded for an ISA As similar paragraph is found in the Introduction, but is not really driven by any of the following chapters). For one, it says "It is clear … declines in biodiversity…". The determinations in Table ES-1 refer to "alterations" not "declines". It is not "clear". Further, , where does the ISA support that a defining attribute of the Anthropocene is global, human-driven, mass extinctions? (Of interest, the Smithsonian, http://www.smithsonianmag.com/science-nature/what-is-the-anthropocene-and-are-we-in-it-164801414/#YdZiMUWAtJ4V8cOS.99, says "According to the International Union of Geological Sciences (IUGS), the professional organization in charge of defining Earth's time scale, we are officially in the Holocene ("entirely recent") epoch, which began 11,700 years ago after the last major ice age." From Wikipedia "As of August 2016, neither the International Commission on Stratigraphy nor the International Union of Geological Sciences has yet officially approved the term as a recognized subdivision of geological time,[3][5][6] although the Working Group on the Anthropocene (WGA) voted to formally designate the epoch Anthropocene and presented the recommendation to the International Geological Congress on 29 August 2016." (I did not see that either body has done so at present, either.) While the paragraph in the Introduction chapter does cite Rockman et al., this paragraph is a bit more alarming than how I read the cited manuscript. Rockman et al. cite the Anthropocene as the era "in which human actions have become the main driver of global environmental change". This is a better characterization. Another term that is used is "richness", which presumably means number of species present, or do they mean diversity, indicating a balance? Richness also can have a more

subjective inference, i.e., that having more species present is good, while what is probably good is diversity. As they note, Salt Lake is not very biodiverse, but that is normal. Generally, every effort should be made to make the ISA void of what might be perceived as being biased in one direction or another. Also, try not to use absolute terms (e.g., "must") unless the absolute is quite absolute. For example, 1-13;28 has "must be provided by regional models … in conjunction with satellites". Not sure must is correct, and certainly they do not absolutely need satellite data. Using the two together may be the best way we currently have, but is it a "must"?

The Aquatic Acidification Index (AAI) was a scientific construct developed as part of the 2008 review. This needs to be further discussed as part of the ISA as to its weaknesses and utility, and the reasoning for EPA's decision to go forward, or not, with this construct. The IRP does not provide such a discussion. It is important that the panel, early on, is fully informed as to the potential constructs of metrics (indicators) that may be used for setting a secondary standard.

All of the chapters could use a synthesis section at the end, linking the chapter to how the topics fit together, and further how they fit in to the overall picture. This is driven by the overall length of the chapters and document, and that it is not always apparent from how the chapters are structured and presented how the information fits together. This is potentially followed by a summary, but the synthesis is the more important of the two. Yes, I know this potentially adds length, but maybe aspects of the chapters can be trimmed down if a good synthesis is developed. The ISA itself, could also use a synthesis section, which is different from an executive summary. (It is interesting to note that the website says about the ISA: "This draft ISA document represents a concise synthesis…")

<u>Chapter 1.</u>

Overall, Chapter 1 does a good job of introducing the ISA. Edits should strive for conciseness as synthesis. Chapter 1, more than the others, could benefit from an overall synthesis section, as well as a summary.

See comment above about the paragraph on the Anthropocene.

I like Tables 1-2 and 1-3.

Chapter 1 could use some more figures. For example, the relationship between ANC and fish diversity.

Chapter 1 might have a section on models used that is more inclusive and separate from 1.6.1.2 and 1.7.1.3. Those sections are currently mis-titled as they only cover some of the models being used. Another approach would be for each section, where applicable, have a "models" subsection, appropriately title "Models used for …" (e.g., for estuarine euthrophication).

Section 1.12 What about other ecosystem services (e.g., fish)? And what about non-N services (S, PM)?

1-81; 2,12 The use of SRP is inconsistent with the Acronyms and Abbreviations section.

Deposition maps in appendix (A-1…A-25) have scales that have 0 at the top… usually at the bottom.

Should have difference plots as well for deposition maps in the appendix (A-1…)

Map of N-deposition and of N- critical loads, together 2would be good. Again, with a difference map.

1-2 Given the split of treating welfare effects of PM between ecological and other welfare endpoints between this and the PM NAAQS reviews, further policy-related documents should make sure this is fully discussed as to the potential implications.

1-1; footnote Should be $NO3^-$ (add the "-")

1-3; 35 Modeling should be added, and it is not apparent why "gradient studies" is singled out here. (Maybe: The research includes laboratory, field and modeling studies.)

1-12; 5 replace impaction by turbulence and diffusion, along with impaction and gravitational settling for PM.

1-13;1 Might add fires (and do you mean wild fires or wildland fires?). According to Chapter 2, they are comparable with lightening and EGUs.

1-13;4: Should bring in organic-N containing PM given the growing body of evidence of its importance.

1-15; 11 Really could use a map to make this point. Also, really could use a map of the differences between decades to go along with Figs 1-3 and 1-4

1-21; Awkward sentence.

1-20; 29 Given the finding that NOx deposition leads to HABs and disease, this should be in the NOx-primary ISA. Is it?

1-37; 34 Awkward (uses analyses twice).

1-37;2. Sections of Hawaii and the Southeast are also heavily forested. What is meant here by abundant? The most total land area or the fraction covered?

1-39; 3 …abundant … abundance…

Chapter 2

The first sentence here omits PM.

Chapter 2 could also use a synthesis.

If there is any thought of using something akin to the AAI, the science should be delved in to in Chapter 2 in an integrated fashion. If not, that should be noted up front.

Section 2.8 really could use a map of overall difference in N-deposition to help the discussion (as well as other difference plots throughout).

Chapter 2, Page 2-13/2-14. First, the discussion of NH4NO3 equilibrium should be moved up a bit as the preceding paragraph uses that principle. Second, the preceding paragraph should consider Weber et al., (2016) Nature Geoscience 9, 282–285 that shows that thermodynamically, at NH3 levels as low as typically found in the US, the particles should have an NH4 to SO4 ratio below 2. In the following paragraph, one might also note that the difference in particle size/composition will also impact pH.

Section 2.3.2 seems to go directly to DON, not ON in general. Non-dissolved organic nitrate can be important.

Chapter 2: They use the Schwede and Lear (2014a) approach to fusing model results with observations to estimate deposition. While this is a reasonable choice, the approach should be better detailed in the ISA, with the important statement from that article; "Admittedly, this method does not maintain mass balance whereas Eulerian models such as CMAQ do." Indeed, a weakness here is that they should assess the lack of mass conservation, if for no other reason than to provide a bit more confidence in their results.

Chapter 2: The section on transport is rather long. What is really important here? They do not ever bring this section together in terms of the topic at hand. Ditto for deposition section.

Chapter 2. In the measurement methods section, the use of low-cost sensors should be discussed.

Chapter 2. Measurement section is not well organized. There are discussions of various N measurement techniques from some different networks, then there is a section on measuring nitrate and sulfate from other networks. Why not have the methods organized by species/group?

Chapter 2. It would be better to use the same methods to produce the spatial plots of atmospheric concentrations of various species over the US (e.g., an observation-model fusion approach).

Chapter 2. The paragraphs on how dry deposition is parameterized is overly long.

Chapter 2: Should include HNO3, NO3, SO4, NH4 deposition velocities. (Similar to Fig. 2-33)

<u>Other Comments</u>:

Table 2-1 (and throughout) Be consistent in using significant figures. For example, should on-road SO2 be 0.0 when off-road is 0.05?

Fig. 2-1 is not overly effective. It should show NOx as being emitted, not just part of the box, not sure why it spits O-N and In-N that way, particularly since it shows an organic species in the box on the LHS. It should include Na as reacting with HNO3 as well. The cycling shown is mainly used to show ozone formation, not NOx oxidation.

2-1;11 I would not use "higher order". I think the term desired is more oxidized.

2-1; second paragraph. Should include organic N as well, here.

2-4; 8: No fuel oil at all being used? (Just checking).

2-5; 31 "States" might be a more appropriate unit than "counties". I suspect there are plenty of counties out east with little SO2 being emitted from coal burning, but there will be SO2 from most counties across the US from vehicles, fires and other ubiquitous processes. Given the smaller size of counties out east, there might even be more counties with low SO2 emissions (were they rank ordered to check out the statement?).

2-7; 21 Might any of the organic N be N(III)?

2-9; 8 Just in the sunlit portions of clouds? What about on a cloudless day?

2-10;2 Is this process important: Put it in perspective. Throught the document, the "science" discussed should be relevant and apparently so.

2-15; 11. What about $NO2^-$? It may not be as plentiful as $NO3^-$, but it is there.

2-16; 1-3 This paragraph seems out of place.

2-12; 9 What is the relative amounts via each pathway? Provide cites.

2-23: 18-28 What is the point here? Not sure the relevance to how EMEP is doing some measurements.

2-25:21-… Start out with a more general discussion of how HNO3 and NO3- are measured, then get to specifics. Ditto with other sections.

Figs. 2-11, 12, 14 Provide the method used to spatially interpolate.

2-42; 5 Add molecular diffusion, remove the "s" on motions.

Fig 2-17 This figure could be improved to include deposition processes, e.g., turbulence, molecular diffusion and settling.

2-49;26 The negatives ("-") should be hyphens (-)

Appendices

Appendix A: There really should be a lead in to this Appendix describing how the maps are developed.

Nice plots visually.

As noted previously, there should be difference maps between the 2000-2002 and 2011-2013 spatial plots

I would invert the scales such that the values go up as the scale goes up.

Appendix B: Much better: it has a lead in with pertinent information.

Any way to have Fig. B-1 in color?

Can spatial information on Hg deposition be provided?

Appendix C: Good case studies. Nice graphics.

Dr. Stephen Schwartz

This review is focused mainly on Chapter 2 dealing with sources, atmospheric processes, and deposition.

GENERAL COMMENTS

Precision of standard

I expect that there are historical reasons why the nitrogen oxide secondary standard is set at 0.053 ppm (annual average), and not a round number such as 0.05 or 0.06, the implied precision being some 2%. (this precision contrasts with the several other standards: 0.5 ppb for SO_2, 15, 35, and 150 µg m^{-3} for PM). This precision would seem to require a justification. As is made abundantly clear in the document there are issues of measurement, modeling, source strengths, deposition rates, chemical reaction rates and the like that have much greater uncertainty than the 2% implied in the precision of the standard. The threshold for damage to natural and managed ecosystems, structures and the like also has much greater uncertainty. Thus a standard with such precision seems to me virtually impossible to defend. In a rational world one would set the standard to a rounder number, but perhaps there are institutional or legal reasons why such a change would be difficult to support, it taking more justification to change a standard than to leave it unchanged. That said, perhaps some explanation could be given as to why such precision should be maintained.

Following up on that, I note that in Section 2 page 2-54 estimates are given for uncertainty in deposition flux of particulate matter:

> Uncertainties in depositional flux estimates in this approach result from the combined uncertainties in the satellite-derived surface concentrations and model-derived deposition velocities used in the flux calculations; average relative uncertainties are estimated to be ~30 % for both NO_2 and SO_2 over land.

Similar uncertainty is given for dry deposition of NO_2 and SO_2, page 2-78. Such uncertainties, which in my opinion may well be optimistic, certainly raise question to the appropriateness of a standard with the precision implied by a numerical value of 0.053 ppm.

A perhaps more general question is whether a standard on atmospheric abundance (mixing ratio) is the most appropriate *form* of the standard, as opposed to, say, deposition flux. (The four elements of a standard are nicely delineated at page xlviii, line 13.) ES page lxi states that the main findings of this ISA are related to N and S deposition. I suspect that there is good reason for retaining atmospheric abundance as the form

rather than deposition flux (ability to measure mixing ratio, versus great difficulty in measuring deposition flux); also phytotoxicity is probably more related to atmospheric abundance than to deposition flux. But the question of form does not seem to be explicitly addressed in the document.

It seems as if some justification is required for the very different averaging times for the two substances, specifically for NOx, annual arithmetic average whereas SO_2 is 3 hour average, not to be exceeded more than once per year.

Presentation of uncertainties

It is fundamental in science that in presentation of measurement results or model results, it is imperative to provide uncertainties associated with the measured or modeled quantities. I thus express the concern here that that requirement is frequently not met in presentation in the Chapter under examination. I elaborate on this concern below. I am particularly concerned with maps presented showing detailed geographical distribution of deposition (e.g., Figure 2-21, page 2-63) without accompanying map showing uncertainty in the quantity. In a few instances, such as Figure 2-33, page 2-79, such maps of uncertainties are presented. Such uncertainty maps should accompany all maps of concentrations or deposition.

An alternative means of assessing uncertainties associated with concentrations and deposition fluxes calculated by chemical transport models (alternative to examining the effects of propagated uncertainties in model inputs) is comparison of the results of two or more models. Although this approach is not given prominence in the Chapter, the comparisons between results from CMAQ and CAMx reported on page 2-80 indicate substantial differences:

On an annualized basis, mean normalized errors (MNEs) in gas-phase concentrations ranged from ~25 to ~100%. MNEs in dry deposition were much larger and ranged from ~50 to >300% and MNE in wet deposition ranged from ~40 to ~100% with no clear preference for one model over another. MNE for NH_4^+ in dry and wet deposition ranged from ~35 to 70%.

Unless one or the other model can be established to be erroneous, inter-model differences serve as a measure of uncertainty in modeled quantities. To the extent that results from either model are used as a basis for standard setting, cognizance should be given to the uncertainties implied by these differences.

Separability of particulate matter from sulfur and nitrogen oxides

The title of the present document is "Integrated Science Assessment for Oxides of Nitrogen, Oxides of Sulfur, and Particulate Matter— Ecological Criteria" and it is stated (Executive summary, p. lxi) that

> The ecological effects of forms of PM, which are not related to N or S deposition, are included in Appendix D. The nonecological welfare effects associated with PM, such as visibility, climate, and materials effects, are considered as part of a separate review of PM (81 FR 87933, December 6, 2016).

This separation may be required for reasons not explicitly stated here. However, much of the generation of secondary organic particulate matter derives from reactions of natural and anthropogenic gaseous organic compounds reacting with oxidants (ozone, hydroxyl radical, NO_3 radical) whose concentrations are greatly enhanced by anthropogenic nitrogen oxides. Likewise, virtually all of the atmospheric sulfate results from anthropogenic emissions of SO_2 and sulfate, and much of the atmospheric oxidation of SO_2 to sulfate is influenced by oxidants (OH, H_2O_2) whose concentrations are enhanced by nitrogen oxides. Consequently there is an intrinsic coupling of the atmospheric chemistry governing sub-micrometer particulate matter and the nitrogen oxides and sulfur oxides under examination here. This coupling may require re-examination of the role of these oxides in formation of PM when the nonecological welfare effects of PM, which may be result in standards that are more restrictive than those related to ecological effects, are examined in the separate review referred to above.

DETAILED REVIEW OF CHAPTER 2

This chapter starts out, appropriately, with emissions. But it does not state why emissions are required (e.g., as input to models for concentrations or deposition), how they are required (location, seasonal, time of day) and how accurate they need to be. All of these requirements need to be set out quantitatively prior to any reporting of current emissions. Then emissions estimates should be presented with time and space scales appropriate to the requirements, and with uncertainties specified. I would think that for primary particle emissions it is insufficient simply to specify mass emission rates, but rather that a more differentiated picture needs to be presented, at minimum number and mass distributions of emissions. I would think that for some considerations size-distributed composition is important. Also optical properties, such as absorption coefficient. The appropriate unit might be $m^2\ s^{-1}$, evaluated as $m^2\ g^{-1} \times g\ s^{-1}$, which would be very pertinent to black carbon emissions from diesels, on- and off-road vehicles, combustion facilities, wildfires. To my thinking emissions is squarely in the bailiwick of EPA as other agencies rely on emission inventories from EPA, for example in modeling the climate effects of

tropospheric aerosols. To my thinking the treatment of emissions in the present report falls far short, either by inclusion or by reference, of adequately describing both needs for emissions inventories and the present status of such inventories relative to those needs.

Page 2-2, line 13 and throughout: "major species include..." Use of "include" here and in general is sloppy and reflects laziness. It implies that the authors have not done the necessary work to provide an exhaustive list. Better: "Major species are..." Then the reader has confidence that these are the major species and there are not others lurking out there. Even better if some indication of whether there are other species that the authors are aware of, and how much they might be contributing.

Page 2-3, line 3. Nitrous oxide is not included because of lack of reactivity, as stated correctly at page 2-1, line 19.

Table 2-1 presents emissions of several species by process, often to two significant figures, sometimes with a large leading digit, e.g., 0.74 for NOx by off-highway vehicles, which implies rather precise knowledge of the quantity, 1 part in 74, or 1.4%, which seems highly optimistic to me. It seems essential that such a table include estimates of uncertainty with clearly stated meaning of the uncertainty range. I call attention to the notation developed by IPCC in AR5 in which uncertainty ranges are qualified as to "likely" by which it is meant that it is likely that the actual value lies within the range (66% likelihood, i.e., central 66% of the pdf of the quantity, roughly ± 1 standard deviation) or "very likely (central 90% of the pdf, roughly 1.6 s. d.). It is wholly unacceptable not to present uncertainty estimates here. And it is essential that there be a transparent chain of reasoning and citations going back to the process by which these emissions are estimated. That is, it needs to be made clear that the quantities presented a summation of emissions calculated as emission factor times process rate, summed over processes (at least that is my supposition). So for each addend in the sum there needs to be an uncertainty, itself reckoned as the uncertainty in the emission factor times the uncertainty in the process rate. These uncertainties would then be propagated into the totals presented.

Further on emissions, the table is introduced (page 2-3, line 18) by the statement that the emissions are compiled from the NEI (National Emissions Inventory), with indication that the numbers come from states and other entities, but without indication of the *process* whereby the NEI numbers are generated. This should be explicitly laid out. The same para states that "For most sources, estimates are generally available for all 50 states." This raises the question of what is the magnitude of omitted emissions, requiring

statement more explicit than "most". Is it estimated that the emissions not included are 10%, 20%? This needs to be stated and justified. Line 19 states "emission estimates developed by U.S. EPA from measurements by source sector " but without any indication of what is meant by "source sector". Caption to Table 2-1 refers to "source categories". Are sectors and categories two words for the same thing, or different things? If the same, then decide which word to use. If different, then both have to be defined and distinguished.

Page 2-5, line 7, states "emissions from mobile sources have been overestimated by ~50 to 75% in the 2005 NEI " but does not get into the implications of this, other than the vague "raise concerns" at line 18. One hopes that this is addressed later. Para should state where in this document this issue is returned to and examined.

Staying with emissions some statement needs to be made at the top of the section as to why one wants total US annual emissions in the first place. Is it as input to models? In that case emphasis should be made on emissions as a function of location (and season, time of day), not just aggregated emissions. Last para on page 2-6 refers to chemical transport models in which emissions are optimized by minimizing a cost function containing contributions from the difference between model predictions and observations. This may well be a better approach, but it requires an inversion to yield emissions by location and activity to compare with bottom up models. If this is done, my guess is that the results are not highly constrained. And if it is not done, it seems to rule out any possibility of sensibly comparing modeled concentrations with measured, as the modeled concentrations are essentially derived from measured. So this needs to be spelled out. Some of these issues are recognized in that para. But the hard assessment of the accuracy of emissions relative to the requirements is not done here.

page 2-7 The para that introduces the discussion of atmospheric chemistry should make it clear *why* the discussion of atmospheric chemistry of nitrogen and sulfur species is being presented. Just says "included here"; "briefly recounted here".

Page 2-9 starts off: "Reactions producing more oxidized forms of nitrogen (NO_Z) " but in fact has reactions on which NO_2 is on the left hand side, reacting to form still more oxidized substances. The key reaction producing NO_2 is not even in the list of reactions: $NO + O_3 \rightarrow NO_2 + O_2$. This reflects poorly on the document. That said, it is not clear why these reactions are presented here in the first place.

The table of Henry's law coefficients 2-2 is accurate in the main, but misleading, as the uptake of acidic or basic species such as HONO and NH_3 is governed not just by Henry's law solubility of the gaseous species but also by the ionic dissociation reaction, which is quite rapid and the extent of which depends on solution pH. The Henry law coefficient given for HNO_3, 2.6 x $10^{6,}$ probably reflects acid dissociation. Values about an order of magnitude lower are given in the compilation by Sander Atmos. Chem. Phys., 15, 4399–4981, 2015 (note different units). To the extent that this number is used in models, this discrepancy should be noted and any implications discussed.

Page 2-15 presents observations regarding organic nitrogen but not clear why. Are the results of Cornell and Jickells consistent or inconsistent?

The top para on page 2-16 is qualitative and speculative. Is there any evidence to support this, and what fraction?

Section 2.4 commencing on page 2-16. Not clear why this is being presented. It seems very qualitative and old (figure from 1968). Seems like material being rehashed. What is the point?

Page 2-24, l 9 ff refers to so-called hybrid approach using satellite data together for column NO_2 with a model to get surface mixing ratio relative to measured column amount. (This method is used also for SO2, below). What is missing is comparisons of surface NO_2 mixing ratio by this approach versus in-situ measurements to assess the accuracy and biases, whether these biases depend on mixing ratio, etc., which can be examined and displayed only by suitable graphical comparisons.

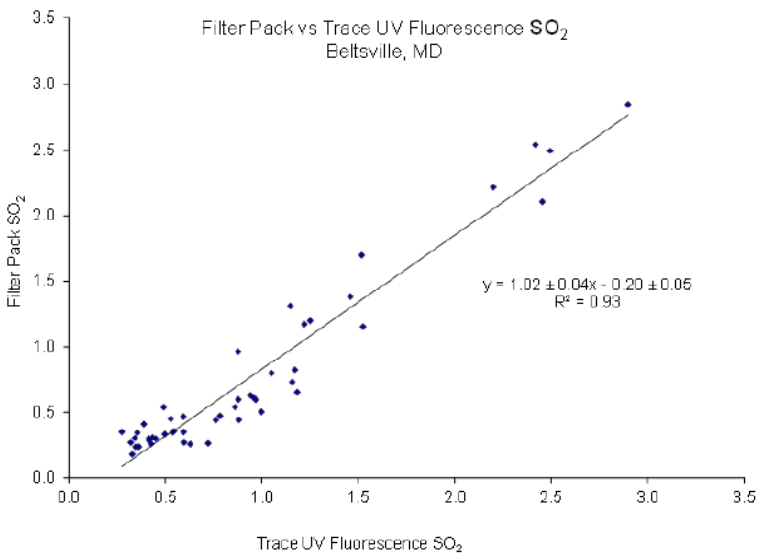
Page 2-30 presents results at rural areas noting that the pulse fluorescence method gives large relative errors at low mixing ratio of SO_2. I would ask whether this matters for purpose of compliance with standards which are much higher. Discuss.

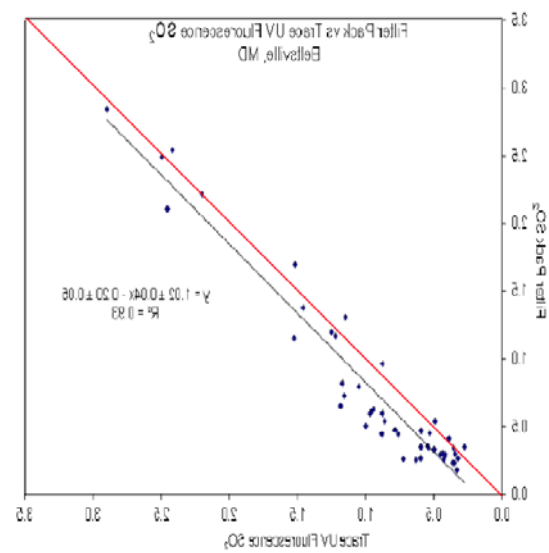
Page 2-31, line 3 states: "As can be seen in Figure 2-8, SO_2 is measured by the CASTNET filter pack by IC analysis of extracts from the cellulose filters. ". I cannot see this from the figure, which compares SO_2 measurements by two techniques. I might add that the figure scales should give the unit (ppb?). The language introducing the figure is hardly informative:

As can be seen in Figure 2-8, SO_2 is measured by the CASTNET filter pack by IC analysis of extracts from the cellulose filters. Because the nylon filter adsorbs some of the SO_2 (Sickles et al., 1999; Sickles and Hodson, 1999), SO_4^{2-} is also measured on nylon and added to the SO_2 (expressed as SO_4^{2-}) collected on the backup cellulose-fiber filters.

Results of an intercomparison of weekly average SO_2 data (ppbv) collected by the CASTNET filter pack and trace level SO_2 monitors during all of 2014 at Bondville, IL and Beltsville, MD are shown in Figure 2-8. (AMEC Environment & Infrastructure, 2015).

What is required is a statement of whether the two methods are consistent, inconsistent, which one is thought to be more accurate, and why. As a matter of style in graphics, the figure that compares two techniques should be squared up so that the physical lengths of the ordinate and abscissa scales are the same and the 1:1 line should be drawn so that one can see whether the data fall above or below that line. Then the implications should be discussed. Is there a systematic bias? How much? Is it important? Which is thought to be more accurate. Just presenting the figures is not very informative.





Left is as presented in the draft document; right is after scales are adjusted to be equal; red line denotes 1 to 1 line. Note how this presentation clearly shows the bias of one measurement against the other not evident in the original.

The document presents a lot of facts, for example, page 2-31:

In addition to the above in situ methods, satellite-based measurements have also been used to measure tropospheric SO_2 and to infer surface SO_2 concentrations with the aid of the GEOS-Chem chemistry-transport model (Nowlan et al., 2014; Lee et al., 2011). Tropospheric column abundances of SO_2 are obtained by the Ozone Monitoring Instrument (OMI) on the Aura satellite or the Scanning Imaging Absorption Spectrometer for Atmospheric Chartography (SCIAMACHY) on Envisat and are combined with results from the GEOS-Chem, global-scale, three-dimensional, chemistry-transport model to derive surface concentrations of SO_2 (as they are for NO_2). Lee et al. (2011) associated annual mean surface mixing ratios of SO_2 derived from the hybrid satellite/model technique with ambient measurements of SO_2, (R^2 = 0.66 and 0.74, slope = 0.70 and 0.93, n = 121 and 115, for OMI and SCIAMACHY, respectively).

But the significance of those facts is not presented. What are the implications of the slopes being 0.7 and 0.9. Are they consistent or not? Which is right? The model or the satellite? Can the satellite measurements replace surface networks? Why does this matter? Of all the studies in the published literaure, why are the authors presenting the ones they have selected?

Reference is made to comparisons of annual mean surface mixing ratio from the hybrid technique compared to ambient measurements. One would certainly like to see the xy comparisons plotted as the example shown above. values of n are given: 121, 115 for two satellites; one wonders whether these are for different locations. One would wish to see whether there is bias at the high values of mixing ratio; for standard setting it would seem that the low values are less important. But perhaps even more instructive would be to see individual point measurements at specific times compared to satellite to get a sense of the spread of the data. Just stating the R^2 value and the slope conveys little information.

page 2-33 refers qualitatively to sources of error in retrieval of column measurements. But what needed is the sysematic and random error in the surface mixing ratio derived from the column measurement. And an assessment of the utility of the saellite measurements for the inferring the surface mixing ratio given those uncertainties together with issues of the verticcal profile. Just listing sources of uncertainties is insufficient. If this method is to be used with confidence, it is essential to see the comparisons. The discussion of the satellite measurements concludes, page 2-33:

> The errors in the column measurements result mainly from uncertainties in the vertical profiles of NO_2 and SO_2, cloud fraction, cloud pressure, surface reflectivity, and particles used in the calculation of air mass factor. A correction is required to account for NO_2 in the stratosphere (produced from N_2O oxidation and cosmic ray interactions dissociating with N_2). The SO_2 offset correction refers to a global background correction arising from issues in spectral fitting, such as spectral correlations with O_3 and stray light within the instrument.

But it is not clear what to make of this. What is the intended use of the satellite measurements, especially as the standards in question are for surface mixing ratios, not column abundances.

Again section 2.5.6, p 2-33 to 2-35 presents a lot of facts. But what is not clear is why. Is the question whether data from the other networks are of sufficient quality that they can be used to ascertain whether standards are being met? And after all the presentation of the methods, how do the measurements compare with EPA approved methods when side by side? Are the other methods good enough? I don't think the reader wants to know if it is a nylon filter or deionized water. The reader wants the bottom line. And that is missing.

Section 2.6 page 2-36 is headed "**Geographic Distributions of Species Relevant for Deposition**, which gives a hint to the motivation of the section. First sentence line 1 says "Maps of the average distribution of atmospheric species are presented in this section. " but it needs to state why they are presented. This should be explicitly spelled out. Once the motivation is presented then that dictates the requirements. How accurately must mixing ratios or concentrations be known, and of what substances and for what purposes (compliance or ascertaining of exceedances; calculation of dry deposition; human health exposure?), on what sorts of time scales? Deposition velocities are very much a function of atmospheric stability, surface roughness, and for vegetation, photosynthetic activity and the like. It would be a great error to calculate a deposition flux as the product of a long-term mean concentration times a single deposition velocity. Authors need to make clear the requirements.

Perhaps more importantly, in view of the use of models here, a short (two page?) description should be presented here of what the model consists of, what are the inputs, the processes modeled, the outputs, the uncertainties that can be ascribed to the modeled quantities. And clear distinction needs to be made between model output and observation. It seems to me that that is essential before presenting any blended product. Comparison between modeled and observed quantities is always good, including pointing out limitations of each. Perhaps then appropriate to show blended product.

Page 2-36 lines 2-4; The fact that this is modeled mixing ratio should be stated in first sentence; not in second sentence as a "However". Otherwise it is misleading to the reader. Start with "Modeled". Height (surface or height above surface) should be specified. It would seem a matter of taste whether to present modeled results before or after measurements, but I think that both should be presented, as well as any blended product.

Fig 2-9. It would be helpful to have a reminder what NO_Y consists of, especially for comparison with NO_2 in Fig 2-10. What fraction of NO_Y is NO_2? As Fig 2-9 is a model output and as individual species should be available from the model, why not show NO_2 explicitly so that there can be an apples to apples comparison with satellite derived NO_2 in Fig 2-10?

Page 2-36, line 8-11: Particles and gases have very different deposition velocities (up to several orders of magnitude), for gases strongly dependent on the substance and substrate, and for particles strongly dependent on particle size. The text implies that concentrations of nitrates are presented as TN = HNO_3 + pNO_3^-, gaseous nitric acid plus particulate nitrate. From a deposition perspective does it even make sense

to present the sum. Authors should report separately or justify. Again as fig 2-9 is a model output, show all NO_Y species individually as mixing ratio. Then it is straightforward to compute deposition flux for each of the several species, and it should become apparent that the flux per mixing ratio (mean deposition velocity over the period) differs substantially for the several species. Actually the mean dep velocity so calculated would be a kind of hybrid quantity because of correlations of mixing ratio and dep velocity. Again if the motivation is calculation of dep fluxes all this needs to be spelled out in detail.

page 2-37, line 5; reference is made to "hazard". Hazard by what mechanism? Rather strong word. but in any event if maps of each species one would not be speculating about the composition of the sum. At line 3 "subjected to concentrations < 1 ppb" seems rather inappropriate; "exposed" seems more neutral.

page 2-39 figure shows mass concentrations. Why not convert to mixing ratio so that the comparisons with mixing ratios in other figures is more transparent.

Figs 2-11 and 2-12 are the scales chosen to be the same to permit comparison? This might be stated. The text p 2-37 describing these and other figures in this section is much too sparse, frequently just stating what is evident in looking at the figure. What is the significance of the quantities being reported? The reader is at a loss because the motivation of the presentation of the geographical distribution has not been given. I contrast the same scales used in the above figures with the quite different scales (and mode of presentation) between Figures 2-13 and 2-14. Would it be useful to show a map of the fraction of NH_3 + NH_4^+ that is one or the other to make the point.

p. 2-40, line 9, increasing at a rate of over 5% per year. Is this worth a figure? One would like to see the trend over a period of time. Ditto for other quantities.

page 2-42 ; line 1: 15.07 µg m^{-3}. Four significant figures? More importantly, what is the distribution? Is it one high value; are the high values spread over the year or seasonal. A time series here would seem especially apt.

Page 2-44: "fusion of monitoring data obtained at Clear Air Status and Trends Network sites (black dots) and Community Multiscale for Air Quality model system results." Some explanation seems required here. How is this fusion done? One would hope that the values are anchored by the measurements and that the model is used to interpolate. Is the interpolation done on a day by day basis? Other details seem essential. What governs the scales here an in Fig 2-16. They are quite different. Is this to encompass the range of

observations or to facilitate comparison. What is the significance of the blank area at the Idaho Oregon border; also Big Bend area,? No conversion of Sulfate to ppb is given; why not? I would remind the authors that mixing ratio by mole is equally valid for particulate matter as for gases. But the reader should not be required to do this in his/her head. Suggest mixing ratio throughout.

Pages 2-48 to 2-52 give a nice précis of dry deposition theory and measurement approaches. About the appropriate length and detail for this Assessment. (This précis stands out in comparison to the lack of such a précis for the modeling on which presentations of results are given in figure 2-9.). The précis also speaks to the compromises and concerns with various measurement techniques. What seems to be absent, however, is an assessment of the uncertainties and systematic errors that result from the approaches taken. One is thus presented with Tables 2-4 and 2-5 without a sense of uncertainty associated with the quantities. For SO2 comparison of the two tables shows that the dep velocity ranges from 0 over snow in winter, 0.1 over grassland in winter, 0.6 for dry grassland in summer, 1.0 for wet grassland, 3.0 for wet deciduous forest in summer, and so on. So it is clear that the value 0.8 given in Table 2 as an average has quite a range of variability. One assumes similarly for other species. So the question that does not seem to be addressed is how is this variability treated. Clearly there are two or three terms that contribute to the variability: surface properties, canopy properties, and atmospheric properties (mainly stability). So the variation in reported deposition velocities is a consequence of all three. There may be compensation point issues with respect to NH_3. One would hope that the calculations do not simply use the seasonal and vegetation type overall deposition velocities, but work with the variations in the governing resistances to calculate fluxes. The text (p 2-53, line 1) acknowledges the possibility of positive or negative correlations of concentrations with dep velocity and resultant errors, but does not seem to quantitatively address the magnitudes or consequences in calculated deposition fluxes. Some estimation of these would seem essential.

p 2-53, lime 7, "relatively short." Always better to be quantitative. For example, if transport velocity is 5 m s-1, then the distance scale is 500 km; maybe relatively short on a continental scale, but relatively long on an urban scale and relatively long compared to time scales of dispersion of point source plumes. Ditto "nearby sources" line 8. Much better if one were to say that that distance sets the scale of influence of sources.

p 2-53 line 9 to 2-54, line 3. These points (regarding temporal variability and correlation) are quite important, but what is missing is a description of how cognizance of these points is reflected in modeling

of deposition pertinent either to calculation of the rate of removal of substances from the atmosphere or to calculation of deposition fluxes.

page 2-54, line 14. The authors are commended for presenting estimates of uncertainties here. One might like to see apportionment of the uncertainty to atmospheric concentration and deposition velocity. Also some description of how the time variation of both, referred to in the previous para, is dealt with in the calculations. Uncertainty estimates such as these get to the heart of the question of whether the implied precision of a standard expressed as 0.053 ppm is justified.

page 2-55, Figure 2-18. I am puzzled by the figure from Lin. It would appear that the deposition velocities should be proportional to the ratio of flux to concentration; as all quantities are on log scale, that ratio should be proportional to the distance between the curves for mass flux and mass conc. This does not seem to be the case in this figure, so perhaps it is not the best figure to illustrate the point.

Page 2-56 – 2-57. The assessment of the state of understanding and model representation of deposition velocity of particles seems fair. The question is then the implications of these uncertainties.

Page 2-58, line 4: The acidity may be much more a function of geographical location (Arizona vs New England) than of whether the clouds are precipitating.

Page 2-58. Not clear why the discussion of throughfall; suggest motivate or omit.

Page 2-61, line 5-6, corrections such as those noted here for stickiness, here of nitric acid on inlet tubing by a factor of 1.62 should be viewed with caution. Is this important in the interpretation of deposition? If so, perhaps it should be flagged.

Page 2-61, line 7 ff. This section presents deposition maps generated by the hybrid approach (model and observations). There seems to be much merit in this approach. One is interested in the deposition as a function of time and location, but the measurements are sparse. Hence the value of using a model as an interpolation mechanism. However, If the approach described here is important to the assessment, then it would seem to require much closer scrutiny. Terms like "bias corrected modeling results" need to be fully explained and the approach assessed. Ditto "fusion of data" from different networks. There are a variety of statistical tests to assess bias between different data sets. It would seem necessary to assess such bias before fusing the data sets. Systematic differences would need to be resolved or otherwise would

contribute to measurement uncertainty. What seems to be missing in all this is estimate of bias and uncertainty, at time and space scales relevant to decision making on suitability of standards.

I am concerned about the approach used in Schwede and Lear (2014a) that is the basis of the results presented here and that presumably are used for standard setting. Examination of Figure 2 of that paper shows that weekly average dry deposition velocities (from CMAQ output) are used with weekly observed air concentrations to calculate weekly average dry deposition for each species. Because of correlations (anticorrelations) between deposition velocity and concentration over such extended periods there are inevitable errors associated with such a procedure. The question would be the magnitude of such error. I could well anticipate that it could be factor of 2 or more. This sort of question is quite amenable to examination from the time series of the deposition velocity from the model together with time series of mixing ratios of say NO_2 or SO_2 available from real-time instruments. Such an analysis would seem essential either to be included in the present document or by reference to primary literature. The magnitude of the uncertainty and bias resulting from such a procedure seems essential to inform any standard setting based on this procedure.

Page 2-61, line 26 ff. Similar concerns as with the estimation of dry deposition amount. The statement "estimates of dry deposition could be obtained using CMAQ evaluated by comparison with monitoring results" raises the question of whether this has been done; what are the results. Again the magnitude of uncertainty and bias seems essential to the use of the results.

Page 2-62, line 1 ff. I note concern regarding the maps of deposition produced by the procedure described in the foregoing paragraphs and in Schwede and Lear. Evidently the authors of the assessment are similarly concerned (lines 7-8): "it should be remembered when viewing these maps that model estimates are subject to uncertainty, and for many parameters, comparison to observations is still needed." That said, it is essential that such comparisons as are available be shown here. Only by such comparisons can one get a sense of the magnitude of errors and biases in the approach. Further, it seems to me that a statement that comparisons are needed is inappropriate here; this document is meant to be an Assessment of present knowledge and understanding pertinent to standard setting, not a statement of required work.

The map of deposition shown in Figure 2-21 is exemplary of many maps shown in the body of this chapter and in the Appendix. Rather high spatial resolution showing patterns over a variety of geographical scales as described in the text on page 2-63. However ***the recipient of this document should not be misled into***

ascribing the quality of the formatting of the map to the quality of the data being presented in the map. ***It is essential when presenting results of measurements or calculations or hybrid quantities such as those presented here to show the associated uncertainty. It is thus essential to show a map of the uncertainty associated with the quantity itself. I would assert the necessity of such an uncertainty map for each deposition map that is presented in this chapter.*** The paper of Schwede and Lear that serves as the basis for the deposition maps presented here is likewise silent on the magnitude of uncertainty associated with the calculated quantities. Although systematically examining uncertainty that results from a procedure such as that presented by Schwede and Lear is non trivial, nonetheless it is essential that such uncertainties be estimated and presented along with the results.

I note that uncertainties are given for dry deposition flux in Figure 2-33. This proves it can be done.

Page 2-64: Are these two figures complements of each other? If so omit one.

Page 2-66, line 7: "uncertainties for dry deposition are likely much larger than for wet deposition. " This statement really calls for quantitative assessment specifying the estimated uncertainty and justifying the estimate.

2-66, line 9 . "the assumption was made that 80% of pNO_3^- is in the fine mode and 20% is in the coarse mode ". Presumably this is based on observations, but it might be expected that this proportion is not a constant but varies with time and space. This would suggest the utility of ascertaining whether the deposition flux in critical areas is appreciably affected by this assumption, as noted in the remainder of that para. What is missing is the consequences of the assumption, which goes beyond the additional uncertainty noted in the conclusion of the para. Yes there is uncertainty in the actual deposition, but there should be certainty in the consequences of the assumptions in the model, and an assessment, ultimately, of sensitivity of the proposed standard to those assumptions.

Page 2-67, Figure 2-24. This is a very informative figure. The implication is that for most of the CONUS the great majority of deposition is due to explicitly modeled species, presumably NO, NO2, NH3, HNO3, nitrate. It would be valuable to have the text explicitly state this rather than the reader having to infer from the list of what is not explicitly modeled. The text at line 2 refers to "oxidized nitrogen species, whereas the figure caption says total nitrogen; this needs to be clarified. But my read of the figure is that deposition by the explicitly modeled species is at least 70% of the total in most of the CONUS. As the modeling is probably no better than that 30%, perhaps to first order deposition by species whose deposition is not

explicitly modeled can be neglected. Perhaps the assessment might explicitly state that. The text does state (lines 4-5) that the not explicitly modeled species can contribute substantially to the total in the vicinity of large urban areas. It does not address the consequences of this for the purpose of setting the standard. I am somewhat surprised at the finding, as most of the species whose deposition is not explicitly modeled are secondary, including organic nitrogen species, which I would have expected to be a larger proportion of the total well downwind of source regions. Perhaps this can be discussed.

Page 2-69, Figure 2-26. I am rather surprised at the high proportion of sulfur deposition by dry deposition coming out of the model calculations. It would be valuable here to have references back to observations that support this conclusion.

Page 2-69. The two page spreads showing the 2000-2002 and 2011-2013 panels would be more effective with a third panel showing the difference. As above some indication of uncertainty should be shown. Because of cancellation of systematic errors it might be that the uncertainty in the difference would be less than in either of the quantities themselves. Such difference plots are provided for wet deposition, Figures 2-28 – 2-31.

Page 2-71, Figure 2-27. ***This figure is an astonishing tribute to the effectiveness of the clean air act amendments, and should be an icon to the effectiveness of this legislation and to EPA***.

Page 2-79, Figure 2-33. I commend the authors on including estimates of uncertainties here. It appears as if dry deposition flux uncertainty is roughly proportional to flux. In this case would it make sense to express as fractional uncertainty to get a much smoother field? Appears to be about 30% for NO2; 50% for SO2, similar to statements at page 2-78, line 11. It would be useful to state how annual average dep velocities are calculated. And for that matter is annual dep flux calculated as mean dep velocity times mean conc, or is it the sum of dep velocity times conc for shorter intervals; and if so, how short, and how is the shorter term anticorrelation dealt with? To what extent is the uncertainty in dep flux due to uncertainty in conc, and to what extent uncertainty in dep velocity. This assessment might guide future research.

Page 2-80. I am quite uneasy over the utility of transference ratios based on annual average concentrations and deposition. I suggest that any such results be carefully scrutinized. Evidently the authors of the Assessment are similarly skeptical. The strong difference in transference ratios between the two models in Figure 2-34 is further suggestion that the approach not be used in standard setting.

Setting aside the transference ratio approach, with respect to the use of modeled concentrations in the Assessment, I note the observation at line 30 of mean normalized errors in gas phase concentrations, apparently between simulations in CMAQ and CAMx of 25 to 100%, and in dry dep 50 to 300%. These errors should on face be the source of grave concern in using the modeled quantities for standard setting. I am concerned at the apparent lack of taking cognizance of differences between the models of such magnitude throughout the chapter. It would seem essential to examine the differences between the modeled concentrations and deposition fluxes in scatter plots similar to those given in Figure 2-34 for transference ratios. This seems essential. It would be useful also to show maps of the concentrations calculated by each of the models together with a map of the differences.

WHAT IS MISSING FROM THIS CHAPTER

What seems to be missing from the Assessment is bottom line analysis of present status (mixing ratios, deposition fluxes) relative to a situation that meets various standards or other requirements such as avoidance of some level of negative effects on ecosystems. Are concentrations in compliance with present standards or out of compliance, and by how much? What are the implications? To achieve compliance do emissions need to be reduced, or alternatively, is there latitude to allow some increase in emissions. Answering the latter questions is a most suitable application for models, provided cognizance is taken of uncertainties. Almost certainly there are multiple ways in which compliance can be achieved: trade-offs between more or less stringent emission requirements in different regions. To my thinking it would be of enormous value to the policy-making community that is the customer for this assessment that the assessment show how far we need to go, and how to get there in order to achieve compliance with present or proposed concentration standards or maximum deposition fluxes (or alternatively, by how much and where emissions can be increased without incurring exceedances). As well, the assessment should provide an evaluation of the current state of the art in such modeling, e.g., 10%, factor of 2, or the like, taking into account uncertainties in the parameters in the models and various structural uncertainties in the models.

Terminology and style

Page 1-12, footnote. The footnote appropriately justifies the use of the term concentration to denote abundance expressed either as mass per unit volume or mixing ratio as "firmly entrenched in the literature."

Page 2-1 line 10. In an attempt to define nitrogen species incorrectly defines "nitrogen oxide" as NO. The common chemical nomenclature for NO is "nitric oxide." In fact the report gets it right usually, e.g., Table 2-1, Figure 2-1, Table 2-2, but occasionally reverts to "nitrogen oxide" section head for 3.5.1, page 3-14. These errors or inconsistencies can only lead to confusion and to an appearance of lack of attention to detail in the document. Please fix.

The term "acid deposition" is to be preferred to "acidic deposition". Here "deposition" is a noun formed from the verb "deposit"; the acid is the implicit object of the verb deposit. This is to be distinguished from deposition being a noun qualified by the adjective "acidic" as to what kind of deposition it is.

Throughout: Concentration seems to be used interchangeably with mixing ratio seems to be used. For example page 2-37, "broader areas of high concentrations (>~5 ppb). " At worst this can lead to confusion; at best it is an indication of sloppiness in presentation. Especially in the context of conversion between concentration and mixing ratio, e.g. p. 2-39: "Concentrations of nitric acid (µg/m3) can be converted to mixing ratios (parts per billion) to rough approximation at normal temperature and pressure by multiplying by 0.38. "

"Elevated levels"; egg p 2-40, line 4. Care needs to be taken so that the reader does not think one is speaking of vertical dependence. Ditto page 2-59, line 29.

Page 2-57, line 26 "cloudwater chemistry"; better "cloudwater composition". Still better: "Cloudwater composition and occult deposition have been measured..." That said, the term "occult deposition" is deprecated; better "Cloud drop impaction (on vegetation)". I note at page 2-62, line 18 the term used is "cloud deposition", much to be preferred.

Figure color bars: In many of the figures, e.g., 2-26, the scale runs from 0 at the top (blue) to large number (here 100) at the bottom (red). The color scale is consistent with expectation, but one generally expects such a scale to run from low values at the bottom to high values at the top, as for y-axes on graphs.

Citations. Citations are generally appropriate. However an exception is the citation Sutton et al. (2011) on page 2-9, which is to a table of Henry's law coefficients adapted from a table in the book by Sutton, for which the citation is

Sutton, MA; Howard, C M; Erisman, J W; Billen, G; Bleeker, A; Grennfelt, P; van Grinsven, H; Grizzetti, B. (2011). The European nitrogen assessment: Sources, effects and policy perspectives. In M A Sutton; C M

Howard; J W Erisman; G Billen; A Bleeker; P Grennfelt; H van Grinsven; B Grizzetti (Eds.), The European Nitrogen Assessment: Sources, Effects and Policy Perspectives (pp. 664). Cambridge, UK: Cambridge University Press.

As this was an obscure source and as I wished to check a particular value I went to some effort to get the book. When I got the book, I found the table on page . The table gives as its source the widely used compilation by Sander, of which the most recent version is readily available,

Compilation of Henry's law constants (version 4.0) for water as solvent. Atmos. Chem. Phys., 15, 4399–4981, 2015 www.atmos-chem-phys.net/15/4399/2015/ doi:10.5194/acp-15-4399-2015

So a lot of trouble to get a readily available citation if the authors had cited the paper by Sander. Much better in general to cite readily available sources rather than much less available book citations.

Production question

In reading the pdf file on the screen I notice pop-up windows that provide explanatory and/or qualifying information regarding the figures. Is there some intent to make this information available in hard copies? Which is the copy of record? Hard copy or electronic?