Preliminary Comments from Members of the CASAC Secondary NAAQS Review Panel for Oxides of Nitrogen and Sulfur on

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

(Comments Received as of August 29, 2018)

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Dr. Edith Allen

Allen, E.B. response to Risk and Exposure Assessment Planning Document
8-24-2018

Ecological Risk Assessment:

1. The overall approach for quantitatively evaluating ecological effects of atmospheric deposition on acidification and nitrogen enrichment of freshwater and terrestrial ecosystems using critical loads and exposure-response curves. [Section 4.3]

Response:
The overall approach is appropriate. Comparing changes in response receptors based on CL to current air quality standards will give a good estimate whether current primary and secondary standards are adequate.

2. The refinement of freshwater critical loads developed in the last review to include new scientific evidence on ANC levels and ecological effects. [Section 4.3.1]

No comment from E. Allen

3. The use of different base cation aluminum (Bc:Al) levels for purposes of deriving forest soil acidification critical loads, in particular, using Bc:Al ratios of 1, 10, or a range of values. [Section 4.3.2.1]

No comment from E. Allen

4. The use of the exposure-response functions to evaluate acidification and nitrogen enrichment effects on individual tree species and communities in case study areas. [Section 4.3.2.2]

Response:
The criteria for selection of case study areas are appropriate, but I have questions about one criterion, that sites recently affected by air pollution be emphasized. Are there are enough sites with recent air quality declines to apply the criterion for recent declines in air quality. Will most of these be in the drier western and central US? For long-lived tree species, this is not an appropriate criterion. It will be easier to assess the choice of case studies when those chosen are listed.

5. The approach for assessing variability/co-variability and characterizing uncertainty in the assessment of ecological effects of atmospheric deposition on acidification and nitrogen enrichment of freshwater and terrestrial ecosystems using critical loads and exposure-response curves. [Sections 4.3, 4.5.2 and 4.5.3]

Response:

The discussion on uncertainty is good. I agree that uncertainty for mycorrhizal response will be high, as data are limited, but will be better for other species. A qualitative assessment of uncertainty may be the best approach, based on quantitative assessment, and thus is appropriate (p. 4-23).
Table 4-3: Should CL for tree survival and growth response be included in this table (from Horn et al. in review)?

4.3.2.3.4 Forests and Grasslands p. 4-20--This paragraph does not add much--could be shortened and combined with 4.3.2.2.1 Trees and 4.3.2.3.3 Herbs/Shrubs. Or if there is a goal different from Trees, Herbs/Shrubs, specific to grasslands, then describe the goal here.

Overall, Section 4 would benefit from a summary paragraph or table. This could include a list of all receptors (trees, lichens, soil acidification, etc.) to be analyzed using steps 1-8 on pp. 4-9 to 4-10, and a list of models and data to assess CLs.

Dr. Praveen Amar

Comments of Praveen K. Amar on August 2018 document titled "Review of the Secondary Standards for Ecological Effects of Oxides of Nitrogen, Oxides of Sulfur, and Particulate Matter: Risk and Exposure Assessment Planning Document"

August 28, 2018

This note provides my preliminary comments on EPA's August 2018 "Review of the Secondary Standards for Ecological Effects of Oxides of Nitrogen, Oxides of Sulfur, and Particulate Matter: Risk and Exposure Assessment Planning Document".

EPA has asked the CASAC to comment on the REA Planning Document focusing on: 1) the overall analytical approach, 2) the ambient air quality analyses, and 3) the ecological risk assessment. CASAC, however, is not being asked to develop a consensus report as a result of this review of the REA Planning Document. Instead, Panel members are to discuss the responses to the REA Planning Document charge questions (see below) and then provide members' individual comments to EPA.

The details of the three areas for CASAC to comment on are included in our Charge and are outlined below:

Comments on the Overall Analytical Approach

1. The introductory and background information and, in particular, the conceptual model and key technical issues. [Chapter 1]
2. As the context for the quantitative assessments described in Chapter 4, the identification of limitations and/or uncertainties related to ecological risk and exposure as assessed in the previous NAAQS reviews and the extent to which they may be addressed by currently available information, tools and methods, thus supporting a conclusion that new or updated assessments of risk and exposure may be warranted to provide estimates with appreciably reduced uncertainty to inform decisions in the current review regarding the adequacy of the existing standards in protecting public welfare from adverse effects, and, as appropriate, similar consideration of potential alternatives. [Chapters 2 and 3]
3. The overall analytical approach for the Risk and Exposure Assessment (REA) and its appropriateness for linking ambient concentrations, atmospheric deposition and ecological effects of interest. [Section 4.1]
4. The proposed criteria and approach for selecting case study areas to evaluate potential risks and exposures in freshwater and terrestrial ecosystems. [Section 4.4]

Comments on the Ambient Air Quality Analyses

The approach for the assessment of atmospheric deposition to rely on measurements of atmospheric concentration and deposition where available, and chemical transport model simulations to provide data for chemical species and locations where measurements are either not available or limited. [Section 4.2]

1. The approach described to develop a spatially complete set of annual average deposition across the continental US for 2014-2016, as well as the proposed method of analyzing potential error and uncertainty. [Sections 4.2.1 and 4.5.1]
2. The approach for using statistical models to estimate deposition response factors that relate a change in ambient concentration to a change in atmospheric deposition, to be used for adjusting air quality in study area locations, and for quantitatively assessing the uncertainty and variability in the adjusted air quality and deposition in various study locations. [Sections 4.2.2 and 4.5.1]

Comment on the Ecological Risk Assessment.

1. The overall approach for quantitatively evaluating ecological effects of atmospheric deposition on acidification and nitrogen enrichment of freshwater and terrestrial ecosystems using critical loads and exposure-response curves. [Section 4.3]
2. The refinement of freshwater critical loads developed in the last review to include new scientific evidence on ANC levels and ecological effects. [Section 4.3.1]
3. The use of different base cation aluminum (Bc:Al) levels for purposes of deriving forest soil acidification critical loads, in particular, using Bc:Al ratios of 1, 10, or a range of values. [Section 4.3.2.1]
4. The use of the exposure-response functions to evaluate acidification and nitrogen enrichment effects on individual tree species and communities in case study areas. [Section 4.3.2.2]
5. The approach for assessing variability/co-variability and characterizing uncertainty in the assessment of ecological effects of atmospheric deposition on acidification and nitrogen enrichment of freshwater and terrestrial ecosystems using critical loads and exposure-response curves. [Sections 4.3, 4.5.2 and 4.5.3]

My comments follow:

General Comments on Executive Summary

Executive Summary is well written. Some comments follow:

Page ES-1 (Line 21): Please replace: “given the contribution of nitrogen compounds to PM including but not limited to those related to oxides of nitrogen.” by a sentence that clearly states the contribution of oxides of nitrogen AND reduced nitrogen compounds such as ammonia.

I strongly agree with the major finding of this document, that based on large amount of measurement data available since the 2008 assessment, as well as advancements in analytical tools (including TDEP), chemical transport models (CTMs), data fusion approaches including satellite data, plus substantial decreases in emissions of NOx and SO2, and increases (substantial, perhaps?) in NH3 emissions since 2008, EPA needs to undertake an updated, detailed and quantitative risk and exposure assessment.

The write up on Page ES-2 (full page) is a good description of what EPA intends to do regarding quantitative analyses of aquatic and terrestrial ecosystems effects (acidification and nitrogen enrichment) using latest data (since the 2008 ISA and REA) on critical loads and exposure-response functions. I support the need to undertake this REA at both of the two scales: national scale as well as at the scale of case study areas. There is a high probability that the uncertainties associated with the quantitative assessment of this REA would be

much lower than those in the previous REA because of the availability of more extensive air quality and deposition data as well as the availability of new analytical tools that EPA proposes to apply.

To reiterate, I agree with EPA's succinct conclusion (page 3-28, 3-29, Chapter 3) that "it is appropriate to develop a risk and exposure assessment based on the newly available air quality and ecological information, to inform the current review".

Comments on Chapter 4 (Sections 4.2, 4.2.1, 4.2.2 and 4, 5.1)

Chapter 4 describes a plan for quantitative assessment under the REA. The quantitative assessment methods for considering current conditions for national scale air quality concentrations as well as future air quality scenarios are covered in Section 4.2. (4.2.1 and 4.2.2). The subjects of variability and uncertainty as they relate to air quality are discussed in Section 4.5.1

Chapter 4 notes that the goal is to "characterize ecological risk and exposure associated with NOx, SOx, and PM for current air quality conditions as well as for when air quality is just meeting the current standards, and if appropriate, when meeting alternative air quality standards". It is obvious in this Planning Document for the future REA to be developed that the words "current standards" and "alternative standards" mean NAAQS (primary and secondary) for NO2, SO2, PM2.5 or PM10 (Table 4.1). However, as this document and the second draft ISA clearly note that the nitrogen deposition is now dominated by reduced nitrogen ("Ammonia"), it is not clear how the approach outlined in Section 4.2 will deal with ammonia under "current standards" and "alternative standards" scenarios when ammonia is simply not a controlling variable or pollutant or a "controlling standard" (the words used in this Chapter). As I see it, this may be the lost opportunity to address ammonia as an explicit "controlling standard" at least for scientific analysis.

There are a number of references in this Section to "adjusting air quality" at the scale of case study areas such that current air quality standards or potential air quality standards are "just being met". It is not clear to me what are the methods and approaches to "adjust air quality". Are we adjusting all pollutants (NO, NO2, SO2, PM2.5 (sulfates and nitrates?) or some of them? How do we decide? Are there more robust and rigorous techniques to accomplish REA's mission?

Regarding the "standard" application of CMAQ model, is CMAQ capable of calculating wet deposition of various N and S compounds? Or, does it only calculate dry deposition estimates that are then combined with estimates of wet deposition from measurements at NADP and other measurement sites? This REA notes at different places that CMAQ is capable of only estimating dry deposition and at other places, it notes that CMAQ is capable of estimating total (wet and dry) deposition. (see lines 21-22 on page 4-5).

Page 4-6 (Lines 16-17) and Page 4-7: I found this section to be very difficult to understand conceptually. This section recognizes "unique technical challenges in adjusting air quality to reflect just meeting multiple standards, particularly given that the indicator pollutants for those standards are related through emissions, chemistry, and transport." This section then goes on to describe three challenges (pollutants have different spatial variability, some are emitted directly, others are not; sensitive ecosystems are located far from large emission sources; and some components of PM that do not contribute to N and S deposition, etc.). And, on top of that, reduced nitrogen/NH3 is not even explicitly mentioned.

I think the mathematics of finding an "optimum" solution for the way we have defined this problem of so "many" variables consisting of "many" contributing sources, "many" primary and secondary pollutants, and

"many" primary and secondary NAAQS (current and future scenarios) is rather complex and not my area of expertise. However, what is being proposed does not sound very convincing to me as a robust approach.

Ms. Lauraine Chestnut

Lauraine Chestnut, preliminary comments on the REA Planning Document, August 28, 2018

Overall Analytical Approach

Applying CL from the literature: It makes sense to be planning to use the extensive CL information available in the literature. However, when deciding which CL evidence to use, it is important to include consideration of what the ecosystem services are that the CL is protecting. Why does the change that the CL would protect against matter?

CL is essentially a threshold model. Everything is not necessarily lost because of an exceedance. How can this be factored into a policy assessment? Is any change adverse? Geographic scope of the analysis is an important dimension as well as the severity of the effect.

Using case studies is appropriate when there is so much variability in the ecosystems across the county. It will be important to be able to say something about the relevance of the conclusions for any given case study to other similar areas, even if only qualitative statements can be made.

It is good to include consideration of uncertainty and the many ways it enters the analyses. Many of these uncertainties cannot be quantified, but their significance can be assessed. What matters most is if there are uncertainties that if known would likely alter the conclusions in a substantive way. Sensitivity analysis can help with this determination.

Dr. Mark Fenn

Comments on OVERALL ANALYTICAL APPROACH:
---I conclude that new and updated assessments of risk and exposure are warranted, considering more recent data, tools and information available for this purpose. Considering the lack of deposition data, especially for dry deposition, the approaches suggested for the overall analytical approach for the REA and its appropriateness for linking ambient concentrations, atmospheric deposition and ecological effects, employs effective leveraging of the available data in order to accomplish the objectives of the REA.

---page 4-21: Regarding criteria for selecting case studies, the first point mentioned is the influence of current air quality on deposition-related effects; that is to select areas in which historical deposition isn't the driving stressor. I imagine most or most likely all of the case studies selected will in fact have major historical deposition impacts, but the saving grace is likely the existence of spatial deposition gradients, or in some cases that the CL is based on N amendment studies.

Also, in the section "Diversity and Occurrence" on page 4-21, may want to also mention the value of including case studies across a diversity of climatic conditions, because responses and mechanisms of response, sensitivity of receptors, as well as mechanisms of deposition vary greatly with climatic conditions.

---Section 4.1.1 (pp 4-1 to 4-2): Certainly for many areas that are meeting the secondary and primary standards for NO2, SO2 and PM the CL will be exceeded for the lower CL values (i.e., for sensitive receptors), largely because of the combined deposition of NOx and NHx, and possibly in combination with S deposition. How is this accounted for---especially the major effect that reduced N can have?

****small scale variation in deposition can be very large with land cover, vegetation species variation, elevation and topography. In some habitats this can occur at scales much smaller than even the 4-km grid scale of the finer resolution models.

---p 1-6, lines 20-21: Shouldn't the contribution of S compounds to PM be also mentioned in this sentence?
---p 1-9, Figure 1-2: I would just comment that direct exposure studies have been done for lichens as well as plants (e.g., NH_3, for which a critical level has been established for lichens by Cape et al. 2009), but based on data in Europe, so I can see why this could be left out of the conceptual model since the focus is on the U.S.
---p 4-2, lines 8-12: Won't the available air quality data be biased toward urban sites? Or is this referring to CastNet and AMoN sites? Where is this air quality data from?

Comments on AMBIENT AIR QUALITY ANALYSES:
---The greatest challenge for the assessment of atmospheric deposition is the fact that there is no gold standard for dry deposition to which the widely-used simulated dry deposition can be compared. And even for the best available methods for calculating the dry deposition flux (e.g., inferential; eddy flux), there is also considerable uncertainty and very little data in the US. Similarly, fog or cloudwater deposition can be an important deposition component in montane and coastal sites and for this we have

relatively little data and the models don't capture this component well either. In contrast, for wet deposition there are ample reliable NADP/NTN empirical measurements for model comparison across the country, although considerable interpolation is still needed as coverage is still limited (particularly for certain habitats/regions), notwithstanding this being our most extensive network.

---I would just comment that because of the limitations mentioned in the previous paragraph (and other limitations such as the need for broader spatial coverage), we increasingly rely on simulated atmospheric deposition results that are rarely validated or compared to empirical deposition data. Ecologists using such simulated deposition seem to get lulled into "believing" the "nice" deposition maps, and rarely acknowledge the considerable uncertainty and errors in the models.

---p 4-20, lines 33-34: Here and elsewhere potential alternative air quality standards (if appropriate) are mentioned. This begs for some explanation or description of how the alternative standards compare to current standards and what determines if this is appropriate.

Comments on ECOLOGICAL RISK ASSESSMENT:

The overall approach for quantitatively evaluating ecological effects is based on the best available relationships. If the Horn et al. study on tree mortality and growth responses across CONUS is published and can be used, this would be a valuable and major addition to the assessment.

---Assessing Uncertainty: On page 4-23, lines 29-32 it states that uncertainty will be assessed by examining the difference between modeled values and measurements. This appears to be true for wet deposition, but I find it troubling that dry deposition will be assessed three different ways that all involve variants of model simulation. Considering the high uncertainty in the models, this is problematic. I'm not sure what the solution is for this, but it might be worthwhile to compare model values of atmospheric concentrations with atmospheric concentration measurements from monitoring networks, such as CastNet, IMPROVE and AMoN. One advantage of the IMPROVE data is that it includes some higher elevation or wilderness sites, where model performance is particularly uncertain and rarely evaluated. Of course, this approach wouldn't compare deposition per se, but does allow for evaluation of how well the models are estimating concentrations of key pollutants, from which deposition is determined.

---Section 4.3.1.2 Freshwater Nitrogen Enrichment: I think the approach of using the CLs, and where possible, the response functions from Baron et al. (2011), Williams et al. (2017), and Nanus et al. (2017) is promising. However, the high level of uncertainty in estimating deposition in high elevation catchments, widespread in the western US, should be acknowledged and accounted for. This is true for empirical measurements and model estimates. An example of a place that does have more intensive and comprehensive measurements of the array of nitrogenous pollutants (at least in the short-term), and associated N deposition calculations in Rocky Mt. NP (if I'm not mistaken at Niwot Ridge).

--CLs determined with one deposition method (i.e., throughfall) and exceedance with another (typically TDEP or CMAQ model).

---p 4-12, lines 7-9: This is presumably a result of declining emissions of NOx and SO_2 to form acidic SO4 and NO3 that can react with NH3 to form NH4 particulates, as described in the following paper:
Butler, T., Vermeylen, F., Lehmann, C.M., Likens, G.E. and Puchalski, M. 2016. Increasing ammonia concentration trends in large regions of the USA derived from the NADP/AMoN network. Atmos. Environ. 146: 132-140.
This can even result in increasing NH3 concentrations without increases in NH3 emissions.

---p 4-13, line 24: What is a "target CL"? I'm assuming this refers to a policy-derived acceptable CL, but seems the use of this term needs to be made clear.
---p 4-13, Footnote 48: Lake chemistry data for Baron et al. 2011 are from surveys from the mid 1980s. This data is quite dated---this should be acknowledged.

---p 4-25: In Table 4-3, in the second row (Lichens), last column (and also in the text of the document), it should be noted that the use of IMPROVE $PM_{2.5}$ N data in relation to lichen response is really a critical level not a critical load, because this is the response to an atmospheric concentration (not to deposition perse) of N in $PM_{2.5}$ (Root et al., 2015). In this study Root et al. (2015) also reported critical loads (kg N/ha/yr) for lichen responses by estimating throughfall N deposition from lichen tissue N concentration using the relationships given in Root et al. (2013).

MINOR EDITORIAL SUGGESTIONS:
---p 1-2, line 11: I would change the last word, 'with' to 'as'.
---p 1-2, lines 14-15: Edit to read: "….standard is 50 $\mu g/m^3$ as an annual arithmetic mean averaged over three years, and the secondary….".
---p 4-2, lines 7-8: Aren't the the values for the primary and secondary standards the same? If so, is this sentence needed?
---p 4-3, footnotes at bottom of page: Second footnote should be number 45.
---p 4-9, line 21: I believe this should be Table 4-2, not Table 4-1.
---p 4-15, line 30: The word "to" is missing from this sentence.
---p 4-18, lines 31-32: This is redundant---already stated that Wilson et al. 2013 includes all 94 species from Horn et al (in review).
---p 4-19, line 7: The publication by Allen et al. 2016 isn't found in the References section.
---p 4-22, Footnote 51: Inasmuch as the California case study includes Sequoia Kings Canyon NP which is in central California, I'd suggest renaming this case study as "southern/central California".
---p 4-25, line 15: This should be Table 4-3.
---p 4-28, line 19: Change 'as' to 'has'

Dr. Ivan Fernandez

Review Comments: REA Planning Document NAAQS Secondary Effects 2018 8.26.18

Ivan J. Fernandez, University of Maine

The draft REA is well written and organized, and lays out a logical approach, based on the ISA, for prioritizing quantitative analyses to be conducted. The document supports the planned REA with the foundation of science captured in the second draft ISA, as well as the work of the 2009 REA. This draft REA mentions the importance of changing patterns of temperature and precipitation in determining future risk in several instances, but can strengthen the discussion of the importance of these environmental trends in determining future risk from N and S pollution while still confining the discussion to the N and S focus. The use of case studies and the strategy to identify them is a good approach. Some of the criteria mentioned appear to favor sites without significant deposition history or sensitive species. If this eliminates whole regions that are particularly impacted by atmospheric N and S deposition, the case study criteria should be revisited.

PDF Page	Line	Comment
Executive Summary		
13	15-18	This is a complex sentence with what appears to be some awkward wording (e.g., 'when just meeting the current standards'?) that I think can be clarified.
Introduction		
15	27	'…atmospheric transformations and loading [of N] to ecosystems.'
20	Footnote 11	Same as above.
20	35	Should this be 'Impacts to public welfare…'?
21	11	Put comma after 'below'
22	Fig. 1-2	In the conceptual diagram, do the lines between ambient air and plant surfaces, soils and surface waters imply the direct deposition? That is, in a forest, does throughfall enhancement of deposition loading to the soil fall within the arrow to soils?
Chapter 2		
24	28	'Sources [of] total loading of N and S [were] discussed…'
28	25	'…[and] ANC concentrations…'
29	29	'…indicated that [a] large…'
30	7	'…for each ecoregion(s)…'
30	24	'…to a[n] ANC…'
33	7	'…watershed [is] small…'
34	29	Change 'was' to 'were'
34	32	This paragraph refers to Bc/Al as a ratio, value, and level. I would be consistent and call it a ration throughout.
35	4-9	This whole paragraph should be in the past tense.
38	6	'…whose influence [have] not been fully…'
Chapter 3		
41	5	For the record, humans have significant control over factors that govern NOx emissions from wildfires and soils. The use of this term here may be technically informed elsewhere in documentation for a particular meaning.

41	9	'…atmospheric transformations and loading [of N] to ecosystems.'
42	6	Delete ', which'
42	14	Delete 'Both'
43	31	Change 'has' to 'have'
49	16-18	This sentence appears to be missing something or is awkwardly worded.
51	1	Mountain(s)
51	5	Adirondack(s)
53	3	Northeast
53	21	Fix subscript/superscript issues (NO3-) throughout the document.
53	29-34	Can more be said about how this decision will be made to move beyond the 'unsure' status?
54	3	Comma after 'N'; also, what is the intended meaning of 'strength' as used in this sentence?
54	10	Change 'has' to 'have'
57	21	If the study included interactions with temperature and precipitation, this would be a good place to note the interaction with climate as well as acidification.
58	11	Change 'are' to 'is'
60	26	Change 'is' to 'are'
64	18-25	This section might be clarified. It begins by suggestion the literature search, not included in the ISA, deals with references that are not about nitrogen or sulfur deposition. This raises some uncertainty as why they are included. In lines 24-25 it states these references do provide linkages to deposition.
65	7	Delete 'some'
Chapter 4		
72	30	Perhaps state under 'current conditions of N and S deposition' since 2014-2016 includes I believe the first, fourth, and thirty fourth record warm years in the US and so are unique environments over the last century overall.
73	14	Delete second 'the'
74	36	Comma after 'relationships'?
77	20	'Examine if the residual [is] lower…'
79	5	'…[total] precipitation…'
81	10	It could be useful to add a sentence here noting the literature that points to rising DOC in many surface waters of the most heavily impacted regions like the Northeast, underscoring the importance of including the BCS metric in this analysis.
82	29-30	What is meant by 'atmospheric deposition patterns'?
83	18	Comma after 'N'

84	1-9	A general comment here on the use of Bc/Al ratios, well grounded in the literature. Often these use data from nationally available data on soils, usually NRCS, that is represented as an aggregated datum across the various horizons of soil, mostly mineral soil. Metrics like Bc/Al might be calibrated differently for organic O horizons, or forest floors. It would be useful to be clear on what 'soil' is for these analyses. Likewise, a metric like ANC applied to a somewhat more homogenous lake water body is conceptually similar to the Bc/Al application to soils, but is notably different on specifics. This is less critical for terrestrial aquatic linkages concerns, but more critical when considering the biological response of higher plants and soil microbial communities. It is also worth noting that the relatively rapid declines in deposition of acidifying
		compounds means soils may be in a state of recovery, which proceeds chromatographically from the top down.
85	15	Change 'this' to 'these'
87	21	Change to 'We note in our evaluation that, …'
93	32	Delete the 'to'
94	34	Change 'bases' to 'basis'
95	19	Change 'as' to 'has'

Dr. Frank Gilliam

Comments on the Ecological Risk Assessment

Frank S. Gilliam

1. *The overall approach for quantitatively evaluating ecological effects of atmospheric deposition on acidification and nitrogen enrichment of freshwater and terrestrial ecosystems using critical loads and exposure-response curves. [Section 4.3]*

   The overall approach proposed in Section 4.3 appears extremely valid, especially in providing what is likely the first synoptic assessment of the efficacy of the Clean Air Act that has ultimately resulted in notable declines in atmospheric deposition of N and S, as well as the acidity associated with both. The use of models (a bit out of my area of expertise) seems particularly promising, especially the MAGIC model that has been used with such success in the past and is, indeed, well associated with our understanding of that the text calls 'pre-acidification' conditions as a benchmark for current and future recovery.

2. *The refinement of freshwater critical loads developed in the last review to include new scientific evidence on ANC levels and ecological effects. [Section 4.3.1]*

   The use of CLs has gained significant traction in North America over the past decade or so, becoming essentially as well-developed here as it is in Europe. The proposed use of combining CL data/information from models with empirical data from the most recent peer-reviewed literature is excellent. I agree that use of ANC, although not always causally linked with the response of aquatic organisms, is a valuable metric for overall stream health. Extensive data are available to support this.

3. *The use of different base cation aluminum (Bc:Al) levels for purposes of deriving forest soil acidification critical loads, in particular, using Bc:Al ratios of 1, 10, or a range of values. [Section 4.3.2.1]*

   I am aware of some way-earlier dialogue regarding the validity/usefulness of BC:Al and its real meaning in the context of forest health. I have long thought that it is both quite valid and useful in integrating several factors that significantly impact the growth of not only trees, but also (though less often considered, unfortunately), forest herb communities. The proposed range (1-10) has a lot of justification, considering the several studies in northeast US. I would state as a caution, however, that the proposal should be clear to articulate on what basis (molar versus mass) the ratios are derived, for I have seen both in the literature and different critical ratios result, depending on how they are calculated.

4. *The use of the exposure-response functions to evaluate acidification and nitrogen enrichment effects on individual tree species and communities in case study areas. [Section 4.3.2.2]*

   This section is at once the most interesting to me and the hardest to assess. This proposed effort relies heavily on work (Horn, et al., in review) that is not available. Thus, it is difficult to make critical comments. Furthermore, although the study sounds very impressive in assessing 156 species of trees, its use in the proposed work appears to be heavily contingent on its being accepted.

5. *The approach for assessing variability/co-variability and characterizing uncertainty in the assessment of ecological effects of atmospheric deposition on acidification and nitrogen enrichment of freshwater and terrestrial ecosystems using critical loads and exposure-response curves. [Sections 4.3, 4.5.2 and 4.5.3]*

   Although this is an area with which I am less familiar, it appears that the proposal has done an extensive job in identifying sources of uncertainly, well summarized in Table 4-3 (although it is incorrectly referred to as Table 4-2 (page 4-25, line 15). I would strongly suggest that recent work of Ruth Yanai at SUNY—ESF on this topic of uncertainty would inform this component quite well.

Dr. Daven Henze

1. The approach for the assessment of atmospheric deposition to rely on measurements of atmospheric concentration and deposition where available, and chemical transport model simulations to provide data for chemical species and locations where measurements are either not available or limited. [Section 4.2]

General: I was hoping the multi-model estimates would come into play in assessment of deposition values themselves, rather than only for use as uncertainty quantification.

4-2, Fig 4-1: This figure only lists CMAQ. However, ES-20 says "models", and 3-9 (line 18) also mentions using multiple models. Later, on page 4-24, use of CAMx is discussed. Since advances in modeling are used as part of the justification for focusing REA on air quality information (3-28, lines 6-10), this might be highlighted here.

4-5, 10-14: There is also remote sensing observations (of NO2 and NH3) that could be used for evaluation (remote sensing of SO2 not likely much use here).

2. The approach described to develop a spatially complete set of annual average deposition across the continental US for 2014-2016, as well as the proposed method of analyzing potential error and uncertainty. [Sections 4.2.1 and 4.5.1]

Given this is a published, well studied approach (from Schwede and Lear, 2014), I have no further suggestions on the methods other than to reiterate comments from above (multi-models could be used for more than sensitivity / UQ, and remote sensing might be of value for evaluation).

3. The approach for using statistical models to estimate deposition response factors that relate a change in ambient concentration to a change in atmospheric deposition, to be used for adjusting air quality in study area locations, and for quantitatively assessing the uncertainty and variability in the adjusted air quality and deposition in various study locations. [Sections 4.2.2 and 4.5.1]

I had a few comments regarding the use of statistical modeling to arrive at deposition levels under adjusted conditions:

- 4-6: The adjustment cases seem like they could potentially be inconsistent, in terms of atmospheric chemical state, so it's not clear to me what the realism of these scenarios is – especially those that would entail increases in concentrations where we don't expect them to rise (4-7, 22) -- perhaps more of a counterfactual point of comparison? Was it considered instead to adjust emissions (although some trial and error required to see what adjustments are required to meet standards)?
- 4-7, 32: Lee et al. (2016) estimated areas of influence for Class I areas. If the study areas are not as pristine / remote, the area of influence may not need to be as large.
- In particular, for the statistical modeling:

- 4-8, 35: It seems like empirical statistical relationships were going to be developed, perhaps to be spatially gap-filled using model estimates. However, in the complete description of the steps to be taken, it seems the empirical information will only be used for evaluation of a statistical model built purely on output from a single model. And for this evaluation step, it doesn't sound as if there is a very clear plan on how to use the observational data.
- 4-10: Step 7 seems rather vague; the residuals will always be lower for the grid-cell specific models. The criteria "is representative and does not obscure variability in the transference ratio" is not very clear and could use refinement. It is not known what is meant by "nationally relevant" in Step 8.
- 4-10: I admire the aims and innovation presented here. Still, it seems like there is a risk of over-fitting in this procedure; a more objective approach using performance metrics determined through cross validation or comparison to the in situ measurements seems warranted. Overall, I'm a bit the approach laid forth has yet to be fully developed or tested (unlike the methods discussed for developing estimates of the current state of deposition), and is also lacking somewhat in sophistication. There is a large literature on statistical estimation in the environment, using methods from Bayesian analysis to machine learning, which might be able better (i.e., holistically and objectively) combine a wider variety of inputs, such as meteorological conditions, in situ or remote sensing measurements, as well as estimates from multiple models.

In terms of uncertainty analysis:

- 4-24: The residuals are not an estimate of the deposition response factor; rather, they present an estimate of the error in the statistical model's prediction of deposition. The error in the response factor itself would differ by a factor of transpose(G) * G where the columns of matrix G consist of the vectors of independent data used in the regression i.e., modeled atmospheric concentration time series in each grid cell (or each aggregated spatial region).
- 4-24, 24: Agreed, but what steps taken if this doesn't work out, or work out well / often enough?
- 4-24, 31: "Develop a framework" sounds vague.

Additional comments not directly related to charge:

ES-2, 15: Here it is proposed to evaluate the risk under a scenario under range of potential conditions (meeting current standard and alternative standards). Could EPA clarify how this type of analysis differs from the charge of the RIA? Additionally, could they clarify here which standards are being referred to (i.e., secondary SOx/NOx/PM or others such as primary PM, etc.)?

3-3, line 32: There are also trends estimates from space (IASI) that US NH3 concentrations in have increased by up to about 0.1 ppb/yr (Warner et al., 2017, https://doi.org/10.1002/2016GL072305)

3-4, 17-30: With the point of this section to be, in part, a summary of measurements that are newly available since the previous review, it would perhaps be more effective to include in these descriptions a snapshot of what the measurement coverage used to look like during the last REA, compared to now.

3-4, 17-30: NH3 remote sensing, especially CrIS, is another new development. Even if this has yet to be fully incorporated into the scientific assessment, the measurements exist and the science they will drive is likely on the way.

3-5, 24: NH3 emissions —> NH3 fluxes

3-5, 31: It's notable in Fig 3-4 that there are also increases. Suggest add comments / explanations for these as well. Are we seeing the impact of long-range transport, or other?

Fig 3-7: Increase in North Carolina? Is this owing to rise in NH3 emissions as well? Or encountering a situation where decreases in SO2 emissions upwind is leading to more transport of ammonium nitrate to remote regions (see Paulot et al., ES&T, 2013)?

4-10, line 20: residual lower —> residual is lower

4-12, 6-9, and Fig 4-2: Would it be more consistent with the oxidized example to compare measurements of NHx to NHx deposition?

Dr. William McDowell

Preliminary comments on the REA Planning Document

1. Inclusion of NH3 is a valuable addition to the assessment (as summarized in p. 1-2, lines 24-27), because it plays a role in the creation of oxides of S and N, and also is an important component of total atmospheric loading.

2. The REA raises a critically important point that has been part of earlier assessments: impacts of regulated pollutants must be placed in the context of other pollutant inputs at that site. As stated on page 2-2 lines 24-28, "Thus, some ecosystems may be solely impacted by atmospheric deposition (e.g., high elevation lakes), while ecological effects attributed to N and S in other systems might be largely due to non-atmospheric sources (e.g., high order streams). Sources to total loading of N and S was discussed more in the 2011 NOx/SOx PA in consideration of the standard-setting process." This important point has been carefully considered in the REA, as it should be.

3. Page 3-13 line 5. The REA should consider an additional paper relevant to the establishment of critical loads. These include Zhou et al. (2015). Zhou, Q., C. T. Driscoll, T. J. Sullivan, and A. Pourmokhtarian. 2015. Factors influencing critical and target loads for the acidification of lake–watersheds in the Adirondack region of New York. Biogeochemistry 124:353-369. This paper highlights the importance of, and the difficulties involved in, assigning responses in dissolved organic matter and organic acids to declining inputs of oxides of N and S. It also documents the effectiveness of declining atmospheric deposition of S, compared to declining N, in increasing ANC in lakes.

Dr. Erik Nelson

Erik Nelson comments on the Risk and Exposure Assessment (REA) Planning document.

Comment on the Overall Analytical Approach. Lead discussants are: *Drs. James Boyd, Elizabeth Boyer, Douglas Burns, Lauraine Chestnut, Ivan Fernandez, Erik Nelson, Richard Poirot*

1. The introductory and background information and, in particular, the conceptual model and key technical issues. [Chapter 1]

    - There are many allusions to public welfare in chapter 1 but it is never properly defined. What do we mean exactly by public welfare and how can it be affected by deposition of N, S, and PM? Do we need to give some examples of public welfare and how it can be affected by N, S, and PM deposition?
    - The chapter mentions that the REA would be concerned with how N, S, and PM deposition impacts plant, animal and aquatic life. But our welfare in this context is defined by how we interpret and experience these impacts; the impacts on plant, animal and aquatic life in of themselves are not relevant to our welfare. Therefore, the REA should be concerned with the risks to our experiences and interactions with the physical world caused by changes in plant, animal and aquatic life due to changes in the atmospheric deposition of N, S, and PM.
    - On the key technical issues. I think these should all be considered via their impact on human welfare. Therefore, I would make the bullet point "Impacts to public welfare" part of the text that introduces the "key technical issues" and then infuse all other bullet points with public welfare considerations. (As an aside, the focus on Class I areas seems misplaced; don't we care about public welfare in all places, not just conserved areas?). Belwo are some detailed thoughts in the key technical issues.
        - Source contributions and loading:
            - It would seem to me that we need to define which ecological process we care about and how each process is affected by the various sources of N and S. For example, the aquatic dead zone in the Gulf of Mexico is primarily a N via surface water problem. Maybe we can say:

                "Using analytical tools and data to help understand which pollutants are affecting the various ecological processes Americans care about will be

important, particularly in the case of reduced nitrogen impacts. This will help define which sources of N, S, and PM are relevant to this proposed REA and which sources are not"

- "Historic air quality…"
  - Again we should interpret this issue via welfare. For areas already inundated with deposition the potential to reduce risks to the ecological processes we care amount will be low. But in areas where deposition has been historically low the risk to changing the ecological processes we care about are large.
- "Timescale of effects …"
  - Again we should interpret this issue via welfare. Some ecological processes we care about can be very responsive to changes in N, S, and PM on a short-time scale; others respond very slowly to change in deposition rates.

    "This affects both the exposure period necessary for effects on the ecological processes that people care about to become adverse, as well as the uncertainty associated with the ability of the REA to assess these impacts on public welfare."

- Page 1-8, line 14: We say "As described in chapter 4, the REA for this review will include an evaluation of the relationship between emissions of the three criteria pollutants (and their precursors) and ambient air concentrations, as well as their contribution to direct as well as deposition-related exposures to biota, soils, sediments and surface waters." The don't we have to say something like:

  "Finally, we will evaluate how deposition's impact on biota, soils, sediments and surface waters could affect the ecological process that Americans value and care about, including food production, recreational experiences, aesthetic experiences, and the importance they place on ecological integrity."

- Figure 1.2: This figure has to include a link to the human dimension. How do the deposition exposures affect how we interpret and experience the physical world? How do the deposition exposures affect our welfare?

2. As the context for the quantitative assessments described in Chapter 4, the identification of limitations and/or uncertainties related to ecological risk and exposure as assessed in the previous NAAQS reviews and the extent to which they may be addressed by currently available information, tools and methods, thus supporting a conclusion that new or updated assessments of risk and exposure may be warranted to provide estimates with appreciably reduced uncertainty to inform decisions in the current review regarding the adequacy of the existing standards in protecting public welfare from adverse effects, and, as appropriate, similar consideration of potential alternatives. [Chapters 2 and 3]

   With regard to chapter 3…

   - I would delete the first bullet point on page 3-1; I do not think it is necessary given the second bullet point.
   - I agree with the assessment not to carry out an ecosystem service analysis in the proposed REA. But I do think the proposed REA can be a bit more strategic about the use of the ecosystem services concept:
     - The proposed REA will assess how freshwater and terrestrial systems respond to changes in levels of N and S via atmospheric deposition (and other sources) with exposure-response curves. After assessing the relationships an informed discussion of the links between ecological state and ecosystem services could be had.
       - For example, suppose the REA establishes a mathematical relationship between N and S deposition rates and freshwater quality (e.g., algae abundance, Secchi depth, etc). Then a discussion could be had that describes *in general* the recreational experiences and aesthetic experience at lakes across the quality gradient.
         - There is a large literature that describes recreational and aesthetic experiences across the lake quality gradient
       - Or suppose the REA establishes a mathematical relationship between N and S deposition rates and tree growth and mortality in forests. Then a discussion could be had that describes *in general* the recreational experiences and aesthetic experience in forest across the quality gradient. Further, the links between forest health and wildfire risk could be explored. For example, if fire risk is greater in less healthy forests then the public should be made aware of the potential links between N and S deposition rates, tree growth and mortality in forests, and fire risk. All of

this and more makes the link between physical effects and public welfare more concrete to the reader.

- As the chapter reads now, ecosystem services are depicted as relevant but too difficult to assess analytical, especially in a systematically consistent way across the US. I agree that a consistent, nationwide assessment is not feasible. But that does not mean the issue of ecosystem services has to be brushed aside in the proposed REA. I think an approach that adheres to what I described above could work.
- "Thresholds" is currently a hot concept in ecosystem services science (tis is different than critical loads, which define the range of loadings that have an adverse effect versus those that do not). This is the idea that an ecological system or process can flip from one stable state to another rather quickly after some threshold has been passed. This means that the welfare we derive from these processes can transition from one level to another rather quickly as well. Do academics talk about thresholds in N and S concentrations in ecological systems? If so, should the risk of passing certain thresholds be part of the proposed REA? If so, should the welfare impacts of transitioning to one ecological state to another be discussed in the proposed REA?

3. The overall analytical approach for the Risk and Exposure Assessment (REA) and its appropriateness for linking ambient concentrations, atmospheric deposition and ecological effects of interest. [Section 4.1]

- Figure 4.1 should have one more component: below the "Changes in Ecological Responses" box should be a box called "Changes in Public Welfare." I know that we are not going to measure this in the REA but I think it is important to indicate the ideal analytical framework.
- Shouldn't the box "Changes in Ecological Responses" acknowledge that output will be probabilistic? The phrase "Estimated…changes at different levels of deposition" makes it sound like we just publish the expected or average response. I assume we will assess changes as a function of deposition in a probabilistic manner?
- It appears that the case studies section will be the place where the REA authors can have the more in-depth discussion on the links between changes in ecological responses, probabilistic changes in ecosystem services, and probabilistic change in public welfare that I mentioned above. I am fine with that.

4. The proposed criteria and approach for selecting case study areas to evaluate potential risks and exposures in freshwater and terrestrial ecosystems. [Section 4.4]

- Please make sure that all lands in the US are represented not just public and conserved lands. We need to include at least one urban area, at least one suburban area, at least one exurban area, and at least one landscape dominated by agriculture use.
- Otherwise I agree with the criteria and approach for selecting case study areas.

Dr. Hans Paerl

Comments on EPA: *Review of the Secondary Standards for Ecological Effects of Oxides of Nitrogen, Oxides of Sulfur, and Particulate Matter: Risk and Exposure Assessment Planning Document,* by Hans Paerl

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

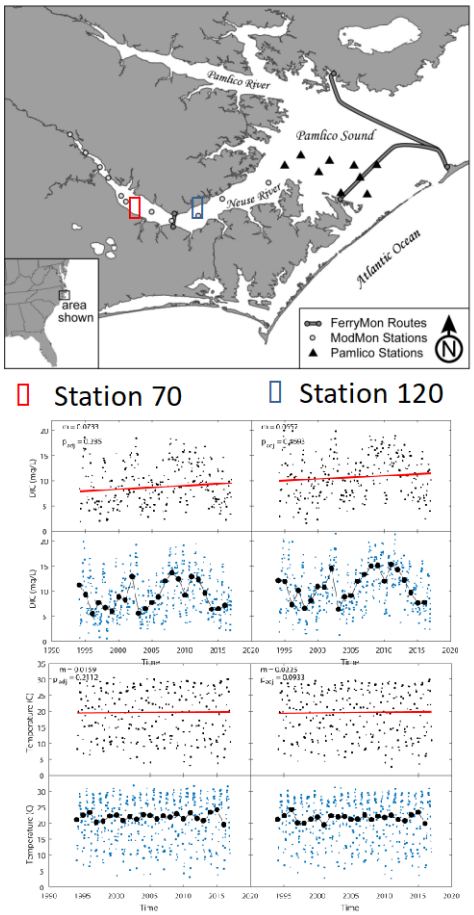
3-23, lines 1-6. It is stated that "Specifically, N has been recognized as a possible contributing factor to coastal acidification because the CO_2 produced by organic matter decomposition in eutrophic waters can contribute CO_2 to the water column along with the dissolution of atmospheric anthropogenic CO_2, decreasing the pH (see second draft ISA, Appendix section 10.5). Given the new scientific information available supporting this effect, the second draft ISA found that the relationship between atmospheric N deposition and increased nutrient-enhanced coastal acidification is likely causal."

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

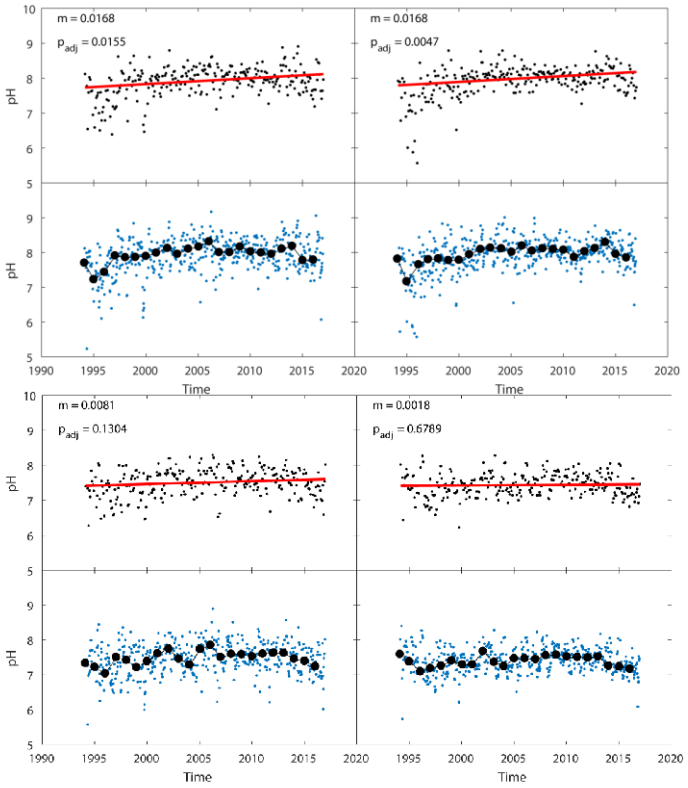
net change in pH. In the Neuse R. Estuary, it looks like pH has risen at upstream station 70, while at downstream station 120 there is no significant trend (see below).

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

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



Long-term changes in pH, DIC and Temp. in the Neuse River Estuary (Data from ModMon Program) http://www.unc.edu/ims/neuse/modmon/



I therefore caution against making the statement "Given the new scientific information available supporting this effect, the second draft ISA found that the relationship between atmospheric N deposition and increased nutrient-enhanced coastal acidification is likely causal."

This caution also applies to similar statements made in the Integrated Science Assessment for Oxides of Nitrogen, Oxides of Sulfur, and Particulate Matter: Ecological Criteria

(Second External Review Draft), APPENDIX 10. BIOLOGICAL EFFECTS OF NITROGEN ENRICHMENT IN ESTUARIES AND NEAR-COASTAL SYSTEMS.

Section 10.1 **Introduction**
Line 8. There is no "hard data" to support the "role of N in nutrient enhanced coastal acidification (Appendix)." (More below on this topic).
P. 10-2, line 8. "altered growth, total primary production" could be changed to "altered (stimulated) primary production"
Lines 16-19, the statement is made "The body of evidence is suggestive, but not sufficient to infer, a causal relationship between N deposition and changes in biota, including altered physiology, species richness, community composition, and biodiversity due to nutrient-enhanced coastal acidification." I do not agree with this statement. The biggest problem isn't acidification, but rather eutrophication and associated detrimental effects (HABs, hypoxia, food web issues, etc.)
P. 10-7, lines 26-27, it is stated that "The authors suggest that increased N deposition may enhance primary production and potentially lead to a shift from N to P limitation in this region" (referring to upper North Pacific Ocean).
This is quite speculative, and there's no real evidence that this is taking place, especially if we invoke denitrification as an important N sink.
P. 10-9, line 22. What is meant by "physical"?

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

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

See Baumann and Smith 2018, Estuaries and Coasts 41:1102-1117.

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

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

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

Mr. Richard Poirot

REA Plan Comments

Comment on the Overall Analytical Approach.

1. **The introductory and background information and, in particular, the conceptual model and key technical issues. [Chapter 1]**

An initial, contrarian comment on the background: On pp. 1-3 & 1-4 it's stated that

> "Section 109(b)(2) of 29 the CAA directs that a secondary standard is to "specify a level of air quality the attainment and maintenance of which, in the judgment of the Administrator, based on such criteria, is requisite to protect the public welfare from any known or anticipated adverse effects associated with the presence of [the] pollutant in the ambient air."

Traditionally, this is taken to mean that the method to determine compliance with the NAAQS must be measured in the ambient air. Consequently, it's reasoned that a secondary SOx/NOx NAAQS must be related to ambient air (concentration) measurements - even though the measured substances are not in the forms (deposition) which are causing the adverse effects. They are merely correlated surrogates or indicators, which require additional model-derived "transfer functions" (fudge factors) to estimate the deposition from which the effects occur. This roundabout approach is clever & innovative, but it also adds considerable complexity, error and uncertainty that would be unnecessary if the best currently available (measure + model) estimates of deposition (TDep maps, with annual temporal and 4km spatial resolution) were used to determine (current conditions &) compliance. If you can argue that measured ambient concentrations are a reasonable surrogate for the deposition of (more or less) these same substances, isn't it equally (or more) true that the measured deposition of these substances is a reasonable (and obvious) indicator of the (immediately preceding) presence of these substances in the ambient air? CAAA Section 109(b)(2) says nothing about measurements. In past NAAQS reviews you've allowed use of surrogates (AAI), direct effects (PM light extinction), and model-only results (Primary SO_2). It's not really a stretch to allow consideration of deposition measurements, with state-of-the-science PRISM & CMAQ model enhancements, to be used as the basis for determining compliance for secondary SOx/NOx NAAQS.

Other than this nit-pick, the presentation of background info, conceptual model and key tech. issues are logically reasoned and clearly presented. Under "Key Technical Issues", the identified issues are all well-justified and concisely described. I could see adding several additional issues including (*1) "Distribution of emission changes among pollutants, sources types and locations".* This is an area (somewhat unique to the REA) where - after reading your more detailed descriptions in the following sections - I still have no idea how you're gonna do it (especially if attainment of PM [& ozone] standards are considered). However you end up doing this, I encourage careful consideration of how emission controls to address NAAQS exceedances should be logically distributed - regionally and among source categories (i.e. would exceedance in the Adirondacks be addressed by NY state emission reductions only)? (*2) "Regional variability in ecosystem sensitivity".* In the past review, the AAI employed a clever mechanism that incorporated regional differences in inherent sensitivity to acidification effects. In this way, the proposed AAI essentially allowed for different AQ standards in different regions. While it's expected that a similar approach might be taken for acidification effects in this review, it's not clear how a similar "regional sensitivity" approach might be employed for N dep. effects.

The brief presentation of the conceptual model is clearly described and nicely diagrammed (Figure 1-2). One additional (external) box that might be added to Fig. 1-2 (and also generally addressed more thoroughly in the ISA and REA) might be entitled something like "Concurrent Ecological Stressors" and

include examples like "other criteria pollutants (ozone), climate change, phosphorus deposition, invasive species, insect/disease, etc.

2. **As the context for the quantitative assessments described in Chapter 4, the identification of limitations and/or uncertainties related to ecological risk and exposure as assessed in the previous NAAQS reviews and the extent to which they may be addressed by currently available information, tools and methods, thus supporting a conclusion that new or updated assessments of risk and exposure may be warranted to provide estimates with appreciably reduced uncertainty to inform decisions in the current review regarding the adequacy of the existing standards in protecting public welfare from adverse effects, and, as appropriate, similar consideration of potential alternatives. [Chapters 2 and 3]**

Generally, I think chapters 2 and 3 do an outstanding job of identifying and then addressing key uncertainties identified in the last review. The clear focus on "here's what we know now that we didn't know then" is the best I've seen in a NAAQS review (this is my 10th)! The need/value of conducting a new REA - based on advances in current understanding is persuasively argued.

3. **The overall analytical approach for the Risk and Exposure Assessment (REA) and its appropriateness for linking ambient concentrations, atmospheric deposition and ecological effects of interest. [Section 4.1]**

As indicated above, I think there are several good reasons why use of deposition measurements (or preferably model enhanced measurements of deposition) would be a superior indicator of both adverse effects, as well as compliance with any NAAQS intended to reduce these effects. Assuming the lawyers overrule me on this, I would also agree that the evidence presented in the ISA and approach proposed in the REA plan clearly support use of ambient air concentrations to asses S & N deposition effects (if that's the best we can do).

4. **The proposed criteria and approach for selecting case study areas to evaluate potential risks and exposures in freshwater and terrestrial ecosystems. [Section 4.4]**

The proposed criteria and approach for selecting case study areas are reasonable (with several objections noted in my line-by-line comments). However, I'm concerned by the "restrictiveness" of some of these criteria and the associated implication that it must be a really big deal to do the required modeling work in these few locations. It would not seem like an efficient use of scarce resources to spend a lot of effort identifying the "controlling pollutant" and/or worrying about the accuracy of "transference ratios". See my related comments on question 1. I would have thought that a more efficient approach might begin with a general evaluation of assorted CLs and CL exceedances. About how much would we need to roll back NOx (and/or SOx) emissions to meet CLs regionally (20%, 40%, 60%)? Then apply those reductions for NOx (or SOx or NHx) nationally (using rational source-specific controls) and see where the chips fall and where the benefits lay.

REA Plan, line-by-line comments

ES-1, lines 20-23: It's not clear what you're getting at here. Why the focus on contributions of N (but not S) to PM? Is this "including but not limited to NOy" a way of saying we will also assess the contributions NHx to PM and N deposition?

p. 1-2, lines 8-11: This isn't really true, as the PM secondary standards (which have no required attainment dates) are, with 1 exception, set equal to the primary standards (which are to be attained within specified time frames). Once the primary standard is attained, subsequently attaining the same

level at a more leisurely pace is nonsensical. These secondary standards protect nothing. The one exception is that the annual $PM_{2.5}$ secondary standard (15 ug/m3) is (irrationally) more lenient than the annual primary $PM_{2.5}$ standard (12 ug/m3). It has no practical protective application to anything, nor was it consistent with CASAC recommendations.

p. 1-2, lines 25-28: You could add ", transport" after "transformations" on line 27. Gaseous SO_2, NH_3^+ & HNO_3^- all dry deposit relatively rapidly - near sources. But their transformation products $(NH_4)_2SO_4$ and NH_4NO_3 are typically transported over longer distances. So NH_3 emissions enhance the transport and more distant deposition of other SOx and NOy species.

p. 2-2, lines 15-16: Do you mean "largest contributor to N deposition" or "largest contributor to acidifying deposition" or both?

p. 2-12, line 31: Is it possible these #s are reversed? Wouldn't the lower (0.6) Bc/Al ratio be associated with a higher (75%) probability of damage?

p. 3-4, line 14: Delete "Both" (you list 3 things).

p. 3-4, lines 31-33: Particulate NH_4 is not routinely measured in the IMPROVE network. It was temporarily measured at about a dozen southeastern sites starting in the late 1990s and ending in 2005. It is measured in the EPA CSN network (where it may have variable biases).

p. 4-1, lines 17-20: An adjustment to concentrations (& deposition) "just meeting current standards" generally implies an increase in emissions. Would this increase be based on bringing concentrations up to the levels of the current secondary standards at only the single worst location (very close to sources and not likely to be especially sensitive)? Or would it be more meaningful to consider scenarios where the current standards were just barely met in some of the broader regions most sensitive to acidification and/or N enrichment (for example the Adirondacks, southern Appalachians, Rocky Mtn., Sierra Nevada,…)? Would additional scenarios be considered for if worst case (or broad regional) concentrations were allowed to rise to the levels of the current primary standards (i.e. if current 2ndary NAAQS were vacated)?

p. 4-3, lines 5-11: When you consider "just meeting" current & alternative standards on a regional basis, would you adjust only regional emissions, or would you adjust emissions nationally to some common, lower level(s) of NOy and SOx? Could you include scenarios where NHx emissions were decreased?

Would the emissions changes used to evaluate alternative standards be based on attaining specific ambient air levels of SOx and NOy - such as those determinable by current ambient air monitoring networks - like CASTNet filter-pack + continuous NOy - that was proposed last time. If so (or if not) will you evaluate the adequacy (or new funding needs) of existing ambient monitoring networks to determine compliance with the alternative standards that are evaluated?

p. 4-5, lines 5-10: Here, and on several other locations, I note an emphasis on particulate NH_4 that I don't understand. Why is it important to know the contributions of gaseous NH_3 and particulate NH_4 to wet NH_4 deposition? Especially since wet NH_4 concentration (and deposition, enhanced by PRISM) is much more densely and accurately measured than NH_3 or PM NH_4? Similarly on lines 26-28: why is it important to know the relative contributions from gaseous NH_3 and particulate NH_4, but not from wet NH_4 deposition? I worry I'm missing something here in terms of the way you intend to use the available data.

pp. 4-6 to 4-8, section 4.2.2.1: The concepts discussed here seem extremely complex and would benefit from more detail or a few examples. I assume some (most) of the "study areas" are likely to be in areas where current secondary and primary SO_2, NOx and PM standards are being attained. So evaluating scenarios where current secondary and/or primary standards are "just attained" would often require increasing emissions, right? If $PM_{2.5}$ becomes the "controlling standard", how do you decide to bump up current $PM_{2.5}$ levels by increasing SOx or NOx emissions, and where those emissions should be increased? Conceivably, increasing NOx to just meeting current NO_2 standards would cause increases in ozone (and possible exceedances of ozone NAAQS) or have indirect effects from ozone enhancements of secondary PM formation rates. Similar kinds of questions arise in terms of how to choose which pollutant and which emissions locations for scenarios evaluating just meeting alternative new lower secondary standards.

p. 4-7, lines 32-36: I don't follow the logic of identifying a single "controlling standard" within each large 3000 km diameter area. Within subsets of areas that large, there may well be several different controlling standards (or current non attainment areas - especially if ozone is also considered, and it seems like it has to be). Possibly the single (highest) controlling standard for a large area might be different than that for the case study area. Again, this makes me wonder how you decide which emissions you increase or decrease and where. If $PM_{2.5}$ is the controlling standard (which I assume it (or O_3) would be for the Northeast (where here's currently some non-attainment and more areas "just under" (same for ozone), do you assume that current $PM_{2.5}$ (& ozone) standard(s) would be attained - in assessing the "at the level of current standards" scenario? Are there necessarily regulatory mechanisms in place that would prohibit NOx or SOx emissions increases from all sources upwind of and contributing to PM non attainment? Any reason not to consider increases in PM as "consequences" of increased SOx or NOx NAAQS?

pp. 4-8 to 4-12, section 4.2.2.2: This seems like a lot of effort to find surrogate indicators of substances that can be measured in the ambient air - when its the total deposition of S &/or N that's associated with the identified effects - and also the metric one would like to see decreased if CLs are exceeded. The annual TDep maps provide the best currently available, lowest uncertainty (measurement + model) estimates of the parameters of concern with 4 km spatial resolution. Why couldn't these be used to determine compliance with alternative standards? Less preferable alternatives would be to rely on CMAQ model estimates of total deposition or NTN measured wet-only deposition. After all, modeling alone is currently employed to determine compliance with the 1-hr primary SO_2 NAAQS.

p. 4-11, Table 4-2: the references to particulate NO_3, SO_4 and NH_4 all indicate a $PM_{2.5}$ size cut, but CASTNET filter packs are open-faced and have no (quantified) particle cut size. Also, I'm not sure why its necessary to consider "deposition response" factors for NHx compounds (unless you're planning to evaluate changes in NHx emissions - which is not a bad idea, but even then, you don't need an air quality indicator, unless you plan to propose an NHx NAAQS). The existing most recent TDep maps for NHX deposition are all you really need to characterize this, right? Possibly, since there is no NHx NAAQS and since NHx deposition appears to be increasing in many areas, you might want to project future NHx increases in some of your modeling (assume recent trends continue).

p. 4-17, lines 10-22: The BCw rate is clearly an important and uncertain factor. Is there any reason to also consider atmospheric dust deposition or wildfire ash. In the ISA, you attribute widespread increases in P concentrations in US lakes and streams to increases in windblown dust. Wouldn't it be logical to expect increasing dust deposition to also contribute to increasing deposition of base cations in soil?

p. 4-19, lines 11-29: Cleavitt et al., 2015 (https://doi.org/10.1639/0007-2745-118.3.304) examined lichen health metrics along a spatial gradient of Northeastern US sites with different levels of N and S

deposition. They compared (aerosol surrogate) estimates of N deposition at their NE sites to the N CL estimates that Geiser et al. (2010) had developed for the Northwest. While varying degrees of damage were noted at all the NE sites, and the Geiser et al. CLs had been exceeded at all the NE sites, they were no longer exceeded at any of the NE sites. It was also noted that the NE lichen health metrics related more strongly to long-term cumulative N deposition (aggregated for 10+ years prior to lichen sampling) than to shorter-term (current year) N deposition.

p. 4-21, lines 8-23: I understand what you're saying, but think this (current but not historical damage) is a very difficult constraint to apply (or even to understand). It sounds almost like an priori determination that "we will ignore historically-damaged areas because these are beyond hope". Meanwhile, the best "chemical recovery" evidence we have comes from areas like acidified surface waters in the Northeast where extreme historical damage was observed. It also seems like a "recent damage only" approach would tend to select for relatively clean locations only - or clean areas with recent new sources? For areas where historical damage has occurred, the effect of current decreased levels of deposition is to sustain the damage and/or prolong the recovery period. We would want to know how (or if) decreasing current deposition would alter recovery periods compared to status quo. I suggest including one or more areas with clearcut historical effects in your case study area collection.

p. 4-21, lines 39-40: Not clear what you mean by "ensuring inclusion of areas that reflect current atmospheric deposition patterns (e.g. areas dominated by oxidized as opposed to reduced forms of N)." I would think "current patterns" would show more areas dominated by reduced N than in the past, and might also expect some areas dominated by reduced N would also show recent increases. Granted, any NAAQS would focus on oxidized N, but it would still be of interest to know how affected ecosystems might respond to changes in reduced N. It could also be useful to show how much greater NOx emissions would need to be in areas with high NHx.

p. 4-22, lines 3-9: Several past considerations of secondary NAAQS (the previous SOx/NOx and previous $PM_{2.5}$ NAAQS reviews) have faulted the lack of adequate monitoring network as an important reason not to proceed with setting a NAAQS. For both SOx/NOx and $PM_{2.5}$ (PM light extinction), the Agency proposed setting up small pilot networks to test monitoring methods - but then never really followed through. So if you limit your focus to only those locations with superior air quality & deposition data, will the results be more broadly applicable. An assessment of how well current (CASTNet + NADP) AQ & deposition measurements compare with CMAQ can be conducted separately, and across the entire network(s).

p. 4-22 lines 10-13: I dislike this logic. No change at all would have complete certainty. So what?