Preliminary Review Comments from CASAC NO_X & SO_X Secondary NAAQS Review Panel on EPA's *Risk and Exposure Assessment for Review of the Secondary National Ambient Air Quality Standards for Oxides of Nitrogen and Oxides of Sulfur: Second Draft (June 2009)*

Comments Received:

Ms. Lauraine Chestnut

Executive Summary

Introduction: Setting the stage for this assessment would benefit from a short paragraph explaining why this secondary standard review is focusing on the ecological effects of NOx/SOx deposition, and not covering other potential welfare effects such as foliar injury from gaseous phases of NOx/SOx or other effects of deposition such as injury to materials.

Page ES-2: The end of this section gives a description of the extent of the problem (a description of the effect and its geographic extent) for the effect of N deposition caused nutrient enrichment in terrestrial ecosystems. Similar descriptions should be added for acidification and eutrophication. For example, the discussion on page 4-9, lines 18-28, does this well for acidification of aquatic ecosystems in the US.

Policy-relevant questions: The document does not appear to attempt to directly answer these question. Is this something that is to be done in the policy document? The first question seems very important, although perhaps a bit too broad. This review is looking only at ecosystem-related welfare effects associated with NOx/SOx deposition. When will the case be made that current standards for SO2, NO2, PM2.5 and ozone (which are currently exceeded in many locations) are not sufficient to remedy or prevent ecosystem-related welfare effects from NOx/SOx deposition?

Page ES-5, lines 18-20: The relationship between the standard and the "maximum deposition load" still seems a little vague. The standard would be the maximum ambient concentration (in the air) that would keep the deposition level at or below the maximum deposition load? The maximum deposition load is determined based on maintaining the ecological indicator at or below the level determined to be acceptable in the policy assessment?

Figure ES-2: The logical flow of this figure is good, but I still have questions about the spatial dimension given that there may be a significant distance between emissions and ambient concentrations and the ecosystems that they ultimately affect. Seems like this type of standard would allow ambient concentrations to be higher in locations where it does not lead to unacceptable levels of ecosystem indicators, which may make sense but is an unusual approach for a NAAQS. There is a sort of source-receptor relationship here that will have to be addressed somehow.

Page ES-6, line 10: Insert "selected" in front of "ecosystems", and add some explanation of why these ecosystems were selected as case studies. They aren't the only sensitive ecosystems—do they have the best data, are they most representative, are they the most sensitive? Any way to give a sense of what share of the problem in the US that these case studies represent for each of the four categories of effects?

Page ES-8: It is good to be working in the concepts of ecosystem services and how that relates to

the CAA definition of welfare effect. Be careful to include nonuse values such as habitat preservation in the descriptions of ecosystem services because there are many aspects of ecosystem services that are important to the public even though they do not involve direct human use. For example, I benefit from knowing that lakes in the Adirondacks support aquatic life without significant loss of quality due to fossil fuel emissions even though I never intend to go fishing there. This type of ecosystem service is mentioned several places in the assessment, but is not mentioned in the sections on aquatic acidification. Even though the only service intended for quantification is recreational fishing, nonuse types of services should also be listed and described.

Page ES-12: Add some sentences on the key conclusions from chapter 3 about the analysis of oxidized and reduced N.

Page ES-13: In what locations are acidification effects a problem in the US? It is not the whole country.

Page ES-18, lines 13-16: Some data are presented on forest recreation usage. It would be good to make a connection between these activities and the effects of acidification. Presumably there is some degradation in quality of experience, and perhaps even loss of area suitable for some types of recreation, due to declining forest health. Similar information for aquatic acidification could also be mentioned, as it is included in Chapter 4. Presumably current usage reflects some loss in quality or quantity due to current degraded conditions.

Page ES-19: How significant and extensive is the problem of eutrophication in US coastal estuaries?

Page ES-20: It is a very significant finding that more than 100% reduction in NOx deposition would be needed to move the case study areas from bad to poor on the indicator scale. How generalizable is this conclusion? Is NOx a comparable share of total N deposition in other sensitive locations? Is this because total N deposition is a small share of the total nitrogen entering the estuaries? Be careful with the wording here. In Chapter 7 there is mention of a "weak" relationship between aquatic nitrogen and the indicator. The weakness is in terms of the effect of changes in NOx deposition on the indicator, I think, not necessarily when all sources of N are considered. Also, be careful to avoid implying that there is no benefit of reducing NOx deposition—it may not alone be enough to solve the problem, but it might be useful if part of a broader policy to reduce N emissions.

Page ES-22: Some explanation of the relationship between lichen and forest health would be helpful here.

Page ES-24, line 14: Restrict this statement to say there is less confidence in the relationship with NOx deposition, not with N enrichment as a whole. Are there any specific circumstances with these case studies that contribute to this result, or would all coastal estuaries be similar?
Detailed comments:

Page ES-2, line 11: Sounds like sulfur can lead to nutrient enrichment and eutrophication. May need to make two sentences here.

Page ES-2, line 16: replace “alters” with “can alter”

Page ES-6, line 10: Insert “selected” before “ecosystems”

Page ES-9, line 6: Line begins “remain unidentified”, should say “remain unquantified”

Page ES-16, line 20: Connect these effects categories to the ANC levels, as is done in Chapter 4.

Page ES-16, line 35: Insert a sentence on the results for lakes at various ANC thresholds as line 37 does for the streams.

Page ES-17, lines 14-15: What does reduction in fine root growth mean for tree mortality, growth, or susceptibility? It says more in Chapter 4.

Chapter 4, page 4-8

The current value of recreational fishing is presumably depressed by reduced quality and quantity due to aquatic acidification. In the absence of these effects there would presumably be more days spent in this activity and many days would have better quality (with more locations available and better quality at locations now being used). The total value today does not say much about what the change in value would be (although it gives a sense of scale). Are there plans to make a quantitative link? Perhaps this is part of the policy assessment?

Dr. Ellis B. Cowling

In preparation for the July 22-23, 2009 CASAC meeting, my individual comments on the Second External Review Draft of the Risk and Exposure Assessment (REA) for the Secondary National Ambient Air Quality Standards for Oxides of Nitrogen and Oxides of Sulfur are organized below in part in response to Charge Questions posed in Lydia Wegman's memoranda to Kyndall Barry dated June 5, 2009. As you will see, greater attention has been given to the Executive Summary and the several Case Study Analyses which were the specific assignments given to me by Chairman Ted Russell.

With regard to the Executive Summary, the principal Charge Questions were in essence:

1) **Does the Executive Summary adequately summarize and characterize the key issues as well as the important findings of this REA analysis?**
2) **Does the Panel have any suggestions for improvement of the Executive Summary?**

My general response is that it is highly desirable to have an Executive Summary in this Second Draft REA. My specific response to question 1, however, is that the Key Issues are presented very well but that the Important Findings are not so adequately presented. Thus, my suggestion for improvement of the Executive Summary is to give much more attention to the Important Findings (such as those that are presented in the "Summary of Case Study Analysis Findings" on pages 3-79 through 3-81 in Chapter 3, "Results for the Case Study Areas on pages 4-56 through 4-71 in Chapter 4, "Current Conditions in the Case Study Areas" on pages 5-18 through 5-43 and on pages 5-58 through 5-81 in Chapter 5, "Nitrogen Addition Effects on Primary Productivity and Biogenic Greenhouse Gas Fluxes on pages 6-13 through 6-28 in Chapter 6, and in the "Synthesis and Integration of Case Study Results" on pages 7-1 through 7-24 and especially the "Conclusions" section on pages 7-24 and 7-25 of Chapter 7.

In my opinion, the Important Findings in this Second Draft REA should be presented in the Executive Summary in the form of as direct answers as possible to each of the eight Policy-Relevant Questions presented for the first time on pages ES-3 and ES-4. It also would be desirable for these Important Findings to be presented in close physical proximity within the text of the Executive Summary to the presentation of Policy Relevant Questions to which these Findings/Answers to Policy Relevant Questions apply.

General Comments on the Chemical Forms of Total Reactive Nitrogen

It was a pleasure to see that both the Executive Summary and all of the main chapters (1 though 7) of this Second Draft REA give more appropriately balanced attention than was given in the First Draft REA to the **chemically reduced** as well as the **chemically oxidized** forms of the **inorganic parts** of total reactive nitrogen (Nr). But both the Executive Summary and each of the main chapters of this Second Draft REA do not give appropriate attention to the **organic** as

well as the **inorganic** forms of **total reactive nitrogen**.

I was pleased to see that the term "Total Reactive Nitrogen" is listed among the "Key Terms" listed on pages xxi-xxviii of this Second Draft REA. But I was disappointed to see that the definition given for "total reactive nitrogen" in the "Key Terms" list is:

1) Not consistent with the way this same term is used in Chapters 3, 4, and 5, and also
2) Not consistent with the generally accepted scientific definition of this very important term.

In this connection, please note the perhaps subtle but very important differences between the definition of "total reactive nitrogen" as presented in the "Key Terms" list on page xxvii of this REA:

> "Total Reactive Nitrogen: All biologically, chemically, and radiatively active nitrogen compounds in the atmosphere and the biosphere, such as ammonia gas (NH_3), ammonium ion (NH_4^+), nitric oxide (NO), reduced nitrite (NO_2), nitric acid (HNO_3), N_2O, reduced nitrate (NO_3^-, and organic compounds (e.g., urea, amines, nucleic acids);" and the

definition of the term "reactive nitrogen" in the now publically available Executive Summary of the report of EPA's Science Advisory Board's Integrated Nitrogen Committee:

> "The term reactive nitrogen (Nr) is used in this report to include all biologically active, chemically reactive, and radiatively active nitrogen (N) compounds in the atmosphere and biosphere of Earth. Thus, Nr includes inorganic chemically reduced forms of N (NH_x) [e.g., ammonia (NH_3) and ammonium ion (NH_4^+)], inorganic chemically oxidized forms of N [e.g., nitrogen oxides (NO_x), nitric acid (HNO_3), nitrous oxide (N_2O), N_2O_5, HONO, peroxy acetyl compounds such as PAN, and nitrate ion (NO_3^-), as well as organic compounds (e.g., urea, amines, amino acids, and proteins), in contrast to non-reactive gaseous N_2."

In Chapters 3, 4, and 5, the terms "reactive nitrogen" and "total reactive nitrogen" are used to refer the sum of wet plus dry deposition of chemically reduced and chemically oxidized **inorganic** forms of air-borne biologically active nitrogen compounds and do not include the additional organic forms of airborne reactive nitrogen compounds (urea, amines, amino acids, and proteins).

In truth, at present, much more is known about the chemically reduced and chemically oxidized **inorganic** forms of reactive nitrogen deposited across the United States than about the amounts of organic forms of Nr deposited across the United States. But, at least in the Neuse River Estuary (which is one of the eight important "Case Study Areas" used in this REA report!), Hans Pearl has presented reliable evidence that as much as one third of the total atmospheric deposition of Nr compounds delivered to this estuary is deposited in the form of organic compounds -- in addition to the chemically reduced and chemically oxidized inorganic forms of Nr. Although it is not known how much organic nitrogen is present in the air sheds of the other Case Study Areas used in this REA, it would be appropriate to admit this uncertainty in the estimates of "total reactive nitrogen" and to use the term "total inorganic reactive nitrogen" or

"total inorganic Nr" (for short) rather than the misleading "total reactive nitrogen."

It also would be desirable to include some discussion (or at least an acknowledgement) about the current uncertainty about the amounts of organic forms of Nr in the air-sheds of all of the Case Study Areas. This would be especially desirable in the various specific sections of this Second Draft REA that deal with "Uncertainty" -- for example, on page 2-18 in Chapter 2, on pages 3-90 through 3-96 in Chapter 3, on pages 4-68 and 4-69 in Chapter 4, on pages 5-40 through 5-43 in Chapter 5, on pages 6-23 and 6-24 in Chapter 6, and on pages 7-21 through 7-24 in Chapter 7).

General Comments on the Major Ecological Effects of Reactive Nitrogen and Sulfur

It was a great pleasure to see very clear divisions and scientifically sound descriptions of the phenomena of excess Nr- and S- induced Acidification, Nutrient Enrichment, and Additional Effects in both terrestrial and aquatic ecosystems that are the object of the reviews of the ISA and REA documents we have reviewed so far in this integrated NOx and SOx NAAQS Secondary Standards Review process.

It is even more satisfying to see the very thoroughly thoughtout general approach to management of the ecological effects of excess Nr and S that is presented for the first time in Chapter 2 and is used as a "reminder symbol" before the title of all the main Chapters of this Second Draft REA document. I presume that the frequent use of this symbol is an indication of the pride that NCEA and OAQPS who collaborated in the creation of this general approach. Your pride in this accomplishment is well-deserved in both my personal and professional opinions.

The effort both organizations have made to develop this general approach – which is necessitated in part by the fact that chemically reduced forms of reactive nitrogen are not (yet?) recognized as Criteria Pollutants, and by the huge variety and complexity of the ecological phenomema that are of concern in the widely scattered geographical areas in which these phenomena are manifested – is especially commendable in light of the rather substantial departure from the dominating past concerns of the USEPA with largely urban- and suburban-based public-health rather than also public-welfare effects of air pollution in our country.

Permit me also to congratulate the authors of this Second Draft REA for their abundant and effective use of color illustrations that display many of the special features and unique challenges of management that will be necessary if the ecosystem protection approach that is necessary for success in establishing improved secondary standards for NOx and SOx as proposed in this REA.

It was particularly satisfying to see that the concept of Ecosystem Services has been so clearly explained and used in the development of this REA document. It also was very satisfying to see that the concept of Critical Loads is not only explained very properly but also used in presenting some of the recommendations in Chapters 4 and 7 in this Second Draft REA document.

Specific Continuing Concerns about Some of the Specific Words and Phrases Used in the

Chapters of this REA

During the now nearly six years in which I have served as a statutory member of CASAC, I have offered persistent suggestions and recommendations to decrease the confusion that often results from the use of terms that have multiple meanings and thus frequently lead to lack of clarity in many of the ideas that are presented in the ISA, REA, and Staff Paper documents used in the NAAQS review processes.

The words and phrases that I once again call attention to in the context of this REA document include the following:

"**Level**" which most importantly is used in NAAQS review documents to mean "EPA designated allowable air concentration of a criteria pollutant." But the word "level" also is used to mean many other things such as:

1) "amount" of anything -- such as the amount of biomass lost, or amount of some chemical constituent in a water body,
2) "extent" of some physical or chemical phenomenon or even the number of people in a population that are concerned about some public health or public welfare impact of pollution,
3) "elevation" when altitude or distance above sea level is intended,
4) "degree" or "intensity" of some biological phenomenon or social concern,
5) "distance above zero" for example in a graph,
6) "Type of interest" such as at the "biological species level" as opposed to "physiological level" or "biochemical level," etc, etc.

In all NAAQS review documents, I recommend that the word "level" be reserved almost exclusively to discussions about the "EPA designated allowable air concentration of a criteria pollutant" and that the great variety of alternative words be used whenever they are in fact what is intended.

"Reduce, Reducing, and Reduction" – These three terms all have both chemical and numerical meanings. Fortunately we have the unambiguous terms "decrease" and "decreasing" which have only a single (always numerical) meaning.

Thus I recommend that the unambiguous term "decrease" be used instead of the word "reduce" when our intended meaning is numerical -- and thus reserve the term "reduce" exclusive for its chemical meaning? I further recommend that the couplet "chemically reduced" and "chemically oxidized" be used when referring to the two major chemical forms of inorganic reactive nitrogen.

A particularly interesting (and at the same time very frustrating!) example of an effort to follow this recommendation in this Second Draft REA document is provided by the wording for the captions of Figure 3.4-1, Figure 3.42-2, Figure 3.4-3, Figure 3.4-5, Figure 3.4-6, and Figure 3.4-7 on pages 3-82 through 3-85 in Chapter 3. Please note that the unambiguous word "decrease" was used in the captions of all six of these figures. For example, the **caption** for Figure 3.4-1 reads as follows:

> "Figure 3.4-1. The percentage impacts of a 50% **decrease** in NOx emissions on total reactive nitrogen deposition in the East." [The **Bolding** was added by me, for emphasis].

What a great disappointment it was to then read the description of this very same figure as it was written on lines 13 through 15 of the **text** on page 3-82 within Chapter 3!. The **text** reads as follows:

> "Figure 3.4-1shows the impacts of the 50% NOx scenario on total reactive nitrogen in the East. In general, a 50% **reduction** in NOx had a 30% to 40% impact (i.e., **reduction**) on total reactive nitrogen deposition. [Once again this **Bolding** was also added by me for emphasis.]

Please also note that **text** description of this figure refers (incorrectly) to the impacts of a "50% reduction in NOx" – which is not the same as a 50% reduction in NOx **emissions** -- as is stated (correctly) in the caption itself !!!

Nitrogen, NOx, NHx, Reactive Nitrogen, Total Reactive Nitrogen, and Total Inorganic Nitrogen . There are many examples in all seven chapters of this REA document where it is not clear whether the intended meaning of the sentence is best conveyed by the word "nitrogen," "NOx," "NHx,""" reactive nitrogen," "total reactive nitrogen," or "total inorganic nitrogen. I list below a few examples where the meaning of the sentence was were particularly puzzling:

1) Lines 13-20 on page 1-10 and lines 5-6 on page 1-19 in Chapter 1.
2) Lines 4-10 on page 2-6 in Chapter 2.
3) Lines 14-20 on page 3-3 in Chapter 3.
4) Line 6 on page 3-6 and line 13 on page 3-7 in Chapter 3.
5) Lines 5 and 6 on page 4-15 in Chapter 4.
6) Lines 5 and 6 on page 5-21 in Chapter 5.
7) Lines 27-31 on page 6-17 in Chapter 6.
8) Lines 7-9 on page 7-8 in Chapter 7.

Dr. Paul J. Hanson

General Comments:

I found the second draft REA to be improved over the first draft. The document is largely successful in defining air quality indicators and ecological indicators that might be used in the context of evaluating exposure metrics for both aquatic and terrestrial in the context of acidification and nutrient enrichment for exposures to SO_x and both oxidizes and reduces forms of nitrogen.

In some cases data and justification are provided for the levels of the ecological indicators that might be considered in evaluating ecological responses, but for the most part levels, averaging times, and forms are not discussed.

While the document makes clear statements about the nature of exposures to US land surfaces and target case study ecosystems, it provides little (if any) useful characterization of the welfare risks involved in allowing current pollutant levels to continue.

To follow specific comments and minor editorial suggestions are provided for discussion and consideration by EPA staff.

Front Matter – Key Terms

Page xxi: Add a definition for ASSESTS

Page xxii: The definition listed for Determined Future Outlook doesn't stand on its own. You might also reference page numbers in the body of the text for all of the definitions related to ASSETS.

Page xxiii: Prior comments on the definition of ecological dose were not addressed. Why this definition limited to microbes?

Page xxvi: The definition of semi-arid region was not changed from the first draft. The rainfall amounts overlap for those Arid Regions, which seems inappropriate.

Executive Summary:

Page ES-2 Line 12: Change "These effects include" to "When fully developed acidification effects include…"

Page ES-2 Line 22: Change to " a well-documented phenomenon indicating…"

Page ES-3 Line 6: Change "the" to 'that'.

Table ES-7 under Terrestrial acidification: "Tree Health" is used as an undefined and unclear term. Use other words or phrases to describe what is really intended (e.g., changes in growth?).

Figure ES-3 and elsewhere throughout the REA: Although some process was conducted to limit the number of case studies for inclusion within the REA that process is not well described. Why were these case studies chosen from an original longer list of possibilities? Should these be characterized as worst-case scenarios? Should the reader assume that these are the only areas of the US for concern with respect to acidification or nutrient enrichment?

Figure ES-4 and elsewhere throughout the REA: "Change in ecosystem structure and process" is used to describe an ecological benefit/Welfare effect. How do (or can) we distinguish changes occurring through natural processes from changes from the effects of acidification and nutrient enrichment? The document should include some discussion about ecological changes in the context of 'background temporal changes" vs. those driven by pollutant exposure.

Pages ES-11 to ES-14 are quite good, but I noted one issue. The term NOy is used in the caption for figure ES-5, but it hasn't been used much in the text. Is it intended to be a placeholder for total reactive forms of N?

Page ES-17 Line 18: In this paragraph sugar maple and red spruce are characterized as being the "most sensitive" to acidification with the implication that all other tree species are less sensitive. Is this really true? Perhaps, other tree species simply haven't been evaluated in enough detail to appropriately characterize their sensitivity. Please reword the beginning of the paragraph to indicate that these species are being highlighted because sufficient data are available to evaluate their response to acidification.

Table ES-2: Does the concept of a policy relevant background Bc/Al ratio belong in this discussion?

Page ES-18 Lines 4 through 16: These paragraphs provide a description of valued characteristics of northeastern forests, but they **do not** provide an indication of fraction of these welfare metrics that are at risk under acidification**.**

General comment: The previous statement is a recurring theme throughout the REA (especially in Chapters 4, 5, and 6). Ecological benefits or measurable welfare metrics are listed for key ecosystems or case study areas with the presumption that all are subject to loss or failure with acidification or nutrient enrichment. In most cases the text (and presumably the available data) do not provide sufficient

information to fully characterize what fraction of a measurable ecological endpoint is likely to be subject to loss under pollutant exposure. I don't believe that it is appropriate for the reader to conclude that 100 percent of a given welfare metric is likely to be lost.

Page ES-21 Line 7: Don't use the term ecological health without an adequate definition.

Page ES-21 Line 12: Can you provide the deposition rate needed to drive mortality? Is it a higher level of deposition or might it alternatively be simply long term cumulative exposure to a lower deposition level?

Page ES-21 Lines 20 and 21: Please provide a range to clarify what is meant by a low C: N ratio.

Page ES-22 Lines 19 to 21: Surface area increases of root systems driven by mycorrhizae are most often associated with the morphological changes driven by ectomycorrhizae, but the authors are using AM as the example. Are they referring to fungal filament exploitation beyond the root systems of plants?

ES-23 Line 13: Are fishing and hunting really a big land use activity for the California Coastal Sage area?

Chapter 1:

Page 1-12 Line 19: Remove the word "not". It appears to generate a double negative that changes the sentence meaning.

Page 1-13: I suggest modifying the sentence by adding the underlined text as follows: "Both are essential <u>elements for vegetation growth and development</u>, and..

Page 1-14: Should "main source" be changed to 'main anthropogenic source'?

Page 1-17: These are all good policy relevant questions. Unfortunately, a number are not addressed within the REA.

Chapter 2:

Page 2-1: This section starts out with a great point – "response to pollutant exposures can vary greatly between ecosystems". Unfortunately, the REA doesn't fully address how to handle extrapolation of responses in case study areas to the balance of the US.

Page 2-1 Line 19: I would remove the word "and" in this line.

Figure 2.4-1: Please add more explanations to the figure caption. What do the arrow widths imply?

General Comment: How do we distinguish ecological effects of acidification and nutrient enrichment from other co-occurring and likely highly correlated pollutant exposures (e.g., ozone)? This issue should be fully vetted in the document.

Chapter 3:

Page 3-2 Line 18: Add the phrase 'other forms of reactive N' after NOx.

Figure 3.2-1: Increase the font size.

Figure 3.2-4: Increase the font size.

Figure 3.2-6: The figure caption uses NOy, but the related text on the previous page (3-11) exclusively discusses NOx. The authors should be consistent and define and use NOy appropriately or not at all.

Figure 3.2-10: The color scale in this figure was inappropriately changed from the scales used in the two prior figures. This isn't a big deal, but by changing the color scale a direct visual comparison isn't really possible.

Section 3.2.5: Should the concept of policy relevant background loadings or deposition levels be introduced and used?

Chapter 4:

Section 4.3.1.2

This section is a good example of how an ecological indicator can be developed for the characterization of a response to acidification. Unfortunately, the other metrics described in the REA are not this clear.

Page 4-43: Is this a true general statement or should it apply only to sugar maple on susceptible sites?

Page 4-47 Lines 9 to 12: This sentence underscores a continuing theme. It is difficult to isolate and estimate the proportion of a given measure of welfare benefit attributable to acidification and nutrient enrichment. See also Page 4-48 lines 4 to 6. Given this reality, how doe we proceed in the development of standards to protect welfare issues without a capacity to judge success or failure in the context of the target pollutants (or combined pollutants).

Chapter 5:

Page 5-1 Lines 6 to 12: This sentence should also appear in the executive summary.

Page 5-6 line 6: Spell out and define NEEA.

Page 5-44 and Section 5.3.1.1: This section is very useful and helpful to the reader. I would sentences to further describe why this discussion is limited to a couple of southern California case studies. Are other ecosystems in the US not impacted? Is there insufficient data to evaluate impacts in other ecosystems? For those that don't live in the region discussed, tell them why or how these results have meaning to broader pollutant exposures across the US.

Page 5-46 Lines 1 to 10: I would remind the reader tat one size doesn't fit all at this point. These data are quite good for lichens, but they may not have quantitative value for evaluating the response for other species or species in other regions of the US.

Section 5.3.1.3:

How is the reader supposed to interpret this information? What fraction of these services is at risk? Much of this information seems tangential.

Page 5-55 line 1: This bullet statement was discussing Eastern United States ecosystems. Do we have semi-arid lands in the eastern US?

Page 5-55 Lines 9 to 19: These statements seem to undermine the discussion. If we don't have the data why are we having the extended discussion?

Figure 5.3-5: A solid connection of this graphic to N deposition isn't made. What fraction of fuel loadings leading to fire danger and frequency can be attributed to pollutant exposure as apposed to natural secondary succession?

Page 5-66 lines 21 and 22: This is a key point.

Page 5-66 line 24: Replace "tree health" with terms that describe what you really mean.

Chapter 6:

Page 6-13 Lines 28 and 29: This statement is written as though it would apply equally to all ecosystems. I'm not convinced that this would be true for all systems at similar time frames.

Page 6-23 Lines 20 to 22: This is an important statement. I'm glad to see it included here.

Chapter 7:

Page 7-10 Line 17: Change "most" to 'known to be'.

Page 7-14 line 27: Add the level of deposition needed to drive mortality.

Page 7-15 lines 17 to 26: Why the focus on these specific metrics for CSS and MCF? Are we to conclude that these are the metrics that are the best fit for all US ecosystems? Where is the discussion about the metrics needed for the development of a US national standard? Is a standard likely to be based on most sensitive systems in the west and then applied to all ecosystems?

Page 7-18 Lines 9 to 30: What is the point of this information? How is it be used? What fraction of each of these welfare metrics are at risk under terrestrial acidification?

Page 7-19 lines 1 to 3: Add some specifics.

Page 7-19 line 29: Should oconic be iconic?

Page 7-22 lines 28 to 30: Again. This is a very key conclusion. How do we use this conclusion in the extrapolation of case study data?

Dr. Rudolf B. Husar

SOx and NOx Risk and Exposure Assessment (REA) Second Draft

EPA is to be commended for the significant improvements of REA, Second Draft. EPA has responded effectively to many of the comments from CASAC. In particular, the inclusion of a section on the CMAQ model description and evaluation is a significant addition (Appendix 1-1), since so many of the key exposure estimates and conclusions depend on CMAQ.

Appendix 1: Description of CMAQ Applications and Model Performance Evaluation

Appendix 1-4, line 5.

'The purpose of these evaluations is to provide information on how well model predictions match the observed data on the regional basis'. Evidently, 'regional' in the context of the REA means the entire Eastern US and the entire Western US. Since the SOx-NOx pattern varies considerably within each model domain, this large scale aggregation makes it difficult to assess the regional model performance, ie. NE, SE, NW, etc.

The criteria for 'acceptability' of model performance based on comparability with photochemical model performances is somewhat dubious. It is my understanding that CMAQ ozone simulations have not improved significantly for the past decade. Hence, photochemical model performance is a rather poor metric for this REA. A more defendable criterion for the CMAQ model evaluation may be the performance on deposition estimates, particularly of nitrogen compounds. The CMAQ model performance for simulating nitrogen (NO_3+NH_4) deposition is modest. Nevertheless, the REA (Appendix 1-5, Line 2) states that 'The model performance results give us confidence that our applications provide a scientifically credible approach for the purposes of this assessment'. EPA's confidence in the nitrogen deposition pattern is not shared by this reviewer.

Chapter 6 Additional Effects

6.1 Visibility, Climate and Materials

According to the charge sheet, this chapter contains results from 'some qualitative analyses for the additional effects, including visibility, climate and materials…' Evidently, the section on visibility, climate, and materials was included at the recommendation of several committee members. However, this Section 6.1 does not reflect the result of any analyses, but merely points to PM Criteria Document as the source where aerosol effects on visibility, climate and materials are treated in detail.

Since EPA has not presented a concise summary of the welfare effects on visibility, climate and materials, I would recommend eliminating this section 6.1 and replacing it with a simple disclaimer such as the sentence in Page 6-2, Line 2.

Certainly, the current phrasing does not give justice to the role of sulfates and nitrates in visibility and climate. For example, (Page 6.1, Line 16) the REA states that impairment of visibility 'can result from atmospheric particulate matter (PM), which is composed in part of sulfate and nitrate..'. In reality, even sub-microgram concentration of ambient SO_4/NO_3 **does** result in impaired visibility.

Also, the statement that 'theoretical and empirical findings suggest that sulfates often dominate the fine particle mass and hence the impairment of visibility' is a dubious formulation of the role of sulfates. The findings on the optical effects of sulfates is not just suggestive, but it is based on firm, direct measurements of both sulfate concentration and light scattering.

6.2 Sulfur and Mercury Methylation.
This section is relevant to the REA. It also properly illustrates the interaction of sulfates with the chemistry of other compounds.

6.3 Nitrous Oxide
The discussion of nitrous oxide is relevant to the climate effects of NO_x. This section appropriately describes the role of nitrous oxide. Unfortunately, the climate effects of sulfate/nitrate aerosols has fallen through the cracks. At a minimum, the recognition of those effects as part of the welfare effects of SO_x/NO_x should be stated along with the pointer to the Climate Section of the PM Criteria Document.

Dr. Dale W. Johnson

Case Study Questions to the panel:

1. Are the uncertainties appropriately characterized across the case studies? Is there adequate information to allow us to weigh the relative strengths of each case study to inform the standard setting procedure?
2. In using the Risk and Exposure Assessment to inform the policy assessment, we plan to focus on aquatic acidification as the basis for an alternative multi-pollutant secondary standard as this is the area where we have the most confidence in our ability to characterize adverse effects. Does the panel agree with this approach?

My answers to these two questions are as follows, with more detailed reviews below:

1. No, the uncertainties are not appropriately characterized and there it not, in my opinion, adequate information to inform standard setting procedures. For the aquatic case studies, there is confusion about how anions and cations interact and considerable uncertainty as to how reduced (or increased) mineral acid anions like SO4 and NO3 affect base cation concentrations directly and immediately through the necessity for charge balance as opposed to much longer term change changes in soils. The document appears to implicitly assume the latter, but without soil data to verify it, we cannot know whether soils have in fact changed at all. For the terrestrial case studies, I believe that there are significant and erroneous assumptions in the simple model used that at least should be acknowledged and discussed before any conclusions are drawn. Also, the case for red spruce decline due to acidification is not as clear as the document would lead us to believe.
2. I think this approach is OK as a <u>focus</u>, but I do not believe that it should occur to the exclusion of all other effects. I assume that this will not be the case.

There are three chapters that deal with case studies: 3, 4, and 5. These case studies all appear to focus on sites either with high levels of deposition of very high sensitivity to increased deposition. This is logical, given that the focus of this report is on potentially negative effects, but care must be taken not to extrapolate the results from these case studies to regional or national scales without first accounting for the many other sites that are not sensitive.

Chapter 3 deals with deposition rates and characteristics of the case study areas, and I really have no comment or issue with anything there.

Aquatic Acidification Case Studies

Chapter 4 deals acidification, 4.2 deals with aquatic sensitivity and 4.2.3 focuses on the Adirondack and Shenandoah sites for case studies. Here, I would comment on the section on page 4-18 that it should include a more detailed discussion of the two possible reasons for the

observed decline in streamwater base cation concentrations: 1) the reduction in the concentrations of mineral acid anions (mainly sulfate), which necessitate a reduction in cations, including base cations, in order to maintain charge balance, and 2) soil acidification. This relates to my earlier review comments as to the critical importance of considering capacity effects (change in the soil, which take a long time and are not easily reversed) and intensity (change in solution, which can take place almost instantly and are very easily reversed). The statement on lines 1-13, page 4-18 implicitly assumes that the changes are of the capacity nature, and ignore the intensity component, which in fact must play a role. Indeed, without soil data, it is impossible to know if capacity changes played any role at all or whether ""base cations buffer the inputs of NO3- and SO42-, which will likely limit future recovery of ANC concentrations." This is a somewhat confused (cations do not buffer anions) and very incomplete statement, because if the base cation decline is due to changes in SO4 and NO3 only, complete ANC recovery can be expected if the levels of those anions are brought back to pre-industrial levels somehow.

I have significant problems with the premises upon which the equations and associated text on page 4-37 are based. First of all, the assumption that the preindustrial rates of base cation leaching are sustainable is completely without foundation; were this the case, we would never find acidized soils in nature (without pollution), and we certainly do find them. The natural genesis of soils with more rainfall than evapotranspiration is to acidify; the question here is to what extent this process has been sped up by pollutant inputs. Secondly, I think that the assumption that nutrient cycling effects by plants can be ignored is deeply flawed, as the literature is full of examples where tree uptake of base cations well exceeds the removal of base cations by leaching. Furthermore, it appears that N cycling is NOT ignored, so this is only a partially imposed and inconsistent assumption.

Specific Comments:

p. 4-25: The legend on Figure 4.2-14 is way too small
p. 4-36, lines 10-12: This is true only in acid soils. In basic soils, inputs of SO4 and NO3 will have little or no effect on the "acid balance of headwater lakes". Anions affect total cation concentration by simple charge balance requirements, but they do not prescribe the type of cation.

Terrestrial Acidification Case Studies

Before specifically going into my assignment, I must comment on Section 4.3.1.1, Ecological Indicators. Here for the first time I see Bc/Al ratios, where Bc includes Ca, Mg, and K. This is only logical if the units are in moles or micromoles of charge (μmol_c), yet no units are given. What are the units? This appears to be based on a report by Sverdrup and Warfvinge published in 1993, but I was unable to get the reference and do not know what it is based on. The more commonly used indicator is that Ca/Al molar ratio of Cronan and Grigal (Cronan**,** C.S. and D.F. Grigal**.** 1995. Use of Ca/**Al** ratios as indicators of stress in forest ecosystems. Journal of Environmental Quality 24: 209-226.

Also, on page 4-44, it is clearly stated that acidification negatively affects red spruce – I admit to not being up to speed on the latest developments, but I do recall that Art Johnson found that climate change, not acid rain, was responsible for the red spruce decline in New England. I think this treatment is a bit one sided and the situation is not that clear. (Johnson, A.H., E.R. Cook, and T.G. Siccama. 1988. Climate and red spruce growth and decline in the northern Appalachians. Proc. NatI. Acad. Sci. USA, Vol. 85, pp. 5369-5373.)

The actual case studies for HBEF, which assumes that net forest increment is zero (I doubt that very much, even if the site has not been recently harvested and would ask for some documentation of that) and the KEF sites, and builds the analysis on the SMB model which has numerous assumptions and the unitless Bc/Al ratio to conclude that "These results suggest that the health of red spruce at HBEF and sugar maple at KEF may have been compromised by the acidifying nitrogen and sulfur deposition received in 2002." I realize that all models are imperfect and yet there may be good reasons to run them anyway, but this seem grossly overstated and I would add many many caveats to this section.

Nutrient Enrichment Questions for the Panel:

1. Section 5.2 and Appendix 6 describe the analyses used to evaluate the effect of aquatic nutrient enrichment. The analysis uses the SPARROW model on one stream reach (Potomac River and Neuse River) to determine the impact of atmospheric total nitrogen deposition on the eutrophication index for the estuary. Does the Panel think that the model is adequately described and appropriately applied?
2. Section 5.3 and Appendix 7 describe the analyses used to evaluate the effect of terrestrial nutrient enrichment. This qualitative analysis describes the impacts due to nitrogen deposition on the Coastal Sage Scrub community of California and mixed conifer forests in the San Bernardino and Sierra Nevada Mountains and larger areas where possible. In addition, the effects on nitrogen deposition in the Rocky Mountain National Park supplemental case study location are summarized. How would the Panel apply threshold values presented in this case study to allow for broader geographic application that accounts for regional variability? Have the associated uncertainties been adequately characterized?

My answers to these two questions are as follows, with more detailed reviews below:

1. I cannot intelligently answer this question. I am not familiar with the SPARROW model and would not be confident in commenting on it without considerably more information as to its structure and premises – more than could or should be included in a document such as this. I will pass on this one to other panelists who probably have more knowledge on the matter than I do.
2. I would not apply the threshold values presented in this case study to a broader geographic region because these case studies, while appropriate for negative effects of pollutant inputs either at high levels or on sensitive sites, do not in any way address the larger majority of ecosystems which are either resistant to negative effects or in fact might benefit from the additional nitrogen inputs. This is in essence a philosophical issue:

should standards be set on the basis of the most sensitive or highly impacted systems or should they be based on regional effects? Since the question asked about regional application, I am assuming the latter and thus my answer is that these case studies are in no way regionally applicable. The uncertainties with this are considerable and not well characterized.

First of all, this section is really mislabeled: it really focuses on nutrient excess, not enrichment and I suggest Nutrient Excess as the title. Nutrient enrichment to terrestrial people often implies something good, and that certainly is not the focus of this section.

Aquatic Nutrient Enrichment

I am not familiar enough with either the SPARROW model or the sites to intelligently comment on that section.

Terrestrial Nutrient Enrichment

The case studies for nutrient excess are good ones, with excellent research programs documenting negative effects of high levels of N deposition. As in my previous reviews, however, I must again go to the mantra of taking a balanced approach to this issue and if this section is still to be entitled Nutrient Enrichment, some mention of the possible benefit to commercial forests in the Pacific Northwest and Southeast should be mentioned. I fully recognize by now how loathe the authors are to do this, but I will continue to make this comment as long as I am on this panel as I think it is important and the omission of it will be greatly regretted later. (for example, see Chappell, H.N., D. W. Cole, S. P. Gessel and R. B. Walker. 1991. Forest fertilization research and practice in the Pacific Northwest. Nutrient Cycling in Agroecosystems. 27: 1385-1314; see also this link for a lay article on fertilization in southeastern pine forests: http://www.ipni.net/ppiweb/bcrops.nsf/$webindex/2476B56D4FDD9EB0852571B1006A6F2E/$file/06-3p12.pdf)

Additional Effects Questions for the Panel

1. In this chapter, we have presented results from some qualitative analyses for additional effects including visibility, climate and materials, the interactions between sulfur and methylmercury production, nitrous oxide effects on climate, nitrogen addition effects on primary productivity and biogenic greenhouse gas fluxes, and phytotoxic effects on plants. Are these effects sufficiently addressed in light of the focus of this review on the other targeted effects in terms of the available data to analyze them?

My answers to this questions are as follows, with more detailed reviews below:

1. The segments on visibility, climate and materials, the interactions between sulfur and methylmercury production, nitrous oxide effects on climate seem to be adequately

addressed, but, for the reasons given in detail below, the segment on nitrogen effects on primary productivity still has some serious problems and issues, as detailed below.

I have serious problems with the assessment of nitrogen effects on primary productivity in this section. First of all, the authors appear to go through significant intellectual gymnastics in order to either ignore or disprove the concept that greater primary production leads to more C sequestration. There may be cases where primary production does not lead to increased C sequestration, and the best example of this is the forest floor, which decreases in mass as mean annual temperature increases despite the increases in primary productivity. However, given that organic C for sequestration is produced during primary production, to cling to the notion that the two are not related defies logic.

Furthermore, the statements regarding nitrogen in this section are largely untrue and often given without citation, and fly in the face of published literature. Examples of this is are on page 6-13, lines 15-17 which states that growth increases due to N inputs are offset by increases in soil respiration and on page 6-14, lines 27-29 where it states that "increased leaf N concentration under conditions of elevated nitrogen deposition may result in higher carbon loss by increasing both autotrophic and heterotrophic respiration". This is simply not true: most studies show that N fertilization causes **decreases** in soil respiration (see for example Tyree, Michael C.; Seiler, John R.; Fox, Thomas R. The Effects of Fertilization on Soil Respiration in 2-Year-Old *Pinus taeda* L. Clones Forest Science, Volume 54, Number 1, February 2008 , pp. 21-30; see also Olsson, P., S. Linder, R. Giesler, and P. Högberg. 2005. Fertilization of boreal forest reduces both autotrophic and heterotrophic soil respiration. Global Change Biology 11 1745– 1753; and Google "Fertilization effects on soil respiration" for many other references**)**

On page 6-14, lines 23-31, the authors state that higher nitrogen concentration in organic matter stimulates decomposition; while this is sometimes true in the early stages of decomposition, the literature is clear on the long-term effects: greater N concentration **increases** the long-term storage of stable organic matter.

See:

B. Berg and C. Mcclaugherty (2003). Plant Litter–Decomposition, Humus Formation, Carbon Sequestration. Springer Verlag, 286 pp., 76 figs.

Sarah E. Hobbie (2008) NITROGEN EFFECTS ON DECOMPOSITION: A FIVE-YEAR EXPERIMENT IN EIGHT TEMPERATE SITES. Ecology: Vol. 89, No. 9, pp. 2633-2644

Berg, B., Ekbohm, G., Johansson, M.-B., McClaugherty, C., Rutigliano, F., and Virzo De Santo, A. 1996. Maximum decomposition limits of forest litter types: a synthesis. Can. J. Bot. **74**: 659–672.

Berg, B., McClaugherty, C., Virzo De Santo, A., and Johnson, D. 2001. Humus buildup in boreal forests: effects of litterfall and its N concentration. Can. J. For. Res. **31**: 988–998.

For the reasons given above, I believe this section to be seriously biased and flawed. I believe that the authors need to better research the literature and present a more balance and accurate assessment of the effects of N on ecosystem C sequestration.

Dr. Myron J. Mitchell

Comments are provided in *italics*

Charge to the CASAC NOx/Sox Secondary Review Panel

Within each of the main sections of the second daft Risk and Exposure Assessment document, we ask the panel to address the following questions, taking into consideration the changes and additions since the first draft Risk and Exposure Assessment:

Executive Summary:

1. In response to the Panel's review of the first draft Risk and Exposure Assessment, we have included an executive summary of this document. Does the Executive Summary adequately summarize and characterize the key issues driving this review as well as the important findings of the analyses? Does the Panel have any suggestions for clarification or refinement of the Executive Summary?

The Executive Summary does a good job of providing a summary of the document. The summary, however, needs attention to detail, clarity and consistency. For example, there needs to be more consistency in the use of the past and present tense. If the focus is on the summary of findings, the past tense is appropriate. However, if emphasis is on the current conditions, the present tense should be used. There is some redundancy in the document such as the mention at various locations of the importance of looking at the effects of total reactive nitrogen versus NO_x. I know that there has been considerable discussion of the importance of using total reactive nitrogen in these analyses, but some of this usage and other areas of duplication should be reduced. A clearer transition and better linkage between Section 2.0 (OVERVIEW OF RISK AND EXPOSURE ASSESSMENT) and previous text needs to be provided.

In my detailed comments, I have provided a number of editorial corrections and suggestions. There should be consistency of whether or not to use direct references in the executive summary. I would suggest, unless there is a very compelling reason, that direct literature references should not be part of the Executive Summary.

There were no specific questions directed at Chapter 2, but the comments provided for the Executive Summary are relevant to this chapter. Much of this chapter is based upon an elaboration and justification of the use of ecosystem services in this assessment.

Air Quality Analyses (Chapter 3):

1. This chapter describes an approach for characterizing the spatial and temporal patterns of nitrogen and sulfur deposition in the case study locations including both oxidized and reduced nitrogen, and both wet and dry deposition of oxidized nitrogen, reduced nitrogen, and sulfur. Are the uncertainties associated with these analyses appropriately identified and described?

2. In response to CASAC's recommendation, the RSM analysis presented in the first draft Risk and Exposure Assessment was replaced by an analysis of results from a new series of CMAQ simulations designed to explore the relative contributions of NOx and NH3 emissions to total, reduced and oxidized nitrogen deposition and the relative contribution of SO2 emissions to sulfur deposition. Does this approach enable us to adequately examine the contribution of NOx to total nitrogen deposition?

3. The CMAQ application and model performance evaluation is presented in Appendix 1, as recommended by the Panel. Is this analysis sufficient to support the use of the model in this review?

Comments in progress

Case Study Analyses (Chapters 4 & 5)

Questions related to the individual case study analyses are presented below. Overarching questions across all the case studies include:

1. Are uncertainties appropriately characterized across the case studies? Is there adequate information to allow us to weigh the relative strengths of each case study to inform the standard setting process?

2. In using the Risk and Exposure Assessment to inform the policy assessment, we plan to focus on aquatic acidification as the basis for an alternative multi-pollutant secondary standard as this is the area where we have the most confidence in our ability to characterize adverse effects. Does the Panel agree with this approach?

Comments in progress.

Acidification:

1. Section 4.2 and Appendix 4 describe the analyses used to evaluate the effect of aquatic acidification. The analysis evaluates the ANC in selected lakes and streams in the Adirondacks and Shenandoah relative to three potential ANC cutoff levels (20, 50, and 100 ueq/L) to determine the impact of current levels of deposition in these areas as well as a larger assessment area. Is this data adequate to establish critical loads of deposition for the case study area?

2. The ecological effect function for aquatic acidification (section 4.2.7) attempts to

characterize the relationship between deposition and ANC. In order to estimate the amount of NOx and Sox deposition that will maintain an ANC level above a given limit requires the knowledge of the average catchment flux of base cation from weathering of soils and bedrock (i.e., preindustrial cation flux (BC0)). How might we generalize from location specific inputs (F-factor approach) to using this approach on a broader scale - watershed, regionally, or some other way - to generalize beyond individual locations? What other methods should be examined for estimating catchment weathering rates nationwide for surface acidity?

This section now does a better job of describing the importance of mobile anions with respect to soil acidification and resultant effects on the depletion of nutrient cations. Many of the figures in this section need to be redone so that legends, axis values, etc. are easier to read.

This section relies substantially on MAGIC model simulations to show various spatial and temporal trends. It needs to be clear which version of MAGIC is being used in these calculations since there are major differences with respect to the ability of the model to predict nitrogen watershed chemistry.

In this chapter the term "acidifying deposition" is used. I assume this is done to account for the role of ammonium inputs that can be nitrified resulting in acidification. However, this is not common terminology in the public policy and scientific literature and it may be preferable to use the more standard term "acidic deposition" throughout the document to avoid confusion.

Within the chapter the term "natural acidity" is used (e.g., pages 4-34 through 4-35). In some of the discussion related to ecological effects of acidification it is not clear what are the consequences associated with some of the assumptions such as the soils not being a sink for sulfur. We know that some soils are strong sulfur sinks and also there is considerable information that there is a net loss of sulfur from soils (e.g., soils as sulfur sources) (e.g., page 4-37).

In the section on uncertainty and variability (4.2.8) a variety of approaches are provided and these appear mostly to be associated with variation in parameter estimates and how this affects model output. Some discussion on the implicit limitations of the model used (e.g., processes not covered, appropriateness of scale both spatially and temporally, etc.) would help place this section in a broader context of the validity of the model results and any possible major limitations.

The section on ecosystem services is a good summary of helpful information related to ecosystem services and acidification issues (4.3.1.3). It is useful that the report explicitly states the problems of estimating directly how ecosystem services are affected by terrestrial and aquatic acidification.

3. Section 4.3 and Appendix 5 describe the analyses used to evaluate the effect of terrestrial acidification. This analysis uses the Simple Mass Balance Model to determine the impact of current deposition levels on Bc/Al levels relation to three potential Bc/Al cutoff levels (0.6, 1.2, and 10.0) for sugar maple in the Kane Experimental Forest and red spruce in the Hubbard Brook Experimental Forest and a larger assessment area based on the FIA database for 17 states. Is this approach adequate to develop critical loads of deposition for the broader terrestrial acidification case study area? Is the regression analysis between Bc/Al rations and tree health sufficiently described and are uncertainties adequately characterized?

In the sections looking at critical loads calculations there is considerable emphasis on the CMAQ model and its application and other issues related to deposition. I am not that there is sufficient balance between these issues and the other issues related to the within system elemental cycles including those processes affecting acidification.

The extrapolation of the critical load calculations for sugar maple and red spruce to other regions (e.g., different states) beyond those of the case studies helped place these results in a broader geographical context. There are some clear issues in looking at these results on state by state basis since these boundaries do not reflect the important characteristics that affect critical loads, but having this information on a state by state basis might be of more interest to policy makers. It is a surprising result that such a high percentage of sites have been compromised with the acidifying total nitrogen and sulfur deposition in 2002 (page 4-62).

In this section there is some use of the use of average critical loads related to three levels of projection. It is not clear if this "average" is meaningful in the context of how critical loads may be applied since the critical load is dependent on specific edaphic features of an area. Does the average take into account how the spatial distribution of edaphic features? Does this "average" apply to specific case study areas or to larger regions? The discussion and analyses that show how specific factors such as parent soil properties affect critical loads calculation is most important.

Nutrient Enrichment:

Comments in progress

Additional Effects (Chapter 6):

1. In this chapter, we have presented results from some qualitative analyses for additional effects including visibility, climate and materials, the interactions between sulfur and methylmercury production, nitrous oxide effects on climate, nitrogen addition effects on primary productivity and biogenic greenhouse gas fluxes, and phytotoxic effects on plants. Are these effects sufficiently addressed in light of the focus of this review on the other targeted effects and in terms of the available data to analyze them?

The section on methyl mercury formation and relationships to sulfur are generally adequate and well done. Some of the wording needs to be changed so that it is clear that this process can occur in areas beyond just surface waters.

The other sections including 6.3 NITROUS OXIDE, 6.4 NITROGEN ADDITION EFFECTS ON PRIMARY PRODUCTIVITY AND BIOGENIC GREENHOUSE GAS FLUXES (including subsections: 6.4.1 Effects on Primary Productivity and Carbon Budgeting; 6.4.2 Biogenic Emissions of Nitrous Oxide; 6.4.3 Methane Emissions and Uptake; 6.4.4 Emission Factors; 6.4.5 Uncertainty), 6.5 DIRECT PHYTOTOXIC EFFECTS OF GASEOUS SO_X AND NO_X [including subsections: 6.5.1 SO_2; 6.5.2 NO, NO_2 and Peroxyacetyl Nitrate (PAN),6.5.3 Nitric Acid (HNO_3)] are adequate in the context of the needs of the current report.

Synthesis of Case Studies (Chapter 7):

1. Here, the case study analyses are integrated and synthesized within the conceptual framework of ecosystem services as shown in Figure 7-2. Where possible, we have quantified select ecosystem services associated with the ecological effects targeted in this review. This chapter discusses adversity by characterizing the degree to which ecological effects are occurring under given levels of deposition to inform the discussion of adversity in the policy assessment and standard setting process. To what extent do you think the description of ecosystem services provides a useful framework in the case study analyses for informing standard setting? Does the Panel have suggestions for additional consideration or characterizations for ecosystem services related to the case studies?

2. Based on the information presented in the current Risk and Exposure Assessment, given adequate time and resources, is there enough information to inform setting separate standards based on the other targeted ecological effects, specifically, terrestrial acidification, aquatic nutrient enrichment, and terrestrial nutrient enrichment? If not, how can our understanding of these ecological effects be enhanced in time to inform the next 5-year review?

Comments in progress.

Detailed Comments and Responses to the Charge to the CASAC NO_x/SO_x Secondary Review Panel by Myron J. Mitchell

Page	Line(s)	Comment
xxiv	4-5	This statement makes no sense: "Ecosystem Structure: Refers to the species composition, distribution, and interactions with some abiotic attributes of the environment s they vary through space and time".
xxiv	21	Change "*This* indicator may either be the actual criteria air pollutant" to "*An* indicator may either be the actual criteria air pollutant".
xxv	2	Delete "As a result".
xxvii	3	Delete "reduced" twice in this line. Why use the term reduced?
xxvii	3	Give the charge for "NO_2" (nitrite) as "-".
xxviii	7	Change to "other *forms of* precipitation".
ES-2	12-13	Change "slower biomass growth" to "lower rates of production".
ES-2	14	Change "In addition to acidification, NO_x acts" to "In addition to contributing to acidification, NO_x acts".
ES-2	22-23	Change "the ecosystem is receiving more nitrogen than it uses" to "an ecosystem is receiving nitrogen in excess of biotic nutritional needs".
ES-2	23	Delete "also".
ES-2	24	Clarify what "This" refers to. Does this mean nitrogen deposition, primary productivity and/or terrestrial carbon cycling?
ES-2	26	Change to "Lichens".
ES-2	27-28	This statement seems out of place. Does this statement have any relevance to lichens? Clarify what aspects of biodiversity have been reduced in grasslands.
ES-3		Figure ES-1. Within the figure change "Soil Process" to Soil solute generation".
ES-4	9	Change "to determining when the" to "to determining when *and where* the"

ES-5	8	Replace “enrich” with “impact”.
ES-5	8	Clarify what “this” refers to.
ES-5	12	Change “while the Ecological Effect Function (box 6) relates the deposition metric into the” to “while the Ecological Effect Function (box 6) links the deposition metric to the relevant”.
ES-5	19	Change “the degradation of” to “deleterious affecting”.
ES-6	2	Change “Because ecosystems are diverse” to “Because ecosystems differ”.
ES-7		In Table ES-1 the Adirondack Mountains should be referred to as the “Adirondacks” and not the Adirondack”.
ES-8	9	Change “ecosystem services is being used as an umbrella term” to “the term ecosystem services is being used as a broad concept”.
ES-8	10	Change “It is a way to help explain” to “The evaluation of ecosystem services helps to explain”.
ES-9	3-4	Change “some of the ecosystem services likely to be affected are readily identified, while others will remain unidentified” to “ only some of the ecosystem services that are likely to be affected can be readily identified”.
ES-9	4-6	Change “Of those ecosystem services that are identified, some changes can be quantified, whereas others will remain unidentified” to “Of those ecosystem services that are identified, only subset of changes will likely be quantifiable”.
ES-9	6-7	Change “Within those services whose changes are quantified, only a few will likely be monetized, and many will remain unmonetized” to “For those quantifiable services only a few will be subject to monetization”.
ES-9	8	Change “A conceptual model integrating” to “An example of a conceptual model of effects on aquatic ecosystems is used to integrate”.
ES-9	12	Change “can be used to inform a policy judgment” to “can be used to in developing policy”
ES-9	16	Change “inform” to “provide”.
ES-9	18-21	Figure caption needs to be changed to indicate that this is an example focusing on aquatic ecosystems.

ES-11	3	The term “magnitude” can be misleading. The correspondence between the actual amount and relative spatial patterns of measured versus modeled concentrations needs clarification.
ES-12	7	Change “information about meteorology and land use in each grid cell of the domain” to “information about meteorology and land use both of which are critical components in affecting dry deposition”.
ES-13	5-7	Change “In the East, high levels of deposition exceeding 18 kg S/ha/yr occur in the immediate vicinity of isolated major sources, as well as in and near areas having a high concentration of SO_2 sources” to “In the East, the highest levels of deposition that exceeding 18 kg S/ha/yr occur in proximity to sources of high SO_2 emission” .
ES-15	13	Change “Acidification can degrade the health of terrestrial and aquatic ecosystems” to “Acidification can have deleterious impacts on terrestrial and aquatic ecosystems”.
ES-15	19	Delete “method”.
ES-15	28	Delete “the additional”.
ES-16	12-13	Change “direct relationship between ANC and fish and phyto-zooplankton diversity and abundance” to “direct relationship between ANC and the diversity and abundance of fish and phyto-zooplankton”.
ES-16	13	Change “MAGIC” to “The MAGIC model”.
ES-16		Within this page direct citations are provided. This does not seem to be consistent with other portions of the Executive Summary. I would suggest that these should be deleted for consistency. This problem is also found in other parts of the Executive Summary.
ES-17	9	Change “Calcium and Al are strongly” to “Calcium and Al concentrations are strongly”.
ES-17	13	See previous comments on the use of citations in the Executive Summary.
ES-17	18	Change “The tree species most sensitive” to “Tree species sensitive”
ES-17	19	Delete “a deciduous tree species”.
ES-17	20	Delete “a coniferous tree species”.

ES-17	22	Change “to” to “on”.
ES-17	23	Delete “both”.
ES-18	6	Change “total removal” to “total harvest”.
ES-18	6	Remove “from timberland”.
ES-18	9	Change “roughly” to “approximately”.
ES-18	10	Remove “from timberland”.
ES-18	11-12	Change “spruce forests are home t o the spruce-fir moss spider (endangered), the rock gnome lichen 12 (endangered), and the Virginia northern flying squirrel (delisted, but considered important).” to “spruce forests are important habitats for endangered species including the spruce-fir moss spider, the rock gnome lichen, and the Virginia northern flying squirrel (delisted, but still considered important).
ES-19	1-4	Change “Some organisms may at first respond positively to an initial increase in nutrients, exhibiting an increase in growth due to fertilization effects. However, as the nutrient load continues to rise, the imbalance can have negative effects either in the organism’s response or in the invasion of new organisms that benefit from increased nutrients” to “ Some organisms may at first respond to an increase in nutrients with increased growth. However, as nutrient load continues to rise, the resulting imbalance can have negative effects either directly on the organism or indirectly by the invasion of other species that are better competitors under high nutrient conditions”.
ES-19	12-14	Change “Nitrogen is an essential nutrient for aquatic ecosystem fertility, including lake, marine, and estuarine ecosystems, and is often the limiting nutrient for growth and reproduction in many of these ecosystems” to “Nitrogen is often a limiting nutrient for lake, marine, and estuarine ecosystems”.
ES-19	15	Delete “of a system”.
ES-19	21	Change to “nitrogen *enrichment* now represents”.
ES-19	26	Change “Due to the cascading impacts and effects of nitrogen enrichment” to “Due to the cascading impacts of nitrogen pollutants”.
ES-20	2	Change “estimation” to “estimate”.

ES-20 5	Change “In this assessment” to “In the current assessment”.
ES-20 14	Change “reductions in additional” to “reductions from additional”.
ES-20 15	Change “resident commercial species” to “resident commercial species important for various fisheries”.
ES-20 24-25	See previous statements about citations in the Executive Summary.
ES-20 29	See previous statements about citations in the Executive Summary.
ES-20 29	Change “that was only about” to “of only ~”.
ES-20 31	See previous statements about citations in the Executive Summary.
ES-21 2	Change “only source of nitrogen to these systems” to “the dominant source of nitrogen to these systems”.
ES-21 5-6	Change “;creating increased growth rates in some species over others, which changes competitive interactions among species; and nutrient imbalances” to “. This higher N availability affects the relative interspecific competitive of plant species resulting in changes in species composition and vegetation structure”.
ES-21 13-14	Change “to cause increased litter accumulation in the soils and carbon storage in aboveground biomass” to increased carbon storage in aboveground biomass and litter”.
ES-21 16	Change “can” to “may”.
ES-21 17	Change “by nitrogen limitation can now better compete and alter species dominance” to “by nitrogen limitation are more competitive”.
ES-21 19-20	Change “ the leaching of NO_3^- in soil drainage waters” to “soil NO_3^- leaching”.
ES-21 20	Change “in stream water” to “in surface waters”.
ES-21 22-23	Delete “; however, these measurements are not always widely available”.
ES-21 26	Change “that nitrogen” to “that increased nitrogen inputs”.
ES-21 28-29	Is the description on the “extent of ecosystems” or the extent of the ecosystems impacted by nitrogen deposition?

ES-22 3-6	Delete these lines.
ES-22 19-23	This seems like a rather detailed finding and could be deleted from the Executive Summary.
ES-23 8	Change “could be quite high” to “is quite high”.
ES-23 11-13	Change “enrichment potentially include decline in CSS habitat, decline in protection of native species, increase in abundance of nonnative grasses, and increase in wildfires” to “enrichment potentially include declines in CSS habitat the protection of native species, and increases in nonnative grasses and wildfires”.
ES-23 16	Change “helps regulate” to helps control”.
ES-23 17	Change “upset” to “disrupted”.
ES-23 19	Change “could” to “may”.
ES-24 1	Change “SO_x deposition on methylmercury production” to “SO_x deposition and resultant change in soil and wetland SO_4^{2-} concentrations in affecting methylmercury production”.
ES-24 3	Change “scope of this review” to “scope of the current review”.
ES-24 9-10	Change “While there are many uncertainties associated with these analyses, from a scientific perspective there is confidence that known or anticipated adverse ecological effects are occurring” to “ Although uncertainties exist, there is strong evidence that known or anticipated adverse ecological effects are occurring”.
ES-24 12-13	Change “Of all the case study analyses, there is most confidence in the ecological responses, effects, and benefits associated with aquatic acidification” to “Within the case study analyses, there is most confidence in the ecological responses, effects, and the deleterious impacts associated with acidic deposition”.
ES-24 13-14	Change “and there is a fair amount of confidence about those associated with terrestrial acidification” to “Similarly, the importance associated with the impacts of acidic deposition on terrestrial systems is clearly documented”.
ES-24 15	Change “benefits” to “deleterious impacts”.

ES-24	18	Delete “However”.
ES-24	20	Change “only” to “dominant”.
ES-24	22	Change “benefits” to “deleterious impacts”.
ES-24	25	Change “terrestrial acidification may be the most useful in terms of developing a secondary” to “terrestrial acidification should be most useful in developing a secondary”.
ES-25		These specific citations should be removed f rom the Executive Summary.
1-2	2	Change “The species of nitrogen and sulfur” to “The chemical species of nitrogen and sulfur’.
1-2	5-7	Change “because NO_x, SO_x, and their associated transformation products are linked from an atmospheric chemistry perspective, as well as from an environmental effects perspective” to “because the atmospheric chemistry and environmental effects of NO_x, SO_x, and their associated transformation products are linked”.
1-2	10	Change “of these two pollutants has been conducted” to “of SO_x and NO_x as well as total reactive N has been conducted”.
1-2	11	Delete “at this time”.
1-2	16	Change “in an ecologically meaningful way” to “that is ecologically meaningful”.
1-3	1	Change “see” to “go to:”.
1-4	13	Why “identical”?
1-4	22	Change “This draft document” to This latter draft document”.
1-5	28	Change “At that time, EPA was aware that SO_x have” to “At that time, EPA was aware that SO_x has”.
1-6	1	Change “specific SO_x concentrations” to “specific atmospheric SO_x concentrations” .
1-8	8	Change “at that time” at the time of the report (1995).
1-8	14	Change “particular relevance to this review” to “particular relevance to the

		current review".
1-10	13-21	Should some mention be made of organic forms of N in the atmosphere including DON? It is noteworthy that in Figure 1.3-1 (page 1-11) that organic forms of N are shown
1-11	9-15	Although the figure shows some of the organic atmospheric S forms. There is no mention of these chemical species in the text.
1-12	19	Change "not high enough" to "not sufficiently high".
1-13	6	Change "Both are essential and sometimes limiting, nutrients needed for growth and productivity" to "Both N and S are essential macronutrients".
1-13	7	Change "Excess" to However, excess".
1-13	1	Change from "These effects include slower growth" to "These effects include slower biotic growth".
1-13	23	Change "Models suggest that" to "Models for the latter study area suggest that".
1-13	28	Change "acidification effects from acidifying deposition" to "acidification effects from atmospheric deposition".
1-14	9	Change "that leads" to "that may lead".
1-14	21	Change "quality in the western United States (U.S. EPA, 2008, Section 3.3" to "quality in the western United States, a region especially sensitive to increased nitrogen atmospheric inputs (U.S. EPA, 2008, Section 3.3"
1-14	23	Change "which leads to eutrophication" to "which may lead to eutrophication".
1-14	27	Change "in highly eutrophic estuaries" to "in some eutrophic estuaries".
1-14	30-31	Change "In terrestrial ecosystems, there are multiple chemical indicators for the alteration of the biogeochemical cycling of nitrogen that is caused by total reactive nitrogen deposition" to "In terrestrial ecosystems, there are multiple chemical indicators that the biogeochemical cycling of nitrogen has been altered by the deposition of total reactive nitrogen".
1-14-15		Change "Nitrate leaching" to "Nitrate leaching from terrestrial ecosystems".

1-15	2	Change “the onset of leaching” to “the atmospheric deposition threshold for nitrate leaching”.
1-15	7	Change “occurring at 3 kg” to “occurring at atmospheric inputs as low as 3 kg”.
1-15	13	Change “this” to “the current”.
1-15	19	Change “In watersheds where changes in sulfate deposition did not produced an effect” to “In watersheds where changes in sulfate deposition did not result in changes in methylmercury generation”.
1-15	20	Change “meaningful” to “substantial”.
1-16	Figure 1.3-3	See previous comments on Figure ES-1 (Is this figure the same as Figure 1.3-3?)
1-17	18-20	This sentence needs to be reworded.
1-18	15-17	This sentence is confusing. What is meant by “the uncertainties in the estimated reductions”? Is the uncertainty on the amount of atmospheric reduction that will occur or uncertainty on the effects of reductions?
1-19	21	Change “Identifying important chemical species in the atmosphere” to “Identifying important N and S chemical species in the atmosphere”.
1-21	6	Change “All of Figure 1.4-1" to “All of the components of Figure 1.4-1" .
2-6	14-15	Change “to total loadings of in the environment” to “to the combined atmospheric loadings of both elements”.
2-7	7	Change “a broad look into the” to “an overview of”.
2-7	8	Change “services that is one tool that can help link” to “services. The analysis of the effects on ecosystem services will help link “.
2-7	10-11	Change “In this Risk and Exposure Assessment, ecosystem services is used as an umbrella term to aid in describing the impacts of ecological effects on public welfare and to help explain how” to “In this Risk and Exposure Assessment, ecosystem services is used to show the impacts of ecological effects on public welfare and help explain how” .
2-8	3	Change “data were not abundant enough” to data were not sufficient”

2-8	19-20	This sentence seems out of place.
2-15	2	Does the statement "The analysis of ecosystem services for the aquatic acidification focused on recreational fishing" indicating that focus for the current assessment of the general analysis of ecosystem services in aquatic ecosystems. I believe the sentence should be changed to: "The current assessment the analysis of effects on ecosystem services from aquatic acidification focused on recreational fishing".
2-15	24	Change "little data is" to "little data are".
2-16	2	Change "ecosystems are addressed" to "ecosystems were addressed".
4-1	26-27	Change "Under natural conditions (i.e., low atmospheric deposition of nitrogen and sulfur), the limited mobility of anions in the soil controls the rate of base cation leaching" to "Under conditions of low atmospheric deposition of nitrogen and sulfur, the naturally produced bicarbonate anion is often the dominant mobile anion with SO_4^{2-} and NO_3^- playing a limited role with respect to cation leaching".
4-1	27-30	Change "However, acidifying deposition of nitrogen and sulfur species can significantly increase the concentration of anions in the soil, leading to an accelerated rate of base cation leaching, particularly the leaching of Ca^{2+} and Mg^{2+} cations" to "Increased atmospheric deposition of sulfur and nitrogen can result in marked increases in SO_4^{2-} and NO_3^- soil fluxes resulting in the concomitant leaching of nutrient (Ca^{2+}, Mg^{2+}) and toxic (Al^{n+} and H^+) cations".
4-2	8	Change to "Criteria for case study selection".
4-2	11	Change "Current conditions for other areas" to "Current conditions for these other areas".
4-2	26	Change "sum of soil and water processes that occur upstream within a watershed, it also reflects the results of watershed-scale terrestrial effects, including nitrogen saturation, forest decline, and soil acidification (Stoddard et al., 2003)." to "sum of terrestrial and aquatic processes that occur upstream within a watershed. Important terrestrial processes include nitrogen saturation, forest decline, and soil acidification (Stoddard et al., 2003)".
4-3	8	Change "certain" to "some".

4-3	10	Change “where strong acids are deposited into the soil” to “where strong mineral acids (e.g., H_2SO_4 and HNO_3) are deposited or generated within the soil.
4-2	13-14	Change “inorganic Al can become mobilized, leading to the leaching of Al into soil waters and surface waters” to “inorganic Al can be mobilized, leading to the leaching of Al from soils to surface waters”.
4-2	15	Change “differently” to “differently to acidic deposition”.
4-2	15-16	Change “on sensitive species” to “on different ecosystems and species”.
4-2	20	Change “migrates” to “leaches”.
4-3	23	Change “maintains the balance of electric charge” to “maintains electroneutrality” .
4-4	1	Delete “further”.
4–4	8	I would disagree that episodic acidification is more important than chronic acidification.
4-4	7-8	Change to “ Short-term (i.e., hours or days) episodic changes in water chemistry have perhaps the most significant biological effects” to “Short-term (i.e., hours or days) episodic changes in water chemistry have important biological effects”.
4-4	9	Change “rainstorms or snowmelt” to “precipitation or snowmelt events”
4-4	10	Change “which tends to provide less neutralizing of atmospheric acidity as compared with” to “than tends to provide less acid neutralizing than water passing through”.
4-4	12	Change “storm runoff or snowmelt” to “events”.
4-5	9-10	Change “receptors” to “parameters”.
4-5	13-14	Delete “Although ANC does not relate directly to the health of biota” and start sentence with “The utility”.
4-5	22	Delete “the”.
4-5	24	Change “Low ANC concentrations have” Low ANC has”.

4-5	30	Change “(Figure 4.2-1, a), which” to “(Figure 4.2-1, a) that”.
4-6	3	Change “has been found in studies” to “has been found in various studies”.
4-6	8-9	Change “Below 100 μeq/L, it has been shown that fish fitness and community diversity begin to decline” to “Below 100 μeq/L ANC fish fitness and community diversity begin to decline” .
4-6	11	Delete “decline; however, the overall health of the community remains good”.
4-6	13	Change “that are sensitive to negative effects on biota that are sensitive to acidification” to “that are sensitive to acidification”.
4-7	2	Change “had to have” to “need to have”.
4-7	9	Delete “primarily”.
4-17	17	Change “fishers” to “fisherman”–this may not be a “sex neutral” term, but fishers refers to a type of animal.
4-8	2	Delete “in these states”.
4-8	13-14	Change “services, such as hydrological regime regulation and climate regulation” to “services associated with hydrology and climate”.
4-8	15	Delete “specific”.
4-8	16-17	Change “delicate aquatic food chains” to “ aquatic food webs” .
4-8	19	Delete “it is worth noting that”.
4-8	20-22	Delete “For example, these biological control services may serve as “intermediate” inputs that support the production of “final” recreational fishing and other cultural services”.
4-9	4-5	Change “The regions of the United States with low surface water ANC values are the areas that are sensitive to acidifying deposition” to “The regions of the United States with low surface water ANC values are sensitive to acidifying deposition’.
4-9	6-7	Delete “at their existing ambient concentration levels”.

4-9	14	Change “surface water data” to “analyses of sulfur waters”.
4-9	19	Delete “are estimated to”.
4-9	21-24	Change “In 2002, Stoddard et al. (2003) took another comprehensive look at the level of acidification within all of these regions. Although improvement in ANC occurred, about 8% of lakes in the Adirondack Mountains and 6% to 8% of streams in the northern Appalachian Plateau and Ridge/Blue Ridge region were still acidic at base-flow conditions” to “Stoddard et al. (2003) suggested that although improvement in ANC had occurred ~8% of lakes in the Adirondack Mountains and from 6% to 8% of streams in the northern Appalachian Plateau and Ridge/Blue Ridge region were acidic at base-flow conditions”.
4-10	9-12	Change “After considering this information, the Adirondack Mountains and the Shenandoah Mountains (referred to in this chapter as Adirondack and Shenandoah case study areas, respectively) were selected. The rationale for choosing these two case study areas is described in the following subsections” to “ Using the rationale described in the following subsections the Adirondack Mountains and Shenandoah Mountains were selected for case study areas”.
4-10	16	Change “The case study area” to This area”.
4-10-11		Delete “, which all draw water from the preserve”.
4-12		For Figure 4.2-3, the axis legends and numbers are too small.
4-13		For Figure 4.2-4, the axis legends and numbers are too small.
4-14	6-7	Delete “because it can no longer be measured”.
4-14	7	Change “Likewise, it is also difficult to determine” to “Likewise, it is also difficult to empirically determine”.
4-15	9	Change “hydrological” to “biogeochemical”–MAGIC is not a hydrologic model.
4-15	10	Change “quality levels” to “chemistry”.
4-15	4-30	The insert on critical loads includes the value of 50 meq/m^2 yr. This value may be confusing in using a load based upon charge versus mass since much of the proceeding discussion including inputs used mass values.
4-17	19	Change “the condition” to “the modeled condition”.

4-18	5-8	It is important to mention that although SO_4^{2-} still dominates the relative importance of NO_3^- is increasing substantially. Also, comparing concentrations of SO_4^{2-} and NO_3^- in surface waters can be misleading since there may be substantial losses of NO_3^- due to biotic processes in watersheds.
4-18	13	Were these declines in Al statistically significant?
4-18	14	Change "significant" to "substantial".
4-19	1-5	Reword this sentence it makes not sense.
4-19	7	Change "is" to "was".
4-19	10	Change "are" to "were".
4-21	2-5	Change "Percentage of Adirondack Case Study Area lakes in the five classes of acidification (i.e., Acute, Severe, Elevated, Moderate, Low) for years 2006 and 1860 (preacidification) for 44 lakes modeled using MAGIC. Error bar indicates the 95% confidence interval" to "Percentage of Adirondack Case Study Area lakes in the five classes of acidification (i.e., Acute, Severe, Elevated, Moderate, Low) for years 1860 (preacidification) and 2006 for 44 lakes modeled using MAGIC. Error bar indicates the 95% confidence interval". (Make similar changes in other figure captions including 4.2-19)
4-19	8-11	Change "Sites labeled by red or orange dots have less buffering ability than sites labeled with yellow and green dots, and hence, indicate those lakes that are most sensitive to acidifying deposition, due to a host of environmental factors" to "Sites indicated by red or orange circles have less buffering ability than sites labeled with yellow and green circles, and hence, indicate those lakes that are most sensitive to acidifying deposition".
4-22	2-7	In figure caption change "dots" to "circles". *Make similar changes in all figure captions and text.*
4-23	7-9	Change "In considering the future responses of lakes to current emissions and given the current condition of the lakes, the question becomes whether lakes can recover to healthy systems (i.e., ANC > 50 μeq/L)" to "In considering the future responses of lakes, the question becomes whether lakes can recover to healthy systems (i.e., ANC > 50 μeq/L) under current levels of deposition".

4-24	4-11	Change “Based on a deposition scenario that maintains current emission levels to years 2020and 2050, the simulation forecast indicates no improvement in water quality”.“Based on a deposition scenario that maintains current emission levels to up to years 2020 and 2050, the simulation forecast indicates no improvement in water quality over either of these periods”.
4-24	15	Change from “will likely not improve the acidification of lakes” to “will not likely improve the recovery from acidification”.
4-24	15-17	Delete this sentence.
4-24	24-25	Change “At this time, it is unclear why ANC initially improved and is now declining” to “It is not known what has caused this temporal pattern of ANC in this case study”.
4-25	Table 4.2-4	Indicate what “+/-“ columns signify.
4-26	3	Change “changed statistically” to “did not significantly differ”.
4-26	19	Change “industrially generated acidifying deposition” to “acidic deposition”.
4-26	21	Change “is” to “was”.
4-31	3	Change “Based on a deposition scenario that maintains current emission levels to 2020 and 2050" to Based on a deposition scenario that maintains current emission levels to years 2020 and 2050".
4-31	12	Change “country” to ”U.S.”.
4-31	13	Change “across populations” to “across various populations”.
4-31	14	Change “picked” to “selected”.
4-31	15	Change “to make estimates of regional extent of condition (e.g., number of lakes, length of stream)” to “to make regional estimates of surface water conditions”.
4-31	21	Change “to be susceptible” to “to be especially susceptible”.
4-32	24	Change “area” to “areas”.

4-32	32	I don't believe the term "ecoregion" been defined in the document.
4-28	2	Change "industrially generated acidifying deposition" to "acidic deposition".
4-34	10	Change "SO_2" to "SO_x".
4-34	13	Change "One hundred 17 lakes of the 169 lakes modeled for critical loads are part of a subset of 1,842 lakes in the Adirondack Case Study Area" to " Of the 169 lakes modeled for critical loads, 117 of these lakes were within 1,842 lakes in entire the Adirondack Case Study Area".
4-34	14-15	Delete "which include all lakes from 0.5 to 2,000 ha in size and at least 1 m in depth".
4-34	21	Change to "13% *of the total population*".
4-34	22	Change "some lakes would have never had ANC" to "some lakes would have never had ANC".
4-34	24-25	Change "estimate based on the critical load alone" to "estimate based solely using the critical load criterion".
4-34	26	What is meant by "natural"? Does this refer to current conditions or preindustrial concentrations? The term natural with respect to surface water acidity needs to be defined. It might be clearer to discuss the role of DOC in these waterbodies that have historically low ANC.
4-35	20	Change "the same" to "similar".
4-36	12	Change "a host of catchment processes and environmental factors that affect the level of base cations (e.g., Ca^+, Mg^+) concentrations and the sinks of nitrogen and sulfur in the lake and terrestrial catchment" to "a series of biogeochemical processes that produce and consume acidity in watersheds".
4-36	19-21	Change "Although ANC does not directly affect the health of biotic communities, it ameliorates acidity-related biotic stress that provides an "ecological indicator" of overall integrity of the ecosystem" to "Although ANC has not generally been used as a parameter for predicting the health of biotic communities, it provides useful information of the potential acidity-related biotic stress and hence is a useful "ecological indicator".
4-36	22	Delete "then".

4-37	9-10	Change "To convert surface water concentrations into surface water fluxes, multiply by runoff (Q) (in m/yr) from the site" to Surface water concentrations are converted to fluxes by multiplying concentrations by runoff (Q) (in m/yr)".
4-37	12	Change "between plants and soil is ignored" to "between plants and soil is negligible".
4-37	19-21	This sentence is confusing. Certainly the nitrogen and sulfur biogeochemical fluxes and transformations affect acidity.
4-40	1-3	This is not just a problem for the United States. The estimate of weathering rates (including the generation of base cations) is a major limitation for many biogeochemical analyses and interpretations
4-42	22	Delete "a coniferous tree species" and " a deciduous tree species".
4-48	6	Delete "and forest".
4-62		The following statement "Collectively, these results suggest that the health of at least a portion of the sugar maple and red spruce growing in the United States may have been compromised with the acidifying total nitrogen and sulfur deposition in 2002; even with the lowest level of protection, half the states contained sugar maple and red spruce stands that were negatively impacted by acidifying deposition" will receive considerable attention. It is important that any caveats be provided on these results so that the interpretation is placed in the most complete picture of the state of the science.
4-65	10	Change "was" to "is".
4-66	17	Does the "average critical loads" have any real meaning in the context of setting critical loads. I would suggest that the range is the most important and demonstrates and clearly shows how edaphic factors can have a major influence on critical loads.
4-67		Figure 4.3.9 See previous comment with respect of providing the values for average critical loads.
4-68		Figure 4.3.10 See comment above on the use of average critical loads in this figure.

4-69		The discussion of uncertainty is important and highlights some of the issues related to the actual calculation of critical loads. It may be over stretching the uncertainty analyses to suggest that "If all or a large majority of estimates indicate that the critical load of a system is exceeded with current total nitrogen and sulfur deposition rates, the probability is high that deposition is greater than the critical load and that the trees and vegetation in that system are being negatively impacted by acidification". The use of the term "probability" seems out of place and suggests that this approach has a stronger statistical underpinning than is the actual case. The key factor is what are the range of values that affect these calculations and how confident are we in using these values in making these calculations. Similarly the term "certainty" would suggest more confidence in these estimates than may actually be the case.
4-71	3-5	This type of calculation in which it is clearly shown how different values can be obtained for critical loads based upon specific edaphic factors (e.g., parent material acidity) is a useful approach and show how this factor can have a dramatic impact on these calculations.
6-2	31-32	Change "aquatic environments" to "aquatic and terrestrial environments, including wetlands,"
6-2	33	Delete "surface water".
6-11	1	Change "emissions" to "deposition".
6-12	24	Change "its global warming potential" to "its global warming potential per molecule".
6-14	14-15	Change "Nitrogen deposition can affect the patterns of carbon allocation because most growth occurs above ground" to "Nitrogen deposition can affect the patterns of carbon allocation between above and below ground production".
6-14	15	Change "This increases the shoot-to-root ratio" to "Increased nitrogen availability increases the shoot-to-root ratio".
6-11	20	Change "Reducing SO_x" to "Reducing SO_x emissions".

Dr. Ted Russell

Review of EPA's Risk and Exposure Assessment for Review of the Secondary National Ambient Air Quality Standards for Oxides of Nitrogen and Oxides of Sulfur – Second Draft

First, I found the second draft of the REA to be improved over the first draft, being more complete and strengthened. EPA was largely responsive to CASAC's comments on the First Draft REA. While there are areas that could be further strengthened, the current version provides the type of scientific information needed to support further review of the secondary NAAQS for SOx and NOx. The one area that needs the most strengthening is in the area where EPA is "stepping out" and considering approaches that would set standards that account for multiple pollutants (in this case, NOx and SOx, which in this case includes oxidized species beyond just NO2 and SO2, as discussed in the REA), and would also account for the presence and effects of a non-criteria species (reduced nitrogen) for which a NAAQS is not being set. The directions being explored by EPA is directly responsive to our scientific understanding of the system, and the staff is to be complemented for the progress they have made in developing their proposed approaches and the underlying analysis. This approach is directly in line with recent recommendation of various CASAC and National Academy Panels. In addition to being very supportive of the multipollutant approach, the Executive Summary is welcomed, as is the statement of Policy Relevant Questions. Also, I like Figure ES-2 (or 2.3-1) and how it is used throughout the document. I found the case studies enlightening and generally well done.

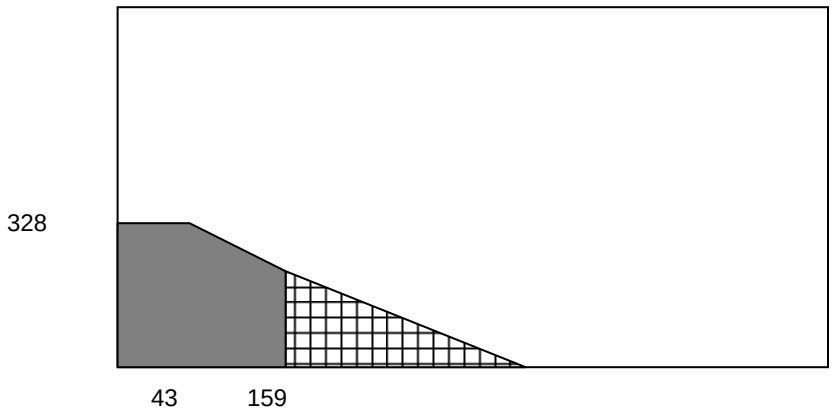
My first primary comment and overriding concern is that the novelty and complexities of having multi-pollutant standards warrant considerable thought, work and review. While I view that EPA staff have made considerable progress – in many ways remarkable progress – I am concerned about the looming schedule and fear that the final thrust will be rushed and there is inadequate time for refinement, review and response. The current REA lays a foundation for how a set of standards can be set, but it has not dealt adequately with some of the complexities, and I am not sure if the few months between this review and the review of the Policy Assessment. In particular, linking the depositional loads to atmospheric concentrations needs to be explored further, and how one might formulate a an air quality standard (i.e., specific to an ambient concentration, including level, form, averaging time, etc.) has not been fleshed out. This will be an initial and important foray in to a multipollutant standard and adequate resources (in this case time) should be allowed such that they get it mostly right.

My primary criticism of this draft, as indicated above, is that it does not adequately deal with linking the depositional load calculations with the atmospheric concentrations implied (or that would be linked to those depositional loads), and how those concentrations vary spatially. While the charge memo states that this has been left for the Policy Assessment, but I believe that such information belongs here. There is certainly quite a bit of science involved in constructing this linkage and this should be reviewed separate from its application as part of the policy analysis. Scientific questions related to risk and exposure include: What is the geographic variability?

What are the risks (fraction of areas that would be subject to ecologically unhealthy depositional loads) associated with varying levels of the standards (particularly given the geographic variability)? How strong is the relationship between ambient concentration measurements and the resulting depositional load?

I was a bit surprised that the Executive Summary did not go in to much greater depth of the material dealing with the Critical Load Function (CLF). This is a major component of how a standard might be formulated, and it belongs in the ES, along with ample discussion of where it comes from and how it links oxidized N and S deposition, and accounts fro reduced N loading as well.

I am a bit confused by Figures 4.3-7 and 4.3-8. For example, in Figure 4.3-8, CLmin(N) is 43, so I would think that the line starts to drop at that point, in spite of what the reduced N loading is, such that the final figure would look like (excuse the artistry):



While I very much appreciated the Policy Relevant Questions being laid out up front, the Executive Summary, and the REA, does not close the loop and answer them (or say that the question has not been answered in the REA and identify where/when/if it will be answered). I was hoping to see this at the end of the ES.

In Chapter 3, I was hoping to see a plot of the annual averaged S and oxidized N deposition divided by the annual average SO2 and NO2 concentrations as simulated by CMAQ to help assess how variable is the relationship between ambient concentrations of the two likely indicator species and depositional loads.

In Chapter 7, I was hoping for more integration across effects. Particularly for a secondary standard, how the various endpoints of concern might add up is important. What I was hoping for is a high level view of what effects are of most concern where, which components are of most concern and where, when acidification and nutrient enrichment are of similar importance,

the response to emission reductions, etc.. I was looking for how various ecosystems would respond to controls associated with reducing ambient SO2 and NO2 levels, and that the chapter would take an ecosystem or regional perspective, in addition to an effect perspective. Table 7.3-1 and Figure 7.1-3 are steps in this direction, though a broader picture was desired.

Specifics (some are suggestions to improve presentation):

Up front material: Please include authors and contributors, preferably noting who worked on which sections.

Page ES-1 Call out box: Should not NH4NO3 be added in the first paragraph?
Page ES-1, footnote: "this" is awkward since you were just talking about the primary reviews.
ES-3,4: Is the list of bullets sufficient? Just asking for now.
ES-5, line 20: Might a better term be found than "maximum depositional load" since the load experienced might actually larger? Possibly maximum depositional threshold?
ES-6, line 4: replace "addresses" with "focuses primary on"
ES-6, line 8.5: Add bullet: Other impacts discussed include: with sub-bullets such as climate, visibility, etc.
ES-8, line 5: replace "addresses" with "providing information necessary to inform setting"
ES-8: I would put "ecosystem services" in "" at this point since you are using it as an umbrella term. (remove the "," after term as well).
ES-10 (and related sections of the report): The REA should be very specific in terms of how emissions and deposition al loads are presented, in this case when presenting emissions, the values should be presented in tons of N (and tons of S), and specified as such, for more ready comparison. Also, metric units are preferred.
ES-10, line 12: rephrase to "…, may appear relatively small by comparison to emissions of NOx and SOx, but are important, particularly in some regions" (NH3 impacts have more than a local footprint)
ES-10, Line 20… The section on "Ambient Concentrations" needs a better lead in and should begin with observations, not model results. If you are only going to present model results, use "Simulated Ambient Concentrations", and should note how simulated values compare with observations.
ES-13, line 3: "… sulfur deposition across…"
ES-14, line 11. Add "Deposition rates associated with PRB levels contribute minimally to the fluxes above those from natural background."
Es-15 line 12: add the closing ")"
ES-16, line 27. How does "sensitive or at risk" map on to, line 19-20 aquatic status categories?
Table ES-2 I think the = should be replaced with "≤"
ES-21, line 17: replace "now" with "then"
Table ES-4. The final row appears to be inconsistent with ES-23, line 1.
ES-24, line 10: change to "… there is strong scientific agreement that …"

1-4, line 28: AQCD should be in parentheses after "Air Quality…"

1-5 Line 18: AQCD (switch C and D.
1-11, lines 12-14: NOx does not combine with water to form nitric acid (or are you talking about gaseous HNO3 becoming aqueous?). Also, this section is on sulfur species, so it may be out of place.
1-16: The first paragraph of the section on "Policy Relevant Questions" focuses on NOx. Broaden the opening sentences of the paragraph to include SOx.

2-12, lines 14-15: Additional explanation of "the use of the common currency of energy" would be appreciated.
2-15, line 2: "…acidification case study focused …"

3-2, line 9 : "NH" got dropped out.
3-2, line 15: H2SO4 is also present at relevant levels.
3-3, line 10: add HO2 and remove the "-" from OH (add a dot if you wish).
3-9, line 13: "… locations of major SO2 emitters." (SO2 emissions are ubiquitous.)

Figures 3.3-6 and 3.3-14: each of these panels can be done in one graph, e.g., using a different color for each season. Also, it is not apparent that the seasonal information is used that much later on.

3-59, line 22: The 2002 value has to be in the range of values observed from 1998 to 2007.
3-81, lines 1-3: Are these in tons N and S or tons of the pollutant emitted? Again, metric tons N or S is preferred.
3-82, line 5: NH is missing
3-82, line 8: NH is missing

4-4, Call out box: You shold not only indicate the species, but what happens to them.
Table 4.2-2 add "(and associated uncertainties)" to the caption
4-17: line 7: "… NOx, and their atmospheric reaction products, have …"

Figure 4.3-1: I would switch the axes.

7-1, Line 21: "the clause "or that society views as beneficial." is awkward as used in that effects may also be disbeneficial.