16 July 2009 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:

Dr. Andrzej Bytnerowicz **2**
Dr. Naresh Kumar **5**
Mr. Rich Poirot **9**
Mr. Dave Shaw **17**

Dr. Andrzej Bytnerowicz

Executive Summary

Does the Executive Summary adequately summarize and characterize the key issues driving this review as well as the important findings of the analyses?

Generally, it does. The section is well written and provides a good overview of the entire document. Graphics, maps and tables are informative and useful. However, some corrections are needed (see my replies to the next question).

Does the Panel have any suggestions for clarification or refinement of the Executive Summary?

a. the second paragraph in the box of page ES-1 should be corrected to: "Substances known as oxides of sulfur, or SOx, include multiple gaseous substances (e.g., sulfur dioxide [SO_2], …."

b. page ES-2 – in the end of the last paragraph a statement that biodiversity changes have also been observed in other ecosystems, such as coastal sage scrub, mixed conifer forests in California or alpine ecosystems of the Rocky Mountains, should be added.

c. page ES-11, text and Figure ES 5 – while the text described NOx, the figure shows NOy. Consistency is needed here. On the same page, line 3, change "Peak SO_2 concentrations" to "Highest annual SO_2 average concentrations".

d. page ES-18, lines 13-16 - this paragraph seems to be out of place. Consider deleting.

e. pages ES-21 through ES-23, section "Terrestrial Nutrient Enrichment" – references should be added to this section.

f. page ES-23, Table ES-4 – zeros for N deposition > 17 kg N/ha/yr do not make sense. Please change to some meaningful values.

Case Study Analyses

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?

Yes. For all described cases (acidification of aquatic and terrestrial ecosystems; nutrient enrichment of aquatic and terrestrial ecosystems) uncertainties have been well described and examples of potential problems have been given. A good example is a statement on page 5-41 describing uncertainties related to the SPARROW model: estimates of N loading based on the CMAQ/NADP overall N loading in the model calibrated against the wet nitrate deposition values. Similarly, for the nutrient enrichment case studies for the terrestrial ecosystems, problems and uncertainties related to air pollution monitoring techniques (such as for ammonia),

those due to limitations in the spatial extent and density of monitoring networks, or uncertainties due to the design and performance of the CMAQ model are listed.

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?

Yes. This research is most advanced and well documented and therefore I agree with such a selection.

Nutrient Enrichment

1. Section 5.2 and Appendix 6 describe the analyses to evaluate the effect of aquatic nutrient enrichment. The analysis uses the SPARROW model on the stream Reach 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?

The SPARROW model and its links to the CMAQ model that provides deposition data and to the ASSETS EI that estimates a likelihood of the current or future occurrence of eutrophication are well described in the Appendices. A better description of the SPARROW model is needed in the main document. Therefore I suggest that most of the information contained on pages 6-31 through 6-33 and Figure 2.2-3 of the Appendices is copied into Chapter 5 of the main document. Otherwise a reader has to read both volumes to understand the SPARROW model and the entire methodological approach.

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 in California and in mixed conifer forests in the San Bernardino and Sierra Nevada Mountains and larger areas where possible. In addition, the effects of nitrogen deposition in the Rocky Mountain National Park supplemental case study location are summarized. How would the Panel apply the threshold values presented in this case study to allow for a broader geographic application that accounts for regional variability? Have the associated uncertainties been adequately characterized?

The proposed approach of an assemblage of a "patchwork quilt" of species and ecosystem types from across the United States is a reasonable and probably the best option considering a general scarcity of data on the ecological effects of atmospheric N deposition. Research results from the ecosystems selected for the case study (coastal sage and mixed conifer forests), as well as from alpine ecosystems in the Rocky Mountains, north-eastern forests, and mid-western grasslands, provide a reasonable representation of the continental US ecological zones, and a high probability that comparable responses would take place in other zones. A range of benchmarks is based on the well characterized sensitive ecological indicators such as changes in lichen communities, especially decrease of nitrophytes at the low end of the sensitivity spectrum; changes in mycorrhizal associations in the mid-range of the spectrum; and nitrate leaching to streams at high levels of atmospheric N deposition. Changes in lichen communities seem to be the most promising N deposition indicators of N deposition effects which could be used nationwide - comparable changes of epiphytic lichens in various ecological zones can be

expected at the similar levels of N deposition since such lichens get their nutritional N from the atmosphere. However, when considering responses of lichens to N deposition, also other factors such as phytotoxic ozone or climatic differences have to be taken into account. Changes in higher plants used as sensitive indicators of N deposition would be characterized by much larger margins of uncertainty because of the edaphic, climatic and many other differences in ecosystems or ecological zones in the US.

The main question still remains – how to translate changes caused by the total nitrogen deposition just into the ambient concentrations of the criteria pollutant (i.e., NOx). Since ecological indicators included in this analysis, as well as the biological systems in general, react to the total reactive N (including its reduced inorganic and organic forms), this seems to be the most important problem to solve.

Additional problem is related to the N deposition data produced by the CMAQ model. Improvements of the model are needed in order to include N organic species in total N deposition estimates as well as the more recent, finer resolution data.

Other Comments

Page 5-30, Figure 5.2-12 – please check the legend. Number of a group should be assigned to each color. Number of estuaries is already listed in Table 5.2-2 and should not be duplicated in the table.

Page 5-32, line 6 – change “reductions” to “decrease”.

Page 5-34, Figure 5.2-15 – drawing a line through these data points is highly problematic.

Page 5-55, lines 3-8 – there is also a need for improving the dry deposition methodologies through more robust models aided by empirical, ground level estimates based on well developed monitoring networks.

Page 5-66, line 11 – in contrast to fire suppression policy and increased atmospheric N deposition, ozone does not contribute to increasing stand density.

Page 5-72, line 3 – add “of exotic species” after “invasion”.

Page 5-79, line 11 – check the units.

Page 5-81, line 20 – add “especially in remote areas” after “networks”.

Page 5-85, lines 10-12 – reference has already been cited above.

Pages 6-29 through 6-42 – check references, many cited in the text are missing.

Dr. Naresh Kumar

The second draft of the Risk and Exposure Assessment (REA) document is much improved from the first draft. However, there are still some areas that need improvement and further analyses that need to be performed. My major comments, with a main focus on the air quality analysis, are:

1. Figure 1.4-1 (reproduced below) appears as the key figure in the REA document. As noted, the Atmospheric Deposition Transformation Function depicted in Box 3 "quantifies the relationship between atmospheric concentrations and deposition of NOx and SOx".

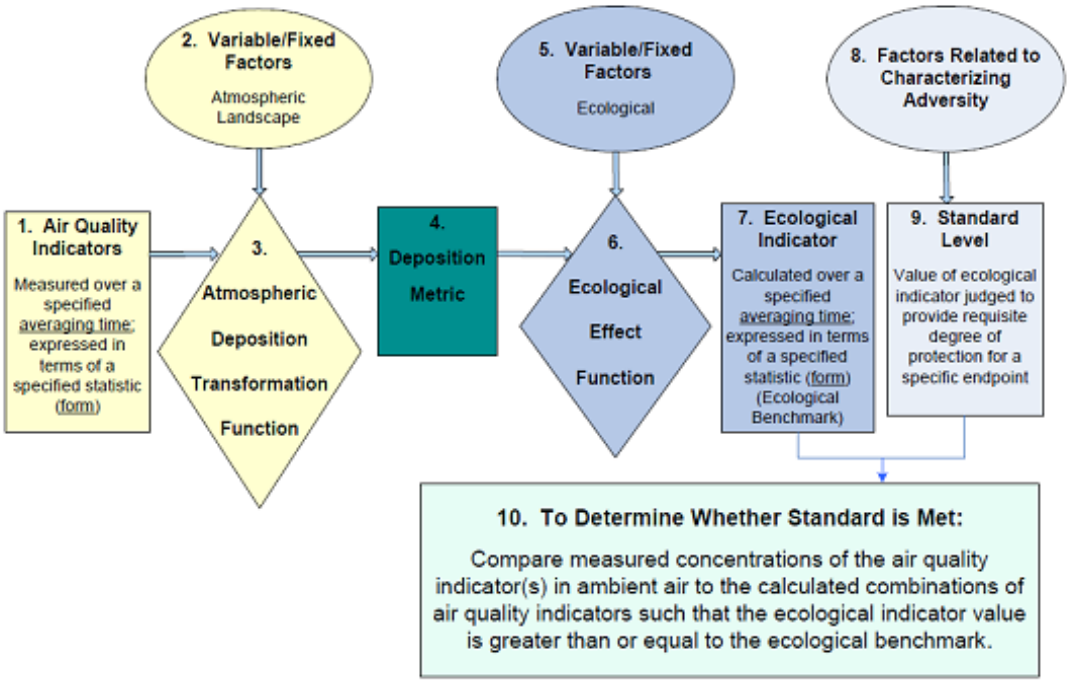

Figure 1.4-1. Possible structure of a secondary NAAQS for NO_x and SO_x based on an ecological indicator.

It appears that the CMAQ model used in the analysis is the "Atmospheric Transformation Deposition Function." However, the CMAQ model transforms emissions, not concentrations. Moreover, it predicts both concentrations and deposition fluxes. As a result, there must be some other form for the atmospheric function depicted in Box 3 that EPA has not shown in the document. The relationship between concentrations and deposition fluxes of NOx and SOx is expected to be proportional, i.e. changes in concentrations would be reflected as proportional changes in deposition fluxes. However, it is likely that the relationship will vary seasonally and spatially depending on regional chemical regime, precipitation and other meteorological variables, as well as land use and terrain features. Because of limited measurements of NOx, it would be difficult to show the relationships between observed concentrations and deposition, but (at a minimum) the CMAQ model results could have been used instead (as they are being used for dry deposition).

It is essential that EPA should show the relationships between concentrations of SO_2 and NOx and the corresponding deposition measures for different regions of the country and for different seasons. These should include:

a. The relationship between measured SO_2 and measured wet sulfur deposition
b. The relationship between modeled SO_2 and total sulfur deposition (measured wet deposition and modeled dry deposition)
c. The relationship between measured NO_2 and measured wet nitrate deposition
d. The relationship between modeled NOx and total nitrate deposition (measured wet deposition and modeled dry deposition).

The EPA should also explore the impacts of combined reductions in SOx and NOx on total N deposition fluxes and their spatial distribution.

2. Since the approach used by EPA relies on model predicted data due to lack of measurements of dry deposition, it is essential to show that the CMAQ model used in the analysis perform adequately when compared against measurement data. The model evaluation presented in Appendix 1 is inadequate for a variety of reasons:

   a. Evaluation is shown for annual averaged quantities that can mask model performance issues, as the compensating effects can cancel out biases and errors in the model. It is a general practice in regulatory applications to choose averaging period commensurate with the measured quantities (e.g., see Morris et al., 2006; Tesche et al., 2006) when evaluating model performance. Therefore, EPA should base model performance statistics using daily or weekly averaged quantities, as appropriate, instead of the annual average quantities. In addition, the statistics should be aggregated for each season, as appropriate.

   b. There is no reason to use normalized mean bias statistics when using annual average quantities. It is recommended to use mean normalized bias for longer averaging periods and fractional normalized bias when using averaging periods of one week or less.

   c. The report does not include a model evaluation performed for NOx purportedly due to lack of available data. However, NO_2 data are available for a model performance evaluation of NO_2. Notwithstanding the issues with the current FRM method for NO_2, it is appropriate to show these model performance results.

      In addition, there are *continuous, highly sensitive and highly precise* measurements of nitric oxide, nitrogen dioxide, total oxidized nitrogen (NOy), and nitric acid available for the SEARCH network in the southeastern U.S. (Hansen et al, 2003; Edgerton et al. 2006) that could have been relied on to conduct a regional evaluation. Although limited in scope to the Southeast, conducting an evaluation using SEARCH data is better than not conducting any evaluation for NOx at all.

Because of the limited model evaluation shown in the document, the statement on Page 1-5 (Lines 2 to 4) of Appendix 1 that "the model performance results give us confidence that our applications provide a scientifically credible approach for the purposes of this

assessment" cannot be supported. Since there is no way to measure the model performance for dry deposition, the estimates of total deposition that rely on predicted dry deposition are highly uncertain.

3. Figure 3.2-1 does not mention or include lightning NOx (LNOx) and soil NOx emissions, so it can be assumed that either those emissions were ignored or were assumed to be negligible (<1% of total). Hudman et al. (2007) show that these emissions can be significant (9% and 27% of total U.S. NOx emissions from soil and lightning, respectively for July 1 to August 2004 period). The magnitude of the lightning source relative to the anthropogenic source in summer 2004 was constrained by the extensive aircraft observations of NOx as part of the ICARTT campaign, so there is high degree of confidence in those estimates. Although these emissions estimates may still have uncertainty associated with them, one cannot ignore a combined source that could be as much as 1/3 of the total NOx emissions over a significant period, especially when estimating regional deposition loads. Additional analysis (Lee Murray and Daniel Jacob, Personal Communications) shows than on an annual basis, lightning and soil NOx may contribute up to 20% of total NOx emissions in the U.S. The exclusion of these emissions used in the CMAQ model simulation further reduces confidence in the modeling results shown in the REA.

   Harvard has conducted preliminary work (Lee Murray and Daniel Jacob, Personal Communications) on estimating contribution of LNOx to nitrogen deposition over several years using the GEOS-Chem model. The results show that LNOx alone (not including the contribution from soil NOx) can contribute ~15% of total annual oxidized nitrogen deposition over the U.S. Another interesting result of their work is that dry deposition accounts for 2/3 of total nitrogen deposition, a result that is different from the results shown in the REA. This indicates that using a single model for a single year to estimate dry deposition could give unreliable estimates. A better approach would be to use multiple models for multiple years.

4. Chapter 3 discusses various uncertainties associated with the analysis, but there is no attempt made to quantify any of those uncertainties. It may not be possible to quantify every source of uncertainty, but there are cases for which this is possible. For example, it is known that ammonia emissions are highly uncertain and studies have been done by EPA to "correct" biases in ammonia emissions using inverse modeling (Gilliland et al., 2003). It is recommended that EPA rerun the CMAQ model with the revised ammonia emissions to quantify the effect of that uncertainty.

5. The REA is not consistent in its use of definitions for NOx and NOy. The first paragraph of Page 1-10 states that "oxides of nitrogen" and "nitrogen oxides" used in this document refers to all forms of oxidized nitrogen compounds, similar to the term NOy used in the scientific community. It is not clear from the document whether the term NOx used in the document refers to ($NO + NO_2$) or to NOy and it seems to be used interchangeable throughout the document (e.g., in Line 1 of Page 1-14, "NOx" seems to be referring to NOy, but on page 3-3 when discussing emissions, "NOx" seems to referring to sum of NO and NO_2). A better clarification needs to be made in Chapter 1 and followed throughout the document. The following instances of inconsistencies are noted as examples:

a. Page ES-11: Line 1 refers to annual average NOx concentrations shown in Figure ES-5, but the caption for Figure ES-5 refers to annual average NOy.
b. Similarly, Page 3-11, Line 27 refers to annual average NOx concentrations, but the caption for Figure 3.2-6 refers to annual average NOy. It is not clear what quantity is shown in the figure.
c. Page 3-21: The policy-relevant background concentrations are shown for NO_2. If the indicator is NOx, shouldn't those concentrations be shown for NOx?

Minor Comments:

1. Page 3-29, Line 19: "These findings show that NOx emissions are much higher than NH_3 emissions in most areas of the country". There is a logical flaw here. The finding that oxidized nitrogen deposition is higher than the reduced nitrogen deposition is a result of emissions that are input into the model, not the other way around.

References:

- Edgerton, E.S., Hartsell, B.E., Saylor, R.D., Jansen, J.J., Hansen, D.A., and Hidy, G.M. (2006). The Southeastern Aerosol Research and Characterization Study: Part III. Continuous Measurements of $PM_{2.5}$ Mass and Composition. *J. Air Waste Mange. Assoc.* **56**, 1325-1341.
- Gilliland, A. B., Dennis, R. L., Roselle, S. J., and Pierce, T.E. (2003). Seasonal NH3 emission estimates for the eastern United States based on ammonium wet concentrations and an inverse modeling method. *Journal Of Geophysical Research*, **108**, D15, 4477, doi:10.1029/2002JD003063
- Hudman …. Hudman, R. C., Jacob, D. J., Turquety, S., Leibensperger, E. M., Murray, L. T., Wu, S., Gilliland, A. B., Avery, M., Bertram, T. H., Brune, W., Cohen, R. C., Dibb, J. E., Flocke, F. M., Fried, A., Holloway, J., Neuman,9, J. A., Orville, R., Perring, A., Ren, X., Sachse, G. W., Singh, H. B., Swanson, A. and Wooldridge, P. J., 2007. Surface and lightning sources of nitrogen oxides over the United States: Magnitudes, chemical evolution, and outflow. *Journal Of Geophysical Research*, **112**, D12S05, doi:10.1029/2006JD007912
- Hansen, D.A., Edgerton, E.S., Hartsell, B.E., Jansen, J.J., Kandasamy, N., Hidy, G.M., and Blanchard, C.L. (2003). The Southeastern Aerosol Research and Characterization Study: Part 1 – Overview. *J. Air Waste Mange. Assoc.* **53** 1460-1471.
- Morris, R. E. , Koo, B., Guenther, A., Yarwood, G., McNally, D., Tesche, T. W., Tonnesen, G., Boylan, J., and Brewer, P. (2006). Model sensitivity evaluation for organic carbon using two multi-pollutant air quality models that simulate regional haze in the southeastern United States. *Atmos. Environ.* **40** (26) 4960-4972
- Tesche, T. W., Morris, R. E., Tonnesen, G., McNally, D., Boylan, J., and Brewer, P. (2006). CMAQ/CAMx annual 2002 performance evaluation over the eastern US. *Atmos. Environ.* **40** (26) 4906-4919.

Mr. Rich Poirot

General Comments on Chapter 3
This chapter summarizes the technical approach for characterizing spatial and temporal patterns in the atmospheric deposition of S and N compounds – originating from anthropogenic SO_x, NO_x and NH_x emissions - in the various case study areas. As a practical matter, these estimates are made with relatively high spatial and temporal resolution covering the entire US, and with flexible modeling methods that will allow scaling of estimated deposition patterns and effects to other sensitive areas, and would also support evaluating potential responses to changes in deposition, air quality and/or precursor emissions.

Generally, I think the chapter is clearly written, conveys useful information, and presents convincing support for the approaches taken to provide the best possible estimates of air quality and deposition in the case study areas and elsewhere within the US measurement and modeling domains. One general criticism is that on several occasions the discussion seems unnecessarily qualitative in places where it would take no more space and provide more useful information if quantitative information was provided. Rather than saying "much greater" or a "vast majority" you could say "5 times greater" or "90%", etc.

The various maps showing emissions concentrations, depositions, etc. are very helpful. Even though effects of aerosol-phase S and N compounds are intentionally (and unfortunately in my opinion) excluded from these secondary NAAQS discussions, I think it would be useful to include maps of sulfate, nitrate and ammonium aerosol concentrations. These should be readily available as CMAQ model output, as well as from the relatively dense IMPROVE + CSN aerosol speciation networks. It would also be useful to show some "ratio" or "difference" maps – for example the ratio of S (or N) deposition to SO_2 (or NO_2) concentration and S (or N) deposition to S (or N) emissions, and the ratio of total reduced nitrogen deposition to total nitrogen deposition. Ratio or difference maps could also be a useful way of communicating differences between measured and modeled gaseous or aerosol concentrations or deposition. In making these maps, it may be necessary to employ some spatial aggregation or smoothing to "show" the information clearly.

1. The first charge question asks if the chapter appropriately identifies and describes uncertainties associated with these air quality-related analyses. The Section 3.5 "discussion of uncertainties" includes a fairly complete listing of the various types of uncertainties associated with the characterization of air quality and deposition in the case study areas. However, the descriptions and discussions of these various causes of uncertainty are so minimally detailed and (intentionally) non-quantitative that I'm not sure much useful information is conveyed.

One exception is a detailed (but graphical only) comparison of the wet N and S deposition estimates derived from simple interpolation of NADP data (the method used in this assessment) compared to similar estimates based on a higher-resolution enhanced deposition model (Grimm and Lynch, 2004), as well as to CMAQ wet deposition estimates for the Adirondack case study area and surrounding region. This is an informative comparison (and responsive to previous CASAC suggestions), but the results could be more useful if somewhat more quantitative comparisons could be included. For example, the maps showing the "similar" spatial patterns might be accompanied by scatter plots providing a more quantitative comparison of the gridded data, perhaps with different colored symbols to indicate grid points (a) within the Adirondack

case study area and (b) within or including the watersheds of the case study lakes. A minor point here is that I don't think the Grimm/Lynch model really accounts for cloud water deposition, which can make substantial additional contributions above elevations of about 600m.

Section 3.3.2 indicates that the discussion of uncertainties in section 3.5 will include a comparison of (2002) results from CMAQv4.6 and v4.7, but no such comparison is actually included there. A brief mention of these comparative results is given in Appendix 1, but this is quite minimal (TNO_3 over-prediction in CMAQv4.7 is about twice that of CMAQv4.6), includes no discussion of possible causes of the differences, and is not especially useful. I recommend adding some quantitative summary (perhaps a table) of the model/measure and model/model comparisons presented in Appendix 1 to Section 3.5. This could be accompanied by a caveat that such quantitative comparisons are not intended to be comprehensive…

To the extent possible, it would be useful if some indications of relative uncertainties could be provided. For example, I would guess that uncertainties in estimated dry deposition are greater than for wet deposition, and that uncertainties in the total deposition of deposition of reduced N are greater than for oxidized N, which are greater than for sulfur deposition. In addition, I would imagine that uncertainties in the characterizations of emissions, air quality and deposition are relatively small compared to uncertainties in the resultant chemical and biological responses in the affected ecosystems.

2. The second charge question asks whether replacing the previously presented RSM analysis with a new set of CMAQ simulations provides an ability to adequately examine the contributions of NO_x emissions to total (reduced and oxidized) N deposition. I'm not sure that the previous RSM approach was inadequate; rather I think the RSM and its uncertainties were not very well explained and documented in the last draft REA. However, I do think the current approach including additional CMAQ runs, which separately explore effects of 50% reductions in NO_x, NH_x and SO_2 emissions and also explore inter-annual variability through use of different meteorological years, provides a clearer and more transparent view of the relative contributions from oxidized and reduced N emissions. This is further supported by the model/measurement and model/model comparisons summarized in Appendix 1.

3. The 3rd charge question asks if the evaluation of CMAQ model performance presented in Appendix 1 is sufficient to support the use of the model in this review. In my opinion, Appendix 1 is presented very clearly and concisely, and does provide sufficient support for the use of the model in this review. One suggestion that might help strengthen this support would be to include some additional references documenting the fairly extensive past performance evaluations to which the model has been subject (i.e. by Appel, Napelenok, Pinder, Gilliand, Dennis, etc.). A second suggestion is to add – either in the appendix or in the Chapter 3 section on uncertainties, a brief summary of the possible implications of the model performance evaluation to the risk assessments being conducted in the selected case study areas.

Specific comments on Chapter 3

p. 3-2, line 18: "$_x$" should be "NH_x". Also you could add ", and their atmospheric transformation products" after "SO_x" and add "the earth's" before "surface properties".

p. 3-2, line 19: You could change "parameters" to "variables".

p. 3-2, line 22: absence of routine occult deposition measurements is not much of an excuse for ignoring what we know very well is an important input at higher elevations and in coastal areas. You could have at least modeled it, as you have done for dry deposition.

p. 3-3, lines 3-4: This is only a half truth. NO and NO_2 are also relatively reactive and may be removed by chemical transformation (to soluble species and/or to compounds like HNO_3 with high deposition velocities) and more rapidly than less reactive more soluble gases like SO_2 under a range of conditions. Also, a high proportion of NO_x emissions are from automotive sources at ground level, and so the potential for long range transport is less than for elevated stack releases.

p. 3-4, line 11: Could you add a parenthetical (XX%) to indicate more clearly what you mean by "the vast majority".

p. 3-5, line 4: Same comment as previous one. It would cost you no space to indicate quantitatively by how much are the NO_x emissions "far greater" in the East than in the West. One easy way to do this would be to add "(total 2002 emissions = XXXX tons/yr)" to the labels of the Eastern , Western and National US NOx emission pie charts in Figure 3.2-1. Also in line 5, instead of telling us that the most of the Western NO_x emissions are from California but are "not shown", why not just add a (XX%) and show us what you mean.

p. 3-5, lines 7-8: So what's the implication (if any) for deposition-related effects of highly episodic NO_x emissions from forest fires? Do we expect episodic acidification or eutrophication? I doubt it.

p. 3-6: I don't understand how confined animal feeding operations greatly increase the <u>volumes</u> of animal wastes. I can see confined feeding increasing emissions densities and the emissions of volatile components of these wastes to the atmosphere (or in runoff to surface waters), but would think the volume of waste produced by 100 cows is about the same weather they are confined or disbursed.

p. 3-8, line 5: You could add "stationary combustion" between "non-EGU" and "sources", as everything is either an EGU or a non-EGU.

p. 3-8, line 10: How much is "much greater in the East"? Again, why not just add the East, West & National totals to the pie chart labels in Figure 3.2-4.

p. 3-10, line 13: add "." after "area".

p. 3-11, lines 27, 28, 32 and p-3-12, lines 1, 4, 7, 10: Text describing figure refers to "NO_x" but Figure 3.2-6 refers to "NO_y".

p. 3-15, lines 21-22: I assume you mean "reduced nitrogen wet deposition was calculated from measurements of wet ammonium (NH_4^+) deposition".

p. 3-16, line 13: You could add "'N" after "measured".

p. 3-24, lines 9-10: Refers to a comparison of CMAQ v4.6 and v4.7 results included in Section 3.5 discussion of uncertainties, but no such comparative results are presented there, and these are only minimally described in Appendix 1 (which includes a footnote on p 1-1 noting that the differences in v4.6 and v.4.7 are small, "as described in Chapter 3". It would be useful to

actually see this description in chapter 3, along with a more detailed comparison in the Appendix.

p. 3-27 and elsewhere "Whiteface" is one word.

p. 3-27, lines 17-18: The recently available 2008 data show a decrease, and you could add a comment to this effect.

p. 3-31, Figure 3.3-3a: I think it would be clearer and shorter to present this data and that from the following 8 pie charts in tabular form and to show both the amounts in kg/ha/yr as well as in %, or you could at least add the totals (X kg/ha/yr) in the figure titles.

pp. 3-49 through 3-57: I would prefer that figures 3.3-6a through 3.3-6i were stacked bar charts showing the absolute (rather than %) amounts of the various components (Wet Re N, dry Ox N, etc.) in each season. We don't need the units to be "% of annual total by season" to see which seasons are most and least important and by how much. Similarly, the charts in figures 3.3-7a through 3.3-7i don't seem to convey very useful information. I think deposition of some species may have different environmental consequences in different seasons, and so it would be more important to no how much of spring N deposition comes from oxidized N than it is to know what fraction of total annual oxidized N is deposited in spring. Also, if absolute amounts were shown rather than percentages of total, it would give a good sense of differences in the various kinds of deposition by site and by season.

p. 3-59, line 10: Whiteface is one word.

p. 3-62: Use consistent terms for the case study areas in the 2 figures (Transverse Range vs. Los Angeles Range).

pp. 3-70 through 3-78: As for the seasonal N deposition charts, I think stacked bar charts showing absolute amounts from wet & dry S dep. by season would convey more useful information, and would also convey the differences in total & seasonal dep. rates in the different study areas. Also, what's the purpose of the large gap between dry & wet dep. bars in figures 3.3-15a through i?

p. 3-81, line 15: It isn't clear what "This" refers to.

p. 3-82, line 5: "$_4NO_3$" should be "NH_4NO_3".

p. 3-82, line 8: "$_x$" should be "NH_x".

p. 3-82, lines 14 & 15: Figure 3.4-4 doesn't really show "contributions of total reactive nitrogen approaching 50%". It shows a nearly 50% reduction in total nitrogen deposition if NH_3 emissions were reduced by 50%. If contributions of 50% were reduced by 50%, total N dep would have been reduced by 25%.

p. 3-82, line 29: "$_4NO_3$" should be "NH_4NO_3".

p. 3-93, line 5: I think "similarities in differences" should be "similarities and differences".

pp. 3-94 – 3-96: The figure captions refer to "total" oxidized N, reduced N or sulfur deposition, but I think they are (and text indicates they are) for "wet" deposition only.

Specific comments on Chapter 4

p. 4-5, line 17: I think you mean "…models do a better job projecting ANC than they do for projecting pH and inorganic Al …"

p. 4-1, Figure 4.2-4: The font labeling the figure axes and legend is too small to read.

p. 4-14, Table 4.2-2: Give units in table (μeq/l, I presume).

p. 4-18, line 7: Table 4.2-2 does seem to indicate that current SO_4^{2-} is "19 fold" greater than NO_3^- but this seems inconsistent with Figure 4.2-6 – which shows current (2006) SO_4^{2-} is about 4 times greater than NO_3^-. Why the difference?

p. 4-24, line 1: Should be "reductions … are" or "reduction …is".

p. 4-25, lines 24 & 25: Table 4.2-4 needs units, and again the observation that SO_4^{2-} concentration is 11 fold greater than NO_3^- (from table) is inconsistent with figure 4.2-14 which shows current (2005) sulfate being about 6x nitrate.

p. 4-40: It would be more effective to show some quantitative results from the sensitivity analysis in tabular or graphical form – possibly in an appendix – rather than just claiming that you did it and that it looked good to you. Without better context and detail, its impossible to know the implications of some of your summary statements – for example what does it mean that "similar results were given for the number of lakes with all realizations above the critical load" or that "changes in critical load values could range from 3 to 24 meq/m2/yr…"

p. 4-41, lines 13-18: Its not clear what's being described "which is on average a 15 μeq/l difference in ANC concentrations or 10%" or what's "on average [an] 8 μeq/l difference in ANC concentrations or 5%".

General comments on Chapter 7

Synthesis and integration of these disparate case study results is an inherently challenging undertaking, and given the difficult task, I think chapter 7 is reasonably well done. I especially like the discussion of uncertainties in Section 7.2 which is organized along the lines of a conceptual model which ultimately relates ecological benefits or welfare effects back to ambient air quality indicators. This conveys the logical progression from air quality to environmental effects and also illuminates the weakest links in this chain. Since the case studies are both diverse and also represent rather extreme conditions, its difficult to envision how the case study results might be scaled to cover a range of environmental effects extending over broader spatial areas. For aquatic acidification, examples are provided for how the case study results might be extended to larger populations of lakes and streams, but its much less clear how this could be done – in ways that would either support selection of specific air quality indicators, or support implementation of secondary NAAQS based on those indicators – for terrestrial acidification or aquatic or terrestrial nutrient enrichment. Some added discussion of the spatial extent of the kinds of environmental effects considered in the case studies would be useful in this chapter.

Another possible missing piece in this synthesis and integration is a discussion of how the different environmental indicators might relate to each other. Using the example conceptual model diagram in Figure 7-2, it seems like there might be (at least) four different diagrams and secondary standards - one each for aquatic and terrestrial acidification and aquatic and terrestrial nutrient enrichment. And each of these would be further modulated by various mixes of S and N deposition and the inherent sensitivities of the effected ecosystems. However, it also seems likely that there would be substantial overlap in areas with surface waters exceeding an ANC threshold and areas where forest soils have Bc/Al ratios below a specific threshold. Possibly the two indicators are sufficiently closely related that a single indicator could be selected that would be "controlling". I wonder if it would be possible to identify a Bc/Al ratio that's the approximate "equivalent" (i.e. would cover about the same spatial area) of an ANC of 50? Or in terms of the severity of environmental effects or the sustainability of some current or improved level of environmental quality, what level of ANC is the approximate equivalent of a Bc/Al ratio of 10, and what kinds of ecological effects from aquatic or terrestrial N enrichment would be considered to be of similar severity? For a given "acid sensitive" area with poor buffering capacity, it would be useful to know whether the aquatic acidification threshold or the terrestrial threshold is more limiting (and whether the answer to this question is based on the inherent environmental sensitivities or on the relative severity of the selected thresholds of adversity).

1. The first charge question asks about the extent to which the description of ecosystem services provides a useful framework in the case study analysis for informing standard setting. While I think the quantitative estimates of (selected) ecosystem services can be one useful consideration to inform standard setting, I think it should be clearly recognized that each presented example of ecosystem services is itself a limited case study. I think the example of considering (only) sport fishing (dis-) benefits from aquatic acidification helps illustrate this point – as it seems only a few degrees removed from David Stockman's infamous 1983 calculation that acid rain controls would cost $6,000 per pound of Adirondack brook trout. I think the presentation of example ecosystem services is useful in chapter 7, but it should be emphasized that only a few examples – with relatively easily monetized benefits - are provided here. An alternative approach to considering "adversity" of effects might be to consider the concept of "sustainability". Given geo-specific rates of soil & bedrock weathering for example, what rates of acidic deposition can be sustained indefinitely without eventually leading to degradation of environmental resources. I think this concept of sustainability is especially important in considering effects which result from long-term, cumulative deposition processes.

Another relevant concept which is not really addressed in this chapter or in previous sections relates to the nature and/or rates of "recovery" of adversely affected ecosystems. A current level of damage that has resulted from past and continuing deposition of acidifying or nutrient enriching S and N compounds may not be fully reversible to pre-deposition conditions and/or may require a substantial lag period between reduction in emissions (and concentrations and deposition) and improvements in environmental conditions. Possibly the "time to" and "degree of" recovery might be variables considered in judging the relative adversity of effects. Since secondary standards have no fixed time requirement for attainment, another possible approach here might be to consider standards requiring a certain rate of progress towards long-term goals which may not be feasible to attain quickly in all areas.

2. The second charge question asks if sufficient information is provided to inform standard setting based on effects other than aquatic acidification (terrestrial acidification, terrestrial nutrient enrichment and/or aquatic nutrient enrichment). As indicated above, I think its possible to identify some deposition thresholds estimates of terrestrial acidification effects, using

something like the Bc/Al ratio, that might be extended over relatively large spatial areas (as has been recently done for New England and Eastern Canada and as is currently being extended to NY). I also think its likely that there will be considerable overlap in regions of, and extent of severity of effects on, aquatic and terrestrial acidification, and/or that there may be a common level of S+N deposition that provides an approximately equivalent level of protection against both kinds of effects. If a specific ANC threshold is considered for aquatic effects, it should also be relatively easy to calculate necessary S+N deposition reductions to attain such thresholds and then to calculate improvements in Bc/Al ratios and terrestrial effects that would result.

Another possibility that should be considered is that if the ambient air quality indicators are required to be in units of the nominal gaseous criteria pollutants – SO_2 and NO_2 – such measures are more related (spatially and temporally) to the emissions that need to be controlled, while the effects of those emissions are more related to the long-term deposition of the chemical transformation products – $SO_4^{=}$ and NO_3^{-} – in combination with inherently sensitive bedrock and soils. These areas of greatest effects are therefore inevitably displaced, sometimes at considerable distance, downwind of the areas of highest emissions and gaseous precursor concentrations. An exceedance of an environmental indicator should therefore logically trigger exceedances of the air quality indicator in relatively large upwind regions. If for example an ANC or Bc/Al threshold is exceeded in the southern Green Mountains of VT – as can be expected given recent TMDL calculations for acid sensitive lakes there, and recent Bc/Al calculations by the NEGECP for that area – a zero-out of all SO_x and NO_x emissions in VT would have little or no effect on reducing the S + N related exceedances of the ANC or Bc/AL thresholds in that region (which is much more dependent on emissions from NY PA and OH). The secondary SO_2 and NO_2 standards should be considered to be exceeded until they are reduced to such levels throughout the eastern US that the deposition-related indicators are not exceeded in (VT and other) acid sensitive areas within that region.

Regarding possible NAAQS based on nutrient enrichment indicators, the various terrestrial effects at different and widely varying N deposition rates as summarized in Figure 7.1-3 provides some basis for considering secondary NAAQS based on several different (low, medium and high) degrees of protection based on total N deposition rates, and calculations could be made of the necessary reductions in NO_2 concentrations and NO_x emissions over broad spatial scales needed to attain those levels of protection. As with acidification-based standards, I don't think its necessary – and may be counter-productive – to try to link environmental indicators and air quality indicators on a point-specific basis, but rather we might consider what levels of total N deposition and effects protection would be achieved in areas with the most sensitive ecosystems if ambient NO_2 concentrations (and NO_x emissions) were reduced by various amounts or percentages throughout the surrounding region.

Considering a secondary NO_x standard based on aquatic nutrient enrichment is further complicated by the large and often dominant contributions from non-air sources. However, calculations could still be made of improvements in aquatic effects that would be expected if oxidized N deposition (and/or NO_2 concentrations) were reduced to certain absolute levels or by certain percentages, with contributions from NH_x and non-air sources taken as a given. This process along with similar assessments for terrestrial nutrient effects and acidifying effects of N deposition might lead to considering a range of secondary standards that would yield a range of beneficial environmental effects.

I think the discussion of uncertainties in section 7.2, organized as a conceptual model linking air quality indicators to ecological effects, can also be used as a guide to areas that would benefit

most from future research and/or environmental assessment for future NAAQS revisions. I also think continuation of the work conducted for this assessment has good potential applications in areas outside the specific SO_x and NO_x NAAQS revision process. Historically, major reductions in SO_x and NO_x revisions have resulted largely through national and regional programs justified by benefits in a wide range of health and environmental effects. This will likely hold true in the future as future revisions to national automotive emissions standards, (whatever replaces) the Clean Air Interstate Rule, future checkpoints for the Regional Haze Rule, and likely lower NAAQS for PM and ozone will all result in, or be strengthened by advance assessments of, the environmental benefits of lower SO_x and NO_x emissions.

Specific comments on Chapter 7

p. 7-1, lines 21-22**:** I don't think you really mean that a welfare effect is what "society views as beneficial". Society views good visibility or a healthy environment as beneficial. A welfare effect occurs when these beneficial welfare or environmental goods are adversely affected.

p. 7-10, lines 1-7: Its not really clear if or how you intend a single ANC indicator threshold to be protective against seasonal or episodic variations. Nor does it seem likely that a given ANC limit, measured at summer low flow for example, would protect all lakes and streams equally against episodic acidification effects from spring snowmelts.

p. 7-11, lines 16-19: For the Bc/Al ratios of 0.6 and 1.2, quantitative estimates of reduced growth are provided (and seem rather "non-protective" if they would allow 75% or 50% of North American tree species to experience growth reductions). By contrast, not much justification is provided for the selection of the more protective Bc/Al ratio of 10 – other than the observation that it's the most conservative value used in some analyses. There isn't really any better justification for the 10 ratio provided in Chapter 4 or in Appendix 5, and I think there needs to be a clearer explanation for why a number of prominent studies have used this ratio. This seems especially important given the large range between 1.2 and 10, and the large spatial areas that will presumably fall within those extremes.

p. 7-12, line 4 "res" should be "red".

p. 7-13, Figure 7.1-2: This figure is not described very well in the text. For example, what are the OEC and OHI scores, or how does the chart show that a greater than 100% reduction in atmospheric N deposition was needed to move from bad to poor.

p. 7-14, line 22: What's a "compared forest health decline"?

p. 7-14, line 26: Its not clear what "(generally at deposition rates of < 10 kg N/ha/yr)" means here. I assume this is a general description of the N additions applied in a particular set of experiments – that have shown transient growth increases followed by increased mortality at deposition rates below 10 kg N/ha/yr, but don't mean to imply that experimental treatments at 10 kg N/ha/yr or greater showed no transient growth increases or increased mortality.

p. 7-19, line 12: I assume that non-use (existence) services would also be affected by aquatic or terrestrial acidification or terrestrial nutrient enrichment.

Mr. Dave Shaw

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?***

I feel that the REA Executive Summary provides a clear summary of each of the chapters with conclusions allowing the reader to extract highlights of the report without reading the entire report.

I would like to see some clarification on critical loads and how it is used in this assessment. I appreciate that the areas of confidence are expressed in the conclusions.

General Comment

I am still committed to the idea that one of the key issues driving this review should be to also identify where data is missing with the intention of filling those gaps. I feel that this should be clearly stated in the Executive Summary, perhaps also listed in the Policy Relevant Questions on Page ES-3. While this may be somewhat addressed in the last two bullets, I feel that it deserves a little more emphasis.

Some other items that would help to emphasize this key issue:

- Improve the link/reduce uncertainties between ambient air quality indicator and deposition.
- Improve/reduce uncertainty in response models (e.g. MAGIC and SMB) or create new ones, to be better predictors (e.g. of pH, Al) and to be better at scaling up to the broader regions. Consider developing Base Cation Surplus as a substitute for ANC.
- The above bullets would require improved input data.

The natural course of action in addressing the above bulleted issues would be to ask EPA to make a separate exercise of identifying future data refining needs. Now that we have the ISA and REA documents to substantiate the needs. The goal should be to increase the confidence in ecological exposure, responses, effects and benefits *to all other areas beyond aquatic acidification*.

I propose that EPA sponsor or provide for a focus group on monitoring and research information that would reduce uncertainty (by improving the input data and improving the models themselves) from these case study areas for national assessments of CL. Uncertainty analysis of those results should be ongoing in the CL process.

Chapter 3: Sources, Ambient Concentrations, and Deposition

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 and sulfur. Are the uncertainties associated with these analyses appropriately identified and described?

The uncertainties associated with these analyses are appropriately identified and described, with a few possible exceptions. In terms of air concentrations, was there an attempt to compare modeled NOx and SOx where measurements are available? Even though there are few NO_2 monitors, and most are in urban areas, it would help to see that at least in urban locations, the model predictions of NO_2 were reasonable. There are still numerous SO_2 monitors at SLAMS/NAMS sites as well.

Section 3.5 does mention some of the issues related to combining measured wet deposition with modeled dry deposition, but leaves out at least one limitation. The spatial analysis using NADP wet deposition data does not capture urban influences well (if at all), whereas the modeled dry deposition will include urban sources and influences (that, of course, are not measured routinely). This methodology may overemphasize dry deposition in the vicinity of large point sources or urban/suburban areas, and we don't have adequate data to assess how important this is.

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 NO_x and NH_3 emissions to total, reduced and oxidized nitrogen deposition and the relative contribution of SO_2 emissions to sulfur deposition. Does this approach enable us to adequately examine the contribution of NO_x to total nitrogen deposition?***

Yes, the 50% NOx and 50% NH_3 reduction scenarios are generally adequate to quantify the contributions of NOx to total N deposition across the modeling domain. However, Section 3.4 has a disconnect with the rest of this chapter of the draft REA. In this section the wet deposition is taken directly from the model, whereas in previous sections the wet deposition is derived from the NADP data. This is necessary in this section since you are trying to estimate changes in modeled wet and dry deposition resulting from changes in modeled emissions.

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?***

The analysis presented in Appendix 1 is generally sufficient to support the use of CMAQ in the draft REA, especially with regard to wet deposition. The seasonal, year-to-year, and broad geographic variations are clearly displayed. On the "dry" side, the model evaluation suggests that ambient concentrations are reasonably well characterized. However, dry deposition is a significant contributor to the total loading of S or N, and it is not clear that the CMAQ dry deposition estimates are valid or even reasonable. While the CASTNet program does not measure dry deposition directly, it still might be useful to compare model predictions of dry deposition at CASTNet sites.

Chapter 4: 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 Shenandoahs relative to three potential ANC cutoff levels (20, 50, and 100 µeq/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?

ANC is a good indicator and these are good cut off levels for selected lakes in the Adirondacks and selected streams in the Shenandoahs. However, these lake standards may not be protective of streams in the Adirondacks. For instance, we know from ongoing research that Adirondack streams do not respond the same as lakes and are not experiencing the recovery that has been detected in lake trends. We are also learning that Base Cation Surplus is a better indicator than ANC to characterize biological impacts in streams. This was mentioned in the ISA (Lawrence et al) and should be reported in the REA.

There are aspects of the Adirondack Case Study Area assessment that need clarification including:

1. Respresentativeness of the modeled waters
2. The treatment of natural acidity
3. Review of the paleolimnological data on preacidification conditions

Representativeness of the modeled waters

The modeling discussion is not clear about the various Adirondack data sets used. There is not enough explanation as to which lakes were used from what survey program and why. For instance (pg 4-14) MAGIC was run on 44 lakes in the Adirondack Case Study, said to be taken from the EPA TIME (n =43 lakes) and LTM (n= 52 lakes). It would be helpful to explain why some lakes were chosen and not others.

It appears that average yearly ANC values were calculated from single measurements, because Adirondack TIME waters are collected only once per year. This should be explained more clearly.

Page 4-16, shows 169 lakes were modeled in the Adirondack Case Study Area. Figure 4.2-9 (pg 4-20) shows ANC values from 94 TIME/LTM lakes, however there are only a maximum of 89 TIME and LTM lakes due to the 6 overlap waters between the two.

The treatment of natural sources of acidity in lakes

Natural acidity is a common phenomenon for the Adirondack Lakes region, this, combined with the observation that DOC is increasing in some areas, demonstrates that more consideration is required than what is provided here. I am unclear as to how a 'natural' ANC concentration of less than 50 determined (pg 4-34 line 26), perhaps more explanation is in order.

Review of the paleolimnological data on preacidification conditions

Were any of the selected MAGIC lakes, part of PIRLA (Paleolimnological Investigation of Recent Lake Acidification) lakes where diatom sediments provide for reconstructed histories of lake chemistry to prior to the onset of recent acidification?

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 NO_x and SO_x 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 (BC_0)). 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?***

The link between deposition and an ANC in surface water is crucial and difficult not only at each specific location but also in order to generalize findings on a broader scale. Determining catchment weathering rates is critical because 'the catchment supply of base cations from the weathering of bedrock and soils is the factor that has the most influence on the critical load calculation and has the largest uncertainty" (pg 4-40 lines 3-5) contributing almost half of the total variability in critical load estimates (pg 4-69 line 16).

As far as methods to examine this and the generalization from specific inputs/sites to apply to a broader scale, I note that current research is underway through the New York State Energy Research and Development Authority by Miller, Lawrence, Weathers and others to examine weathering rates and these regionalization questions. These results and other related work e.g. sponsored by the National Park Service should be incorporated as they are made available

Aquatic acidification is the best place to start because of the weight of the evidence across many sensitive areas. However, while ANC is currently the indicator of choice, other more biologically direct indicators (pH and Aluminum) should be considered along with the newly developed Base Cation Surplus. These will likely be more useful further down the road in the critical loads process. For example, inorganic monomeric aluminum at detectable levels (2 umol/L) is toxic to biota in any environment, aquatic or terrestrial. Its presence is directly associated with anthropogenic acidic deposition.

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 relative 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 Forst 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 ratios and tree health sufficiently described and are uncertainties adequately characterized?***

The approach is adequate to develop critical loads for these case study areas, but I feel that further discussion is needed to determine whether the assessment is adequate to inform about the terrestrial responses of all sensitive areas like the Adirondacks and other US regions (Fig 4.3-3) with potential red spruce and sugar maple sensitivities. It would be helpful to explain how the indicator soil solution ratios at the Kane Experimental Forest (PA) and/or the Hubbard Brook Experimental Forest (HBEF) in NH are going to be translated to protection of sensitive sugar

maple and red spruce ecosystems in other regions? Specifically, how will the uncertainties with misclassification of soil parent material (pg 4-69) be resolved?

While HBEF is a pre-eminent long term forest ecosystem study site, it has limitations representing high elevation northeastern forests. For example, the high elevation forests (greater than 900 m above mean sea level) of the Adirondacks cover over 100,000 acres and according to Miller et al 1993 have accumulated 80% more sulfur than the lower elevation forest at HBEF. A significant portion of the sulfur as measured at Whiteface Mountain in the Adirondacks comes from dry deposition and cloudwater contributions.

A question that may be in order for discussion is: Are the number, location and routine parameters collected by the USFA FIA permanent sampling plots adequate to support the analysis for acidity critical loads to high and medium elevation forests in the northeastern US?

Chapter 5: Nutrient Enrichment

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?***

I feel as if SPARROW is adequately described within the REA and appreciate the detail in Appendix 6. I especially appreciate the use of the New England waters paper by Moore et al.

I believe it would be helpful if in either the REA or Appendix 6 a table with the data type and sources were printed. For example, where was the loads data derived from, or the stream network? The use of the other tables in this section are especially helpful, so this additional table would contribute to the ease of understanding.

Case Study Analyses (Chapters 4 & 5)

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?***

I feel that aquatic acidification is the best place to start because of the weight of the evidence across many sensitive areas. However, while ANC is currently the indicator of choice, I believe that other more biologically direct indicators (pH and Aluminum) should be considered along with the newly developed Base Cation Surplus. These will likely be more useful further down the road in the critical loads process. For example, inorganic monomeric aluminum at detectable levels (2 umol/L) is toxic to biota in any environment, aquatic or terrestrial. Its presence is directly associated with anthropogenic acidic deposition.

Specific comments/questions

Chapter 3: There are several instances in which "NH" is left out – Page 3-2 lines 9 and 19, and Page 3-82 line 5.

page 4-10 Figure 4.2-2 New England is not an ecosystem

Pg 4-15 Figure (a) How were the critical load (green dot) modeling sites selected?

Pg 4-14 line 14. What is the present condition? Here it is defined as 'present (2002 and 2008)' whereas the discussion starting on pg 4-18, calls 2006 the current condition. Which are actual measured and which are modeled current condition values?

Pg 4-39 Uncertainty and variability. How well do the MAGIC model simulations for current ANC concentrations measure against measured median ANC?

Pg 7-1 lines 21 to 27. By transitioning to the "ecosystem services" concept, (we must also preserve) there is a danger of losing the broader definition of effects that are important to people or that society views as beneficial. Specifically it should be recognized that the majority of the sensitive areas identified in these documents are either national or state parks or scientific experimental forests that they have a collective intrinsic value that have been previously identified as exemplary. They deserve the bar of level of protection to be raised. (Protecting biota in a park or a scientific reserve is 'more important' than anywhere else because we have as a society identified these are part of the public good.)

Pg 7-10 lines 5 to 7. Crucial point but sentences are awkward. Not exposure to an annual mean, but on average with an annual mean of 50, the probability is low that the lake will go down to 0 during springmelt. The length of time sentence should be deleted as it does not summarize that section, but a point to be made as part of the background to this discussion.

Appendix 1, Footnote 8: Why is NOy listed as the sum of NO, NO_2, HNO_3, and PAN? It should include also HONO, N_2O_5, ANO_3, and NTR as is listed in the Table 1.1-2 for deposition.

Appendix 1, Section 2.1 and Figures 2.1-1 through 2.2-7: Don't these additional lines indicate a factor of 1.5 around the 1:1 line (not a factor of 2)?

Discuss further the consequences of MAGIC using a calculated ANC rather than measured Gran ANC. How well does MAGIC scale up beyond the watersheds of the modeled lakes themselves?