20 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. Praveen Amar **2**
Dr. Charles T. Driscoll **7**
Dr. H. Christopher Frey **18**

Dr. Praveen Amar

First of all, this second draft of theREA "Risk and Exposure Assessment for Review of the Secondary National Ambient Air Quality Standards for Oxides of Nitrogen and Oxides of Sulfur" is a substantial improvement over the first REA draft. It is very well-written, is more complete, emphasizes important issues of risk and exposure at the right level, and is organized and structured in a manner that makes it much more readable.

In addition to general comments on the Executive Summary, comments are provided on the section of REA dealing with air quality analysis (Chapter 3, entitled "Sources, Ambient Concentrations, and Deposition").

Comments on Executive Summary

The eight policy-relevant questions (page ES-3) do provide a focus on what needs to be done in the next phase of this CASAC review process. What, however, is not clear to me, is how these questions actually will be answered in the next "policy phase" of this process. Should EPA have made the first attempt to answer these questions here in this REA? What is missing in the Executive Summary and in the rest of the REA is a clear roadmap (or, a serious attempt) of how REA will get to the next "policy assessment" phase.

The articulation of the "right questions to ask" right at the beginning of the REA process is an excellent idea, even though it is difficult to formulate and write them out clearly. It is not clear to me why the List of policy-relevant questions in Chapter (Section 1.4) is expanded to fifteen questions from eight questions in the Executive Summary. Some of the questions in Section 1.4 are included in the Executive Summary, but new ones are also added. I recommend that both Lists include the *same* set of policy-relevant questions. If the two Lists which follow each other need to be different, please do provide a rationale. Otherwise, it is confusing to observe that the List of questions is almost doubled right in the beginning of REA.

Also, the sequence of policy-relevant questions should follow the layout of the REA itself. For example, the structure shown in Figure ES-2 (air quality indicators to deposition metric to ecological indicators to establishing standards to determination whether these standards are met) is a good way to re arrange the sequence of policy-relevant questions.

The fifth policy-relevant question (ES-4) should be modified to also include the corollary and related question "Does the available information provide support for considering *joint* air quality indicators for NOx and SOx?" The third policy question needs to address uncertainty in addition to variability "associated with those responses." Also, fifth bullet on Page 1-17 is awkward ("… trying to be protected against?").

A general comment about reduced nitrogen/NH_3/NH_4, etc.: This REA is a substantial improvement over the ISA and previous REA (first draft) in how it addresses the role of

ammonia and reduced forms of nitrogen in total nitrogen deposition (however, it still does not address organic forms of nitrogen). The EPA staff deserves to be commended for this major shift in their treatment of total reactive nitrogen. However, there are references in the Executive Summary and in the body of the document (Chapter 3 and other places, see below) that ammonia is a "local" pollutant or that the ammonia sources are located in "rural" areas or that at four million tons per year, U.S. emissions are "small," compared to emissions of NOx and SO_2. These statements seem to imply that ammonia is not as important as the other two pollutants. Recent work by EPA (Dr. Robin Dennis and others) indicates that even "local" CAFO (confined animal feeding operations) sources have a large regional footprint in addition to a large local impact. Since there is a large regional component associated with emissions, atmospheric chemistry, and transport of the NOx-SO_2-NH_3 system in addition to important "local" component, it is important that this REA, including the Executive Summary, be revised appropriately to better reflect the regional nature of ammonia emissions, and the regional role of reduced nitrogen in deposition and in ecological effects. The results presented in Chapter 3 clearly show, reduced nitrogen deposition is from 20 percent to as high as 50 percent (above 30 percent on average) of total nitrogen deposition at locations of various case studies. Therefore, ammonia/reduced nitrogen needs to be treated the same way in the REA as SOx and NOx.

The Atmospheric Deposition Transformation Function (ADTF) and Ecological Effect Function (EEF) as shown in Figure ES-2 and as described in the Text are good conceptual framework to address "source to welfare effect". What is missing here, however, is an acknowledgement that these functions are simply not some "magic translators" but are very difficult to "derive," once EPA takes into account geographical and seasonal variability of the relationship between concentrations and deposition, as well as uncertainty associated with measurements and model predictions. This needs to be clearly addressed in the context of Figure ES-2 and other places where this Figure appears as the organizing principal of this REA.

Page ES-6: The Executive Summary and the document use "nitrogen enrichment," (please see Page ES-6), "nutrient enrichment," (see Table on Page ES-7) and "nitrogen nutrient enrichment" to mean the same thing. Please choose one (I suggest "nitrogen nutrient enrichment") and use it throughout the document for clarity and for ease in reading.

Page ES-8 (line 6): "broad look" needs to be replaced with something more rigorous. For example, "broad evaluation/investigation" etc. The description on Page ES-9 (top) of monetized and non-monetized benefits is excellent.

Page ES-11: Figure ES-5, Title should say, NOx, and not, NOy.

Page ES-13: As an extension of my comment about reduced nitrogen/NH_3 above, it would be very helpful that the Executive Summary include two additional Figures in addition to the Figure ES-7, one on spatial distribution of oxidized nitrogen and the other of reduced nitrogen, with accompanying Text that highlights the important role that reduced nitrogen plays in conjunction with the role of oxidized nitrogen.

Page ES-15: It is important that this Executive Summary describe how the case studies will be "extrapolated" from "smaller scale to other sensitive areas in the country." This important

concept has been described in previous EPA efforts and needs to be addressed here before the four main ecosystem effects are presented

Page ES-19: Aquatic Nutrient Enrichment: The Text states "human activity has likely contributed to a six-fold increase in nitrogen flux …." Note that REA (Chapter 3) notes that this has happened over the last 100 years. I would speculate that this large increase has occurred over a much shorter time horizon, of say, last 30 to 40 years, over which emissions of NOx from some industrial sources, and especially, ammonia from animal operations have substantially increased at an accelerated pace. If this could be confirmed by EPA, it should be noted in the REA, because this finding may be policy-relevant if it implies a larger and more recent role of reduced nitrogen in total nitrogen deposition.

Page ES-20: The ASSETS index needs to be spelled out and clearly explained here as well under "Key Terms," especially under "Key Terms" in its proper context. I found it hard to understand. For example, it should be clear under "Key Terms" that it applies to nitrogen enrichment. Please also explain the use of the term "pressure" in its proper context. Same for "Determined Future Outlook" for the ASSETS index. The term, ASSETS itself, needs to be spelled out.

In "Key Terms" and in other sections including Executive Summary, the definition of "Uncertainty" needs to recognize that there is more to uncertainty than simply "parameter uncertainty." Please note that in addition to parameter uncertainty, there is equally important, if not more important, the concept of model uncertainty (that sometimes is addressed through expert solicitation or other methods). It would also be very helpful to explain here that uncertainty is different than other terms with which it gets confused (for example, variability, precision, sensitivity, and risk). It would be also useful to explain all these other concepts under "Key Terms."

Page ES-22; The various ecological thresholds for CSS community need to be summarized. This is too much detail for the Ex. Summary.

Page ES-24, Conclusions: This section needs to be redone and rewritten. It does not do justice to an otherwise well-written Executive Summary. The words "confidence," "most confidence," "fair amount of confidence" may be following the recent IPCC format, but they just do not fit here. One suggestion is to write this Conclusion section in the excellent format of "Key Findings," prepared by EPA staff during the earlier phase of the REA process.

Comments on Chapter 3

The f**irst** key charge question relating to air quality analyses deals with treatment of uncertainties associated with air quality analyses in Chapter 3 and whether uncertainties have been identified and described appropriately. Chapter 3 describes EPA's chosen approach that characterizes the spatial and temporal patterns of deposition at locations of chosen case studies. The deposition is

characterized for both wet and dry forms and for total reactive nitrogen (and its two components of oxidized and reduced forms) as well as for total sulfur.

The discussion of "uncertainties" in Section 3-5 is descriptive in nature. It is, however, not helpful in providing a quantitative and relative sense of uncertainties in various components (emissions, wet and dry deposition, sulfur versus nitrogen deposition). For example, it would be useful to state that NH_3 emissions are much more uncertain than NOx emissions that in turn are less certain than SO_2 emissions. It may also be useful to state that dry deposition is simply not measured but inferred from model calculations and has therefore uncertainties of unknown magnitude. For such cases, sensitivity studies could be useful to put bounds on the results. "Uncertainty" in "Key Terms" also needs to recognize that there is more to uncertainty than simply "parameter uncertainty" (see above).

The s**econd** charge question on air quality analyses is related to the replacement of Response Surface Modeling (RSM) in the first draft of REA with a new series of full-blown CMAQ simulations. The REA now contains CMAQ simulations to estimate the relative contributions of emissions of NOx and ammonia to nitrogen deposition (total N, and its oxidized and reduced forms) and a similar analysis for contribution of SO_2 emissions to total sulfate deposition. The second charge question asks, "Does this approach enable us to adequately examine the contribution of NOx to total nitrogen deposition?"

The replacement of RSM model with CMAQ and associated sensitivity runs (impact of 50% reduction in NOx, NH_3, and SO_2 emissions on deposition of nitrogen and sulfur) is step in the right direction. Evaluating the inter-annual variability in emissions and meteorology and their impact on deposition fluxes is an important and useful addition to this REA. The results in Chapter 3 and Appendix 1 are described in great detail in various figures, tables, and charts. My major comment, however, is that a much better and more realistic approach to investigate the impact of emission reductions on deposition fluxes is through the application of " Direct decoupled Method"(DDM) than the "brute force" approach of reducing emissions of one of three pollutants by a set percentage (in this case, 50%). Realistically, emission strategies are not based on "one pollutant at a time" reduction, but are a combination of different reductions in the emissions of SO_2, NOx, and NH_3. The relative sensitivity of various components of deposition fluxes (wet, dry, total, oxidized and reduced nitrogen, sulfur, etc.) should be investigated more meaningfully through the application of DDM or similar approach. It is not clear if EPA would be able to undertake such an effort in the short time available to develop secondary standards for SOx and NOx.

My second major comment (also noted above in the Executive Summary and repeated here) is that this Chapter needs to more clearly address ammonia and its emissions and impacts on deposition. Statements such as ammonia emissions are "local" or "rural" or are not as widespread as NOx and SO_2 miss the important point that ammonia is an extremely important chemical in the context of establishing secondary standards for SOx and NOx. Even though I do recognize that under the current regulatory constraints, REA can only develop a secondary NOx standard with ammonia "embedded" in it, the task is going to get only more difficult if we do not address the issue of ammonia and reduced nitrogen clearly.

The **third** charge question on air quality analyses relates to the CMAQ model performance evaluation undertaken by EPA after the first draft of REA in response to recommendations made by our Panel. This charge question asks, “Is this (new) analysis sufficient to support the use of the model in this review?”

Appendix 1 does provide useful and clearly presented information on the evaluation of CMAQ for annual average concentrations and annual wet deposition fluxes for components of nitrogen (nitrate, ammonium, sulfate, etc.) for the year 2002. It also provides data on model performance by comparing predictions of monthly concentrations and wet deposition fluxes with measurements for the four-year period of 2002-2005. Though model-measurement comparisons are useful for annual and monthly time scales, it may be useful to evaluate model at finer temporal scale (for example, hourly and weekly data where available) for a more “stressed” performance evaluation of CMAQ’s results.

Dr. Charles T. Driscoll

For each chapter general comments are given followed by specific comments.

Overall

The EPA is to be commended for this effort at compiling and completing the REA on ecosystem effects of sulfur and nitrogen oxides. This was a major undertaking and an important new initiative for future air quality management in the U.S. This report lays of a framework for a critical loads approach to guide mitigation of ecosystem effects from air pollution. The REA document is much improved over the previous draft. There are less typos and wording problems. Some of the redundancy has been eliminated. Most importantly the document is more focused than the previous draft

While I am enthusiastic about the initiative and report, there are a few considerations that should be addressed before the REA is finalized. First, there needs to be a more comprehensive and systematic discussion on the front end of the document on the concepts of maximum deposition load, critical load and target load. I have no idea what maximum deposition load is or how it is different from critical load. The term is introduced at the beginning of the report and then not really addressed again, while the term critical load is used throughout the report. Also the distinction needs to be made between critical loads and targets loads. This distinction is ignored in the report, but it has critically important policy considerations. Critical load is a steady-state concept. Unfortunately ecosystems are rarely, if ever at steady state. Steady-state models can be used to determine critical loads and/or empirical models. Target loads are a time –dependent phenomenon, and calculated with dynamic models. An important management consideration in this analysis is the time for ecosystems to reach critical chemical limits or achieve conditions of critical biological indicators following emission controls. Also the time to reach steady-state is important to understand for management considerations. The REA does not consider these time dependent process. The document really needs a clear treatment of these concepts and how they are used in the REA and for ecosystems management of air pollution effects.

Another important consideration is the spatial and temporal compatibility of the atmospheric transport models (i.e., CMAQ) and the watershed effects models. CMAQ is designed to address large spatial scales over short temporal scales. Many of the ecosystem effects and associated models are manifested over smaller spatial scales and over long time frames (e.g., decades to centuries). In the air quality chapter the authors provides some discussion of these considerations. If a critical load/target load is to be used to assess and guide ecosystem effects of air quality a rigorous analysis of the compatibility using transfers between atmospheric transport and watershed/ecosystem models this disconnect needs to be rigorously evaluated. While such an analysis is beyond the scope of the REA some text needs to be dedicated to this issue to set the stage for a future research initiative.

I have some pet peeves with the writing style that I have mentioned in previous reviews. Throughout the document the text is written as inanimate objects are doing something. For example, Figure xxx shows… This chapter discussed… An inanimate object cannot do anything.

A better approach would be: such and such is shown in Figure xx or In this chapter current emission sources are discussed…

I also urge that the word reduced only be used when referring to chemical reduction or a reduced chemical form such as ammonium. In the document the term reduced is used to refer to decreases as well as reduced chemical forms. This makes for some confusing text. Please make this change.

Executive Summary

General Comments

The quality of the Executive Summary is good. It provides a good overview of the report.

ES-2, line 10 - The text indicates that sulfur can limit productivity. I don't think this is a likely situation. The text should be clarified and probably corrected.

ES-5, line 18 – the REA uses a term maximum deposition load. How does this compare with the terms critical or target loads which are used later in the document (Chapter 4). Introducing a new term that is similar to other terms used in the report will confuse the reader. This term should be clarified relative to critical/target loads.

ES-16, text box – In this text box critical load is defined. I would like to see the actual definition of critical load, "the level of atmospheric deposition of a substance below which there is no harm to the ecosystem" or something like that. How does critical load compare with maximum deposition load? Also no effort is made to clarify the time-dependent nature of the problem. Critical load is a steady-state phenomenon; target load is a time dependent value. The text box should be expanded to address these issues.

Specific Comments

ES-2, line 7 – I do not like the term occult deposition. I do not believe it is a very accurate or descriptive term. Why not refer to it as cloud and fog deposition?

ES-2, line 13 – Change to … localized loss and extinction of fish.

ES-16, line 13- MAGIC should be defined or clarified.

ES-18, paragraph 2 – Some mention should be made of the value of sugar maple in fall foliage with respect to tourism.

Chapter 1.

General Comments

None

Specific Comments

1-13, line 4 – I do not like the term occult deposition. It is not a very accurate or descriptive term. Why not just refer to cloud and fog deposition?

1-13, line 6 - The text indicates that sulfur can sometimes limit plant growth and productivity. I don't think this is a likely situation. The text should be clarified and corrected.

1-15, line 21 – Methylation of mercury can occur in virtually all watersheds, not just the northeastern U.S and southeastern Canada. This sentence needs to be deleted or corrected.

Chapter 2.

General Comments

2-10, line 9 - As discussed above I do not like the term "maximum deposition load" it seems redundant with the concept of critical/target load. Why introduce a new term? This term should be clarified with respect to how it is different/ related to critical load. Also the second half of the phrase is not correct. It is not the amount that solves a mass balance equation for an ecological indicator. This sentence needs to be corrected.

Specific Comments

2-12. line 15 – The sentence "Valuation may be an important step…" is very similar to 2-11, line 18 "In addition valuation may be an important step…". You may want to delete one of these.

2-15, line 24- data are…

Chapter 3

General Comments

My hat is off to the authors of the chapter it is greatly improved over the last version. It is much more focused and easier to read. The authors did a great job re-orienting to focus on the ecosystem case studies. I also appreciate the effort made at the end of the chapter to discuss the sub-grid scale issues.

This section could be shortened further by eliminating the text on sulfur deposition for the nutrient case study areas; Potomac, Neuse, and western sites.

The authors should examine the text with respect the writing style. In some sentences the text is written as the present tense, in others it is written as the past tense. The text should be consistent, I would suggest in the past tense.

Specific Comments

3-1, line 21- Change to: were used as modeling…

3-2, line 8 – Change to: emissions, transformations and deposition…

3-2, line 9, 18- A typo? additional x? Should this be NHx?

3-2, line 28- Change to: The emissions to atmospheric concentrations-to-deposition...

3-3, line 17- Change to: are important components…

3-14, line 12- Change to: United States were also used.

3-15, line 16 – Aren't there about 200 NADP sites?

3-22, line 8 – The term non-ambient loadings is horrible. It doesn't mean anything. Please change it. I would suggest non-atmospheric N sources.

3-27, line 16- Should be Whiteface.

3-27, line 17- Change to: show a downward pattern to 2006.

3-48, line 8- Change to: is fairly uniform…

3-48 line 9- Change to: are generally consistent with…

3-59, line 10- Change to : Whiteface

3-80, line 10- Change to: and thus, decreasing atmospheric…

3-80, line 23- Change to: simulations were run.

3-81, line 23- space between to and 50%

3-82, line 5- Change to: for the pattern is that decreasing NOx decreases HNO3 which limits NO3.. There is a 4 before NO3, should this be HNO3?

3-82, line 5- Change to: This change… also NH4+ superscript on the charge.

3-82, line 8- Should this be dry deposition of NHx?

3-82, line 29- Should this be limits HNO3? Also a space between (NO3) and increases.

Chapter 4

General Comments

The acidification chapter is in need of some editing. Several sections are difficult to follow and the wording needs to be cleaned up. The term inorganic aluminum should be changed to dissolved inorganic aluminum, to clarify that the aluminum is in the aqueous phase not particulate phase. Throughout the chapter the authors refer to ANC concentrations. ANC is not really a concentration. It is a measure of acid base chemistry and represents the composite of many solute concentrations. It is possible to have negative ANC values; concentrations cannot be negative. The authors should refer to it as simply ANC or ANC value

4-14 In the section on critical loads, it is not at all clear what calculations are being done. This needs to be clarified. How are critical loads determined? Also critical load is a steady-state phenomenon, but model calculations were done for 2020 and 2050. Aren't these values really target loads? Finally critical loads are discussed here, but in the Executive summary the term maximum deposition load is introduced. How are these concepts different? In addition to the MAGIC calculations, the steady state water chemistry model (SSWC) is introduced on p 4-36. Both models should be introduced in this methods section. It should be made clear how the models are used and how they complement each other. On 4-36 and 37 the description of the SSWC model is very confusing. This seems to come from left field. This section needs to be re-written to clarify what is being done, and why.

Throughout the chapter the term buffering is used incorrectly. Buffering more specifically pH buffering is the resistance to change in pH. pH buffering can be high due to dissolution of aluminum minerals while contributing to acidification. A better term is the ability to neutralize acid.

4-53, paragraph- I strongly disagree with the statements on steady-state vs dynamic models. Agreed critical loads are a steady-state phenomenon, but ecosystems are not at steady state. A more balanced treatment should be given in the text. There are advantages and disadvantages associated with steady-state and dynamic models, which should be stated. Earlier in the text a dynamic model MAGIC is used for "critical load" calculations. (Although I might argue that these are not critical load calculations, rather target loads.) Application of steady-state models are problematic for ecosystems that are not at steady state. As many sensitive forest ecosystems are exhibiting soil exchangeable cation depletion, one might argue that it is not appropriate to use a steady-state model.

Specific Comments

4-1, line 15- Occult deposition is a poor term. I would just refer to cloud and fog deposition.

4-1, line 21- Change to: depleting soil exchangeable base cations…

4-2, line 2 and throughout the document- Change to: dissolved inorganic aluminum…

4-2, line 27- Need to define nitrogen saturation.

4-3, line 14- Change to: ecosystems and biological species…

4-4, line 1- Change to: available base cation pools of…

4-4, line 14- Change to: chronic or base flow chemistry….occasional acidic episodes, with..

4-4, line 41- Need to define base cation surplus.

4-5, line 21- The statement made concerning PnET is not entirely true. ANC can be calculated as base cations less strong acid anions in PnET-BGC, but PnET also simulates ANC by depicting the protonation of bicarbonate, organic anions and aluminum to represent measured ANC.

4-5, line 24- The statement made is incorrect and contradicts the statement made on line 13. Change to: Low ANC coincides with effects on…

4-13 on Figure 4.2-4 and several of the other figures in the chapter (e.g., 4.2-12, 13, 14) it is a challenge to read the labels and scale on the figures. The font size needs to be increased. The quality of these figures should be improved.

4-13, line 17 and throughout the chapter- Change to: trends in sulfate and nitrate concentrations and ANC…

4-14, line 3- Change to: were used...

4-14, line 6- Change to: because historical measurements are not available.

4-14, line 12- Change to: and low concern (Table 4.2-1).

4-16, line 14- Change to: 0 ueq/L (acidic), 20 ueq/L...

4-16, Table 4.2-1, first row Change to: Near complete loss of fish...

4-17, line 8- Change to have decreased (Figure 4.2-3)…

4-18, Figure 4.2-6- If these are mean values for all lakes this should be made clear.

4-18, line 11- Change to: base cation supply neutralizes the inputs…

4-18, line 17- It is not clear which monitored lakes is being referred to here; the LTM lakes?

4-21, line 9- Change to: less neutralizing ability…

4-21, line 15- Change to: the lake could neutralize and still…

4-29, line 2- Change to: have less ability to neutralize acid inputs than sites……

4-31, line 7- Change to: may degrade by 2050…

4-24, line 22- In addition to inputs of organic acids, some watersheds simply have low rates of base cation supply.

4-36, line 19- What is meant by accounting for effects of chloride?

4-37, paragraphs 2 and 3- This section is confusing. For example on line 21 it states maximum deposition load for sulfur is equal to the amount of sulfur the catchment can remove. On line 14 an assumption is that long-term sinks of sulfate in the catchment is negligible. This section needs to be re-written. I thought I understood the SSWC model until I read this section.

4-38, line 14- Change to: all possible combinations of…

4-38, line 17- Change to: each combination of depositions...

4-40, line 24- Change to: ANC = 50 ueq/L.

4-44, paragraph 1- You also may want to mention that regeneration of sugar maple is restricted under low calcium conditions. See Juice et al. 2006.

4-50, line 17- the ability of a system to neutralize acid…

4-50, line 18- Change to: successfully neutralize acidifying…

4-50, line 20- Change to: to neutralize acidifying…

4-50, line 23- Change to: bedrock with low ability to neutralize acid inputs..

4-55, line 16- This statement is not true. Watershed 6 has been cut and the forest biomass has been impacted by climatic disturbance events. See Aber et al. 2002.

Chapter 5

General comments

The nutrient enrichment chapter is well done. It is good to see a balanced description of the effects of fertilization in the beginning of the chapter. The Potomac and Neuse case studies illustrate the difficulty is quantifying the effects of atmospheric nitrate deposition in complex and diverse watersheds.

The chapter would benefit from a brief description of SPARROW, including the limitations of the model. Using a model like SPARROW will not allow for a determination of the time of recovery in response to decreases in nitrogen loading.

Specific Comments

5-9, line 14- Should the sentence say “at which the system is not nitrogen-limited”?

5-19, last paragraph- I don’t understand incremental yield and delivered yield. Can these terms be clarified?

5-20, line 27- Define TNs.

Figures 5.2-7 and 5.2-11. There appears to be a mistake in the figure legend. Shouldn’t the units be kg/ha-yr? Also more descriptive figure titles defining incremental nitrogen yield and delivered nitrogen yield would be helpful.

5-33, line11- Change to: levels by decreasing the oxidized…

Figure 5.2-16. Note that the concentration scale here is very small, not very meaningful differences in terms of measured concentrations.

5-45, line 18- Space between section 3.3). and Changes..

5-45, line 25- Typo: waters, reduced…

5-45, line 28- Typo: uptake (Figure 5.3-1).

5-48, line 27- Change to: from decreasing nitrogen…

5-58, line 18- Typo: analytical

Chapter 6

General comments

The material on the linkages between mercury and sulfur is generally a good overview. There are some inconsistencies in the text that need to be revised. For example on p -2, line 26 it is stated that inconsequential amounts of methyl mercury can be produced in the absence of sulfate. But on p 6-4, line 12 the text correctly indicates that methyl mercury can be produced by iron reducing bacteria. The first statement should be deleted. On p 6-3, line 5 the statement is made “Watersheds with conditions known to be conducive to mercury methylation have been identified in the northeastern U.S. and southeastern Canada..” but is followed by the statement “ whereas watersheds with elevated methylmercury levels… are seen in most of the U.S. The first phrase should be deleted. It is inconsistent with the second part of the sentence and with figure 6.2-5.

Specific comments

6-1, line 24- Change to: stated that decreases in visibility…

6-3, line 17- Should be Driscoll et al. 2007

6-7, line 34- I believe now all 50 states have some sort on mercury consumption advisory.

6-20, line 16- There seems to be part of a sentence missing.

6-21, line 15- What other types of ecosystems were data available besides conifer and deciduous forests?

6-23, line 5- Standard deviation is given for N2O and CH4, can the variability for CO2 uptake response factor be given.

6-23, line 26- Should this be section 6.4.4?

6-25, line 17- Change to: Because of decreases in ambient SO2…

Chapter 7

General comments

Some of the overall suggestions that I made at the beginning of this report could be incorporated in this section.

7-23, line 22- The information presented in Table 7.1-3 is not quantitative and no information on Bc/Al is presented. This sentence needs to be changed.

Specific comments

7-2, Figure 7-1- I find the font difficult to read on the purple background in this figure.

7-5, Line 10- Change to (i.e., rain, snow), cloud and fog, and dry…

7-5, line 12- Change to: Both are essential. Nitrogen often limits the growth or productivity, and species diversity of ecosystems.

7-5, line 14- Change to: acidification and with excess nitrogen to nutrient enrichment.

7-5, line 16- Change to: localized loss or extinction…

7-7, line 22- Change to: nearly complete loss of…

7-8, paragraph 2- Again maximum depositional load. How is this related to critical load?

7-9, line 2- Change to: lake could neutralize inputs of acids and still…

7-10, line 1- Do you mean: 30 ueq/L in summer than in spring…

7-10, lines 2 and 3- Change drops to decrease.

7-10, line 3- Change to: Severe impacts can occur to fish…

7-10, line 5- Do you mean low or high pulses?

7-10, line 14- Change to: mobilization of dissolved inorganic Al…

7-10, line 25- typo on superscript.

7-10, line 26- Change to: concentrations of available Al, as measured by exchangeable base…

7-11, line 5- Need a period at the end of the table title.

7-11, line 12- Sverdrup and Warfvinge 1993 is not in the references.

7-14, line 21- Do you need the word magnesium?

7-17, line 19- red spruce

7-19, line 29- Is oconic a word? Iconic?

7-21, line 8- Change to: Tahoe drain through…

7-22, line 4- Are continuous monitoring units used for NOx?

7-22, line 16- Change to: data are…

7-23, line 7- Change to ANC and aquatic acidification.

7-23, line 8- Change to: between soil exchangeable Bc/Al ratio and terrestrial acidification.

7-23, line 21- Change to: between ANC and fish species…

7-23, line 22- The information presented in Table 7.1-3 is not quantitative and no information on Bc/Al is presented.

7-24, line 13- Change to: sustain terrestrial and aquatic food chains.

References

Aber, J.D., S.V. Ollinger, C.T. Driscoll, G.E. Likens, R.T. Holmes, R.J. Freuder and C.L. Goodale. 2002. Inorganic N losses from a forested ecosystem in response to physical, chemical, biotic and climatic perturbances. Ecosystems 5:648-658.

Juice, S.M., T.J. Fahey, T.G. Siccama, C.T. Driscoll, E.G. Denny, C. Eager, N.L. Cleavitt, R. Minocha, and A.D. Richardson. 2006. Response of sugar maple to calcium addition to northern hardwood forest. Ecology 87(5):1267-1280.

Dr. H. Christopher Frey

GLOSSARY:

The definition of variability in the glossary is weak and misleading. The explanation of variability given on page 2-18 is much better. The glossary should be updated to better reflect accepted definitions of risk assessment terminology.

EXECUTIVE SUMMARY

Please define NO_y in the text box.

Figure ES-2: The term "atmospheric landscape" is confusing. One does not associate a landscape with the atmosphere. Use a more descriptive term.

Page ES-4: It is helpful to list what was analyzed. However, some explanation of what was not analyzed and why would also be helpful. For example, earlier it is indicated that sulfur is a nutrient, and a reader may wonder therefore why sulfur "enrichment" is not considered.

Figure ES-3. Remove the brown background color, because it is not useful.

Page ES-8. The "concept of ecosystem services" is mentioned but not defined. It should be explained to the reader.

Page ES-9 . The allusion to monetization in the first paragraph seems entirely inappropriate for this document. By law, the costs associated with a potential standard cannot be considered in setting the standard. Why ***should*** any of the endpoints be monetized in the context of this document? The issue of monetization related to benefits assessment, which may be relevant to a regulatory impact analysis or to information requested by OMB, but is not relevant to the process of developing NAAQS. Hence, either delete this material throughout the document, or provide a context that clearly differentiates that it is not related to standard setting and is mentioned for other reasons, and enumerate what those reasons are.

Page ES-10. Since CMAQ is a framework, it is non-specific to say that "CMAQ" was used to simulate concentrations. The reader would need to know more about what versions of components within CMAQ were used – i.e. what chemical mechanism, what advection algorithms, what treatment of planetary boundary layer, what "light" model, etc.

Figure ES-5. What layer of the atmosphere is represented here? Is it the lowest layer? Similar comment for Figure ES-6.

Figure ES-7. In lower right, it appears that there is a minus sign but some explanation should be given as to why "Total N – CMAQ dry + NADP Wet"

Page ES-14. For policy relevant background concentrations, please indicate how these were estimated: where they estimated based on monitored values at remote measurement stations, or are they based on model estimates?

Page ES-15. What was the finding regarding whether "area-based risk and exposure assessments are … suitable"?

Page ES-16. The level of detail on this page and a few others that follow seems to ramp up significantly. Who is your audience? If the ES is intended to be read by lay policy people, they will either stop reading here or eyes will glaze over. Try to write in shorter sentences and paragraphs. For example, the paragraph that starts on page ES-15 and goes onto page ES-16 is nearly a page long. The text box contains a run-on sentence and awkward and unclear definition of critical load. There is much repetition in the second paragraph on page ES-16. The phrase "ANC values" or "ANC limits" or similar is repeated three times.

Page ES-17. Top of page: "It was not possible to… a larger dataset" What larger dataset was desired? Larger in what way? This sentence is unclear.

"may aid" does this mean that the writer is not sure if the "connection" helps determine "adverse impacts"? Why so tentative?

The section non Terrestrial Acidification is likewise not likely to be read by a lay policy reader. Is the audience intended to be peer experts in terrestrial acidification? If so, they can probably read this. Doubtful that others can, however. May want to start out differently, explaining and introducing concepts step by step. i.e. soil acidification is bad, and explain why. Then explain that acidification of soil changes the concentrations of Ca++ and Al, and why. Then introduce that Ca++ is part of a "base cation" (explain what that is) that includes a few other ions. Then introduce the ratio of Bc to Al, and explain that it gets smaller or larger as acidification gets more severe. Etc.

Last paragraph on the page. Do the numbers 0.6, 1.2, and 10 have units? The way the sentence is written, it is not clear that these are Bc/Al ratios, since the sentence says that Bc/Al was calculated from the number given. Very unclear to the reader.

Page ES-18. What is a Al/Ca++ imbalance? In what way are Al and Ca++ supposed to be in "balance"? And why is the ratio now inverted? These kinds of inconsistencies are a good way to lose the reader.

Middle of page, the monetization seems entirely inappropriate. See earlier comment.

Page ES-20. Please clarify if "atmospheric deposition" includes only direct deposition or if indirect pathways are included – e.g., does deposition to land and then run-off into estuaries constitute "atmospheric deposition" to the estuary? It is a bit odd and confusing to the reader to say that more than 100% or greater reduction in atmospheric deposition was necessary. How can one have more than 100% reduction in deposition?

Bottom of page… e.g., 1.5 kg N/ha – should there be a time dimension to this number? i.e. over what time period did this amount of deposition take place?

Page ES-22: The term "mycorrhizal community" is mentioned before it is defined. This will confuse lay readers who are not experts in the topic area.

Table ES-3. Is the area really known to 6 significant figures?

Bottom of page ES-22. "The pressures exerted" seems to be a metaphor. In technical writing, avoid technically-based metaphors, as they can be confused for a literal interpretation, depending on the background of the reader. Since my background is mechanical engineering, I tend to think of pressure in terms of force per area. I don't think this is the intended meaning.

Page ES-24. It is not clear to the reader as to why the critically important role of N2O as a greenhouse gas is beyond the scope of this assessment. If they are beyond the scope of this "review," then why were they addressed qualitatively. It is contradictory to say they were beyond the scope but that they were included in the assessment, even if qualitatively. Why is an assessment of the effect of N2O on climate of necessity qualitative? Why can't it be quantitative? What are the key findings of the qualitative assessments of these endpoints?

There needs to be a section that addresses the key findings, conclusions, and implications associated with assessment of uncertainty and variability. What are the largest sources of uncertainty and variability? Given the uncertainty and variability, what findings can be made with a high degree of confidence? Which can't?

CHAPTER 7. My review focused on the section regarding uncertainty.

The section on uncertainty, starting on page 7-22, reads like the typical qualitative "laundry list" approach of acknowledging uncertainties but not characterizing them. This is, frankly, unacceptable.

The National Research Council (NRC, 1994) stated the need to describe uncertainty and to capture variability in risk estimates. Risk characterization became EPA policy in 1995, and the principles of transparency, clarity, consistency, and reasonableness are explicated in the 2000 Risk Characterization Handbook (EPA, 2000). Transparency, clarity, consistency, and reasonableness criteria require analysts to describe and explain the uncertainties, variability, and known data gaps in the risk analysis and imply that decision makes should explain how they affect resulting decision-making processes (USEPA, 2000, 1992, 1995).

On numerous occasions, the NRC has explicitly called for the use of probabilistic risk assessment (NRC, 2007a,b). NRC (1992) recommended that EPA should thoroughly discuss uncertainty and variability in the context of ecological risk assessment (NRC, 1993). NRC (1994), in a major review of risk assessment methodology, stated that "uncertainty analysis is the only way to combat the 'false sense of certainty,' which is caused by a refusal to acknowledge and [attempt to] quantify the uncertainty in risk predictions." NRC (2002) suggested that EPA's

estimation of health benefits were not wholly credible because EPA failed to deal formally with uncertainties in its analyses.

EPA's Science Advisory Board has made recommendations similar to those of the NRC. Parkin and Morgan (2007) urged the Agency to characterize variability and uncertainty more fully and more systematically and to replace single-point uncertainty factors with a set of distributions using probabilistic methods (Parkin and Morgan, 2007). EPA has developed numerous internal handbooks on how to conduct quantitative analysis of uncertainties in various contexts (e.g., EPA, 1995; 1997; 1998; 2000; 2001) EPA (2009) provides a detailed overview of the current use of probabilistic risk analysis within EPA (including 16 detailed case study examples), an enumeration of the relevance of PRA to decision-making, common challenges faced by decision makers, an overview of PRA methodology, and recommendations regarding how PRA can support regulatory decision making. EPA's National Exposure Research Laboratory has recently explored methodological issues for dealing with uncertainty quantitatively when coupling models for air quality, exposure, and dose (Ozkaynak et al., 2008).

There are numerous texts on how to conduct analysis of uncertainty (e.g., Morgan and Henrion, 1990; Cullen and Frey, 1999; Vose, 2008). The World Health Organization has recently released guidance on qualitative and quantitative methods for uncertainty analysis in the context of exposure assessment (WHO, 2008). These guidelines have been used by EPA to support uncertainty assessments of exposure and health effects for criteria pollutants under the National Ambient Air Quality Standards (NAAQS). Hence, the framework is a general one. In particular, WHO proposed guiding principles that are adapted here:

- Uncertainty analysis should be an integral part of the assessment.
- The objective and level of detail of the uncertainty analysis should be based on a tiered approach and be consistent with the overall scope and purpose of the assessment.
- Sources of uncertainty and variability should be systematically identified
- The presence or absence of moderate to strong dependencies between inputs is to be discussed and appropriately accounted for.
- Data, expert judgment, or both should be used to inform the specification of uncertainties for scenarios, models, and inputs.
- Sensitivity analysis should be an integral component of the assessment.
- Uncertainty analyses should be fully and systematically documented in a transparent manner, including: quantitative aspects pertaining to data, methods, inputs, models, outputs; sensitivity analysis; qualitative aspects; and interpretation of results
- The results of the assessment including the uncertainty should be subject to an evaluation process that may include peer review, model comparison, quality assurance, or comparison to relevant data or independent observations.
- Where appropriate to an assessment objective, assessments should be iteratively refined over time to incorporate new data, information and methods in order to reduce uncertainty and improve the characterization of variability.
- Communication of assessment uncertainties to the stakeholders should reflect the different needs of the audiences in a transparent and understandable manner.

The decision context of risk assessment includes: (a) how to prioritize the activities of the assessment, and development of data for the assessment, in order to characterize and, where possible, reduce uncertainty; (b) how best to manage risk. Decision makers often want to know: who is at risk; the magnitude of risk; and tradeoffs between risk management alternatives. Examples of specific questions that decision-makers may ask include (Bloom et al., 1993; Krupnick et al., 2006):

- How representative is the estimate, (e.g., what is the variability around an estimate)?
- What are the major gaps in knowledge, and what are the major assumptions used in the assessment? How reasonable are the assumptions?
- Would additional data collection and research likely lead to a different decision? How long will it take to collect the information, how much would it cost, and would the resulting decision be significantly different?

Generally, from a scientific perspective, it is preferred to quantify uncertainties wherever possible. As WHO (2008) explains (p. 31):

> *Determination of an appropriate level of sophistication required from a particular uncertainty analysis depends on the intended purpose and scope of a given assessment. Most often tiered assessments are explicitly incorporated within regulatory and environmental risk management decision strategies. The level of detail in the quantification of assessment uncertainties, however, should match the degree of refinement in the underlying exposure or risk analysis. Where appropriate to an assessment objective, exposure assessments should be iteratively refined over time to incorporate new data, information and methods to reduce uncertainty and improve the characterization of variability. Lowest-tier analyses are often performed in screening-level regulatory and preliminary research applications. Intermediate tier analyses are often considered during regulatory evaluations when screening-level analysis either indicates a level of potential concern or is not suited for the case at hand. The highest tier analyses are often performed in response to regulatory compliance needs or for informing risk management decisions on suitable alternatives or trade-offs.*

Hence, the Tier 1 (Qualitative) approach is not a default. It should be a justified choice that is consistent with the purpose and scope of the assessment.

WHO specifies a structured approach to qualitative assessment of uncertainty that includes

1) qualitatively evaluate the level of uncertainty of each specified source;
2) define the major sources of uncertainty;
3) qualitatively evaluate the appraisal of the knowledge base of each major source;
4) determine the controversial sources of uncertainty;
5) qualitatively evaluate the subjectivity of choices of each controversial source; and
6) reiterate this methodology until the output satisfies stakeholders

Somewhat amazingly, the document seems to ignore an effort undertaken in the mid 1990s to create a quantitative framework for characterizing variability and uncertainty associated with

acid deposition, which is the **Tracking and Analysis Framework (TAF)**. The details of this framework, and the associated software, are available at http://www.lumina.com/taf/. TAF represents a systematic effort sponsored by the National Acid Precipitation Assessment Program (NAPAP) to develop an "integrated assessment" model that represents emissions, transport and fate, deposition, and adverse effects to terrestrial and aquatic systems, and for which variability and uncertainty were quantified. The philosophy of TAF was to develop a reduced form model that was tractable and that could be used for repetitive analysis, such as "what-if" policy scenarios, sensitivity analysis, and assessment of the effect of variability and uncertainty on estimated end points. This approach is a practical one, and should serve as an illustrative example of how variability and uncertainty can be quantified in an assessment such as needed in the REA. EPA should review TAF and either use this framework, or update it and use it.

A laundry list of uncertainties, as given in Section 7.2 is not useful in interpreting the assessment results unless it is conducted in a systematic manner that puts priority on quantification and that leads to comparative assessments of which sources of uncertainty are important with respect to well defined endpoints. At a minimum, there should be summary tables that categorize sources of uncertainty with respect to a well defined endpoint. What are the implications of uncertainty for interpretation of the assessment results?

Why is there a benefits assessment component to Chapter 7? As noted in comments regarding the executive summary, costs cannot be considered when setting the NAAQS. What is the purpose of providing monetized assessment of benefits in the context of this REA?

References Cited and Bibliography (not all references listed below are cited):

Bloom, D.L., D.M. Byrne, and J.M. Andreson. 1993. Communicating Risk to Senior EPA Policy-Makers: A Focus Group Study. Prepared by Bloom Research and the Office of Air Quality Planning and Standards. U.S. Environmental Protection Agency, Research Triangle Park, NC.

Bogen, K., A. Cullen, H.C. Frey, and P. Price, "Probabilistic Exposure Analysis for Chemical Risk Characterization," Toxicological Sciences, submitted August 20, 2008, revised and resubmitted Jan 2009, accepted February 8, 2009. Available on-line at http://toxsci.oxfordjournals.org/cgi/reprint/kfp036?ijkey=sIBW9Sm3pvbPQnm&keytype=ref

Cullen, A.C., and H.C. Frey. 1999. Probabilistic Exposure Assessment: A Handbook for Dealing with Variability and Uncertainty in Models and Inputs. Plenum Press, New York.

EPA. 1997a. Guiding Principles for Monte Carlo Analysis. EPA/630/R-97/001, Risk Assessment Forum. U.S. Environmental Protection Agency, Washington.

EPA. 1998. Guidance for Submission of Probabilistic Human Health Exposure Assessments to the Office of Pesticide Programs. Office of Pesticide Programs. U.S. Environmental Protection Agency, Draft, 11/4/98, http://www.epa.gov/fedrgstr/EPA-PEST/1998/November/Day-05/6021.pdf (accessed August 8, 2007).

EPA. 2000. EPA Science Policy Council Handbook, Risk Characterization. EPA 100-B-00-002.

EPA. 2001. Risk Assessment Guidance for Superfund: Volume III - Part A, Process for Conducting Probabilistic Risk Assessment. EPA 540-R-02-002, Office of Emergency and Remedial Response, U.S. Environmental Protection Agency, Washington, http://www.epa.gov/oswer/riskassessment/rags3adt/ (accessed August 8, 2007).

EPA. 2007. Letter from M.G. Morgan and R.T. Parkin (SAB) to S. Johnson (EPA), February 28, 2007. EPA-SAB-07-003.

EPA, 2009, Using Probabilistic Methods to Enhance the Role of Risk Analysis in Decision-Making, External Review Draft, Prepared by Risk Assessment Forum PRA Technical Panel Working Groups, U.S. Environmental protection Agency, Washington, DC.

Evans, J.S., J.D. Graham, G.M. Gray, and R.L. Sielken. 1994. A Distributional Approach to Characterizing Low-Dose Cancer Risk. Risk Analysis, 14(1):25-34.

Frey, H.C., and S.R. Patil. 2002. Identification and Review of Sensitivity Analysis Methods. Risk Analysis, 22(3):553-578.

Hattis, D.B., and D.E. Burmaster. 1994. “Assessment of Variability and Uncertainty Distributions for Practical Risk Analyses,” Risk Analysis, 14(5):713-730.

Hoeting, J.A.; D. Madigan, A.E. Raftery, and C.T. Volinsky. 1999. Bayesian Model Averaging: A Tutorial. Statistical Science, 14(4):382-417.

IEC. 2006. Expanded Expert Judgment Assessment of the Concentration-Response Relationship Between PM2.5 Exposure and Mortality, Final Report. Prepared by Industrial Economics, Incorporated for Office of Air Quality Planning and Standards, U.S. Environmental Protection Agency, Research Triangle Park, NC, http://www.epa.gov/ttn/ecas/regdata/Uncertainty/pm_ee_report.pdf.

Krupnick, A., R. Morgenstern, M. Batz, P. Nelson, D. Burtraw, J. Shih, and M. McWilliams. 2006. Not a Sure Thing: Making Regulatory Choices Under Uncertainty. Resources for the Future, Washington.

Mokhtari, A., H.C. Frey, and J. Zheng. 2006. Evaluation and Recommendation of Sensitivity Analysis Methods for Application to Stochastic Human Exposure and Dose Simulation (SHEDS) Models. Journal of Exposure Science and Environmental Epidemiology, 16(6):491-506.

Morgan, M.G., and M. Henrion. 1990. Uncertainty: A Guide to Dealing with Uncertainty in Quantitative Risk and Policy Analysis. Cambridge University Press, New York.

NRC. 1983. Risk Assessment in the Federal Government: Managing the Process. Committee on the Institutional Means for Assessment of Risks to Public Health, National Academy Press, Washington.

NRC. 1994. Science and Judgment in Risk Assessment. National Research Council, National Academy Press, Washington.

NRC. 2002. Estimating the Public Health Benefits of Proposed Air Pollution Regulations. National Research Council, National Academy Press, Washington.

NRC. 2007b. Models in Environmental Regulatory Decision Making. National Research Council, National Academy Press, Washington.

OSTP/OMB. 2007. Memorandum (M-07-24) for the Heads of Executive Departments and Agencies; from Susan E. Dudley, Administrator, Office of Information and Regulatory Affairs, Office of Management and Budget, to Sharon L. Hays, Associate Director and Deputy Director for Science, Office of Science and Technology Policy: Updated Principles for Risk Analysis, September 19, 2007.

Ozkaynak, H., H.C. Frey, J. Burke, and R.W. Pinder, "Analysis of coupled model uncertainties in source to dose modeling of human exposures to ambient air pollution: a PM2.5 case-study," Atmospheric Environment (accepted December 4, 2008).

Parkin, R.T., and M.G. Morgan. 2007. Consultation on Enhancing Risk Assessment Practices and Updating EPA's Exposure Guidelines. EPA-SAB-07-003, Science Advisory Board, U.S. Environmental Protection Agency, Washington.

PCRARM. 1997. Framework for Environmental Health Risk Management Final Report, Volume 1. Presidential/Congressional Commission on Risk Assessment and Risk Management, http://www.riskworld.com/nreports/1997/risk-rpt/pdf/EPAJAN.PDF.
Pearl, J. 2000. Causality: models, Reasoning, and Inference. Cambridge University Press, New York.

RFF (2007), "Uncertainty Modeling in Dose Response: Dealing with Simple Bioassay Data, and Where Do We Go from Here?," Workshop, Resources for the Future, Washington, DC, October 22 - 23, 2007. (http://www.rff.org/Events/Pages/Cooke-Uncertainty-Workshop.aspx, accessed 6/27/09).

Royall, R.M. 1997. Statistical Evidence: A Likelihood Paradigm. Chapman & Hall, London.
Saltelli A, S. Tarantola, F. Campolongo, et al. 2004. Sensitivity Analysis in Practice. John Wiley & Sons, New York.

Small, M.J. (2008) Methods for Assessing Uncertainty in Fundamental Assumptions and Associated Models for Cancer Risk Assessment Risk Analysis, Vol. 28, No. 5, 2008 DOI: 10.1111/j.1539-6924.2008.01134.x

Stahl, C.H., and A.J. Cimorelli. 2005. How Much Uncertainty Is Too Much and How Do We Know? A Case Example of the Assessment of Ozone Monitor Network Options. Risk Analysis, Vol. 25, No. 5, pp. 1109-1120, October 2005.

Vose, D. (2008), Risk Analysis: A Quantitative Guide, Third Edition, John Wiley and Sons, West Sussex, England.

World Health Organisation (2008), Harmonization Project Document No. 6, Part 1: Guidance Document on Characterizing and Communicating Uncertainty in Exposure Assessment, International Program on Chemical Safety, World Health Organization, and Co-sponsored by International Labour Organization, and the United Nations Environmental Programme, WHO Geneva, Switzerland, 2008.
(http://www.who.int/ipcs/publications/methods/harmonization/exposure_assessment.pdf)