**September 24, 2008**

**Draft Comments from Myron J. Mitchell on "Risk and Exposure Assessment to Support the Joint Review of the $NO_2$ and $SO_2$ Secondary National Ambient Air Quality Standards: Preliminary Draft"**

**General Comments** (For these general comments my responses are in *italics.) The document and associated attachments are extensive and it is a major challenge to provide a cohesive approach to presenting this information. I recognize the difficulty and complexity of pulling all of this together. The potential for providing clearer linkages in this document to the "Integrated Science Assessment for Oxides of Nitrogen and Sulfur –Environmental Criteria" may help maintain focus and reduce the size of the document. More consistency is needed in the use of terms such as oxides of sulfur, oxides of nitrogen, sulfur oxides, nitrogen oxides, $SO_x$, $NO_x$, etc. Also, in the document there are differences in criteria for sensitivity of various parameters such as ANC. Careful editing would help focus the document and reduce redundant information. The draft document has many noun trains such as "draft revised criteria document."*

**Responses to Charge to the CASAC $NO_x$/$SO_x$ Secondary Review Panel**

Scope of the review (Chapters 1 and 2):

1. Chapters 1 and 2 provide the background, history, and the framework for this review, including a discussion of our focus on the four key ecological effect areas (aquatic acidification, terrestrial acidification, aquatic nutrient enrichment, terrestrial nutrient enrichment). Is this review appropriately focused in terms of the targeted effect areas and in terms of characterizing the important atmospheric and ecological variables the influence the deposition and, ultimately, the ecological impacts of nitrogen and sulfur? Does the Panel have any further suggested refinements at this time?

*Chapter 1 needs a more balanced introduction with respect to the impacts of S and N gaseous constituents and deposition with greater attention earlier in each chapter on the effects of N on ecosystem structure and function including the alteration of species composition. In Chapter 2 the description of errors in the analyses needs a more rigorous approach that more clearly identifies specific issues related to sources and amount of errors. In its current form Chapter 2 provides very broad descriptions that do not clearly identify the major sources of error that are important to the current assessment.*

Air quality analyses (Chapter 3):

1. To what extent are the air quality characterizations and analyses presented in Chapter 3 technically sound, clearly communicated, appropriately characterized, and relevant to the review of the secondary NAAQS for $NO_x$ and $SO_x$?

*The terminology and analyses associated with "policy-relevant background concentrations" is confusing. Some consideration is needed of the adequacy of the current monitoring network in evaluating air quality especially in relationship to measurements in urban settings.*

2. Section 3.2.1 describes an approach for evaluating the spatial and temporal patterns for nitrogen and sulfur deposition and associated ambient concentrations in the case study locations. This draft document includes the analysis for the Adirondacks case study. Does the Panel agree with this approach and should it be applied to the other Case Study Areas?

*This can be a helpful approach. However, within each of these regions there have been other measurements made of associated parameters including precipitation amount, atmospheric deposition, etc. It does not appear that any substantial attempts are being made in this document to compare these current results with previously published results? There is a major emphasis on providing both spatial and temporal patterns using the CMAQ modeling results. Shouldn't these modeling results be compared with other results of deposition for this region? Having some comparisons with other measurements would be helpful in evaluating these deposition estimates.*

3. Section 3.2.2 describes the relative contributions of ambient emissions of nitrogen and ammonia to nitrogen deposition for the case study areas. To what extent is the approach taken technically sound, clearly communicated, and appropriately characterized?

*Clarification is needed on the relative importance of the emission sources including the accuracy of these emission estimates, atmospheric chemical conversions and the estimates of deposition velocities in evaluating the results of these model outputs. Too much of this section describes model output with little supporting evidence of the relationships noted.*

Case Study Analyses (Attachments 2-6):

*These attachments are extensive and it would be most helpful if they were more targeted to the issues and needs of the "Risk and Exposure Assessment to Support the joint Review of the $NO_2$ and $SO_2$ Secondary National Ambient Air Quality Standards*."

1. Attachment 2 presents a GIS analysis to define geographical areas that are sensitive to acidification and nutrient enrichment. Are the national geospatial datasets chosen adequate to identify sensitive areas? Are there other data sets that have not been identified by this analysis that we should consider? Does the Panel agree with this approach or can they suggest alternatives?

*In general, the selected regions and associated data sets appear to be appropriate for this analysis.*

2. Attachment 3 presents our current progress on evaluating the effect of aquatic acidification in the Adirondacks. It describes the use of the MAGIC model to evaluation ANC levels in selected lakes and streams in the Adirondacks and Shenandoahs. To what extent is the approach taken technically sound, clearly communicated, and appropriately characterized?

*There are some differences in the deposition estimates being used for estimates using the MAGIC versus other approaches in the document (e.g., ASTRAP versus CMAQ). Will this result in some issues relating to congruity of analyses in the document? These analyses have a specific focus on the Grimm and MAGIC models. There have been other efforts to evaluate acid rain/biogeochemical responses in each of these sites (especially the Adirondacks) and hence it would be useful to include some of these other results to indicate either differences or support of these current modeling efforts. In these discussions different levels of ANC are suggested that differ than for other sections in the document. A more consistent approach is needed on setting the ANC limits of concern with respect to sensitivity to acidification and recovery from acidification.*

3. Attachment 4 presents our current progress on evaluating the effect of terrestrial acidification. It outlines a plan to use the Simple Mass Balance Model to evaluate current deposition levels on forest soil ANC for sugar maple in the Kane Experimental Forest and red spruce in the Hubbard Brook Experimental Forest. To what extent is the approach taken technically sound, clearly communicated, and appropriately characterized?

*This portion of the document is highly uneven within some cases substantial detail provided and in other places there are broad generalizations. This makes it difficult for the reader to ascertain the salient points.*

4. Attachment 5 presents our current progress on evaluating the effect of aquatic nutrient enrichment. It outlines a plan to evaluate how changes in nitrogen deposition affect the eutrophication index in two estuaries: the Chesapeake Bay and Pamlico Sound. The analysis will model one stream reach (Potomac River and Neuse River) to determine the impact on the eutrophication index for the estuary. To what extend is the approach taken technically sound, clearly communicated, and appropriately characterized?

*No comment at this time.*

5. Attachment 6 presents our current progress on evaluating the effect of terrestrial nutrient enrichment. It describes an approach to evaluate the effects of nitrogen deposition on the Coastal Sage Scrub community in California and in mixed conifer forests in the San Bernardino and Sierra Nevada Mountains. To what extent is the approach taken technically sound, clearly communicated, and appropriately characterized?

*No comment at this time.*

Additional Effects (Chapter 6):

1. In this chapter, we have presented results from some initial qualitative analyses for additional effects including the impact of sulfur deposition on mercury methylation, the impact of nitrous oxide on climate change, and the impact of nitrogen deposition on carbon sequestration. Are these effects sufficiently addressed in light of the focus of this review on the other targeted effects and in terms of the available data to analyze them?

*These descriptions seem adequate, but certainly the issues go beyond those associated with carbon sequestration.*

The purpose of Chapter 7 is to summarize the Case Study results and characterize the relationships between levels of an ecological indicator and the associated degree of ecologically adverse effects. To what extent is this approach technically sound, clearly communicated and appropriately characterized at this point of the review? Does the Panel have any further suggested refinements at this time?

*In its current form this Chapter is highly descriptive. Future revisions will need to include more specific quantitative results that show clear relationships to the assessment.*

Chapter 8 begins to explore how a secondary NAAQS might be structured to address the targeted ecological effects discussed in the risk assessment. The next draft of this document will include one or more examples of how this structure might be used to related specific levels of air quality indicators with a corresponding ecological indicator for a given location and/or scenario. To what extent is the described approach technically sound, clearly communicated and appropriately characterized at this point of the review. Does the Panel have any further suggested refinements at this time.

*Clearly providing a clear scientific linkage that is policy relearnt between concentrations of $SO_x$ and $NO_x$ and resultant S and N deposition and subsequent translation to ecosystem level effects is a major challenge. There are good linkages between concentrations of $SO_x$ and $NO_x$ and deposition, but the linkage to ecosystem level effects is more complicated due to inherent variability across the United States of the sensitivity of these systems. There are major problems with respect to both having confidence in calculated deposition velocities and how to ascertain "functional forms."*

**Specific Comments in which *italics* indicate specific changes.**

| Page(s) | Lines(s) | Comment |
|---|---|---|
| xv | 3-5 | Why should this definition only include terrestrial ecosystems? |

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| 1-1 | 8 | Change to "The NAAQS *have been* established for pollutants." |
| 1-2 | 8 | Change to "complex interactions among relevant *chemical* species of." |
| 1-3 | 10 | Clarify more specifically who is "we." Do you mean EPA? |
| 1-3 | 19 | "Chapter 2" should be in bold font. |
| 1-4 | 1-5 | This does not agree with provided definition on nitrogen enrichment (page xv, lines3-5) |
| 1-5 | 13 | Change to "*This draft*." |
| 1-5 | 15 | Delete "held." |
| 1-5 | 24 | Change to "identified the critical *components* to be." |
| 1-5 | 25 | Specify the title of the "Administrator" here and elsewhere in the document. |
| 1-6 | 5 | Change "NAAQS for NO2 *was set at* 0.053 ppm." |
| 1-6 | 10 | Change to "secondary NAAQS values for $SO_2$ under." |
| 1-6 | 13 | Change to "of evidence from vegetation effects." |
| 1-6 | 19 | Change to "EPA was aware that $SO_x$ *has* other." |
| 1-6 | 25 | Here and elsewhere be consistent in the use of terms $SO_x$ and sulfur dioxides. |
| 1-7 | 17 | Change "completed by EPA at *this time*." |
| 1-7 | 20-23 | Change to "Administrator's conclusions that (1) based upon the then-current scientific understanding of acid deposition, it would be premature and unwise to prescribe any regulatory control program, and (2) when the scientific uncertainties had been *substantially* reduced through ongoing research efforts, EPA would draft and support an appropriate set of control measures". |
| 1-8 | 3 | Change to "*Due to* the complexities and *substantial* remaining." |

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| 1-8 | 20-21 | Change to "long-term." |
| 1-9 | 28 | Be more explicit with respect to this "Federal Register notice." |
| 1-10 | 7 | Why "beyond full implementation of Title IV….?" |
| 1-10 | 7 | Change "to presented in the *2005 NAPAP* Report." |
| 1-10 | 10 | Be more specific with respect to which "recent reports." |
| 1-10-11 | | It would be preferable to start with a clearer definition of the different forms of nitrogen oxides and then discuss some of the confusing terminology. |
| 1-11 | 6 | Is another group of individuals using different terminology for nitrogen oxides? |
| 1-11 | 12 | Change to "deposition of *reactive* nitrogen" |
| 1-11 | 19 | Change to "In many *regions*." |
| 1-11 | 22 | Change to "assessing *the impacts of nitrogen deposition*." |
| 1-11 | 24-26 | This is not true most research does consider separately the oxidized and reduced forms of nitrogen inputs especially for natural ecosystems. In many cases the total input of reduced nitrogen is less well known than that of oxidized nitrogen. |
| 1-12 | 13-14 | With reduction in S deposition the relative importance of N deposition in contributing to acidification has increased. This should be noted in this introductory information. |
| 1-13 | 5 | The statement "sometimes limiting" with respect to N is misleading. Nitrogen is often a limiting nutrient with respect to primary production in terrestrial ecosystems. Even though N may be a limiting nutrient does not necessarily imply, however, that there may not be substantial ecosystem damage. Changing N inputs can alter ecosystem structure including altering the system from a more desirable state. Such factors are especially important with respect to the evaluation of N deposition in the western U.S. Such secondary effects need to be considered in setting standards. |
| 1-13 | 6 | Change to "acidification, nutrient enrichment, eutrophication, and |

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| | | changes in species composition." |
| 1-14 | 7-9 | Other effects including changes in species composition and community structure need to be included for both terrestrial and aquatic systems especially associated with N inputs. |
| 1-14 | 10-24 | Clearly there have been substantial studies of acidic deposition effects in the eastern U.S., but some consideration is needed of effects in the western U.S. where the effects appear to be substantially different especially with respect to N deposition influences. |
| 1-14 | 26 | Does the reference to "high elevation lakes" refer to the western U.S.? |
| 1-14 | 31 | In this section the issue of changes in "community composition" is mentioned and this needs to be included earlier in the document. |
| 1-15 | 1-4 | This statement on alteration of aquatic communities needs to be made congruous with earlier statements on overall effects of S and N deposition. |
| 1-15 | 22 | Change to "*a*lpine." |
| 1-16 | 5 | Provide some information on what are these "other factors." Do you mean periods of relatively high microbial activity? |
| 1-16 | 8 | Be more explicit on the relationship (e.g., positive or negative) between "oxygen content, temperature, pH, and supply of labile organic carbon." |
| 1-16 | 11 | Be more explicit on what are these conditions. Do you mean for conditions the amount of wetlands and linkages to drainage waters? |
| 1-16 | | Figure 1.3-3 should explicitly have a category for changes in species composition that may differ from altered biodiversity. |
| 2-1 | 9-12 | This listing needs to explicitly include effects on community structure and species composition. |
| 2-1 | 16-18 | This statement begins to raise the issue associated with different effects depending on region in the U.S. |

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| 2-1-2 | | The absence of case studies in the Rocky Mountain west is problematic. |
| 2-4 | | This introduction can be improved with a more focused approach. There is considerable repetition in this part of the document. |
| 2-5 | 4-5 | Change to "data were *not sufficiently available* to perform." |
| 2-5 | 18 | Clarify in scaling-up clarify how this will be done with respect to sensitive areas (e.g., will this be done for all sensitive areas or will the scaling be a function of other sensitive areas with similar characteristics? |
| 2-6 | 27 | Change to "*need* for them." |
| 2-6 | 28 | A citation is needed for the "MEA" here. Later it is indicated that MEA (2005). |
| 2-7 | 3 | Care should be made in the use of the term "regulating" since this term has a different meaning in the context of environmental regulations such as those associated with laws formulated by the EPA. Maybe a clarifying phrase is needed to avoid any confusing recognizing that the term regulation has a different meaning in an ecological context. |
| 2-6 | 14 | Are there different references for the MEA? Here it is indicated: MEA, 2005b. Also see Line 17 (MEA, 2005b). |
| 2-7 | | This section needs some further articulation with respect to the description of ecosystem services. Is this description based solely on what is provided in (U.S. EPA, 2006)? It would be helpful to provide some further references that link these statements to other informational sources. |
| 2-7 | 16 | Change to "interest in the risk assessment *described in the current document*." |
| 2-7 | 20-22 | A better description is needed that clarifies the association of $NO_x$ deposition and eutrophication. |
| 2-7 | 23-25 | This statement is poorly worded. |
| 2-7 | 26-31 | The wording of this section needs much better clarification. It is |

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| | | confusing whether a linkage or several linkages are being evaluated. |
| 2-8 | | Figure 2.2-2 Ecosystem Services: the ecological processes or functions having monetary or non-monetary value to *individuals or society*. |
| 2-9 | 1-2 | Change to "We have begun identifying the primary ecosystem service(s) affected by *either* acidification *and/or* enrichment and for major ecosystem types and components (i.e., terrestrial ecosystems, soils." |
| 2-9 | 4 | Change to "The impacts *affecting* various." |
| 2-9 | 6-7 | Change to "*These impacts on ecosystem services* will be." |
| 2-9 | 10 | Change to "tourism. *Effects on fisheries* (decreased." |
| 2-9 | 21 | Change to "tourism. Effects on fisheries." |
| 2-9 | 27-28 | Change to "hazard mitigation. *Methods for evaluating linkages to measurements and ecosystem* services may include." |
| 2-10 | 5 | Clarification is needed on what is meant by "community." |
| 2-10 | 7 | Change to "changes in stand density, shifts in." Densification is jargon. Change to "shifts in lichen community species composition." |
| 2-10 | 14 | Delete "intake." |
| 2-10 | 15-16 | Why distinguish between "Native Americans and Alaska native Villagers?" |
| 2-13 | 2-4 | Change to "*Our approach in evaluating risk assessment relies upon various analytical tools and techniques, data sources, and other forms of analyses each of which has inherent uncertainties* ." |
| 2-13 | 6 | Change to "affect the *level* of its response." |
| 2-13 | 10-11 | This is not a very succinct or complete description of the errors associated with modeling approaches. |

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| 2-13 | 12 | Change to "involved *with the transformation and fluxes of nitrogen and sulfur constituents*." |
| 2-13 | 14-15 | It would be more helpful to be more explicit in describing the sources of uncertainty. For example with respect to deposition, wet deposition of S and N can be relatively well characterized compared to dry deposition. The major issue associated with the errors related to dry deposition are associated mostly with the modeling of deposition velocities. Also, be more specific in what is being implied with the use of the term "ecological modeling." |
| 2-13 | 14-21 | This section needs to include more specific information. It would be most helpful to explicitly state the major sources of error in risk estimates. |
| 2-13 | 22-30 | These statements are very general. At a minium specific examples need to be supplied that show actual examples of these errors and importance to the assessment. |
| 2-14 | 2-3 | It is anticipated that these discussions will provide sufficient detail on these sources of error in risk assessment. |
| 2-14-15 | | References should also include citations to the actual literature and not rely solely on EPA documents. |
| 3-1 | 10 | Change to "The deposition *results*." |
| 3-1 | 26 | Change to "The total amount of $NO_x$ emitted in the USA in 2002". |
| 3-2 | 3-4 | Change to "primary emitters of $NO_x$, mainly as NO and $NO_2$." |
| 3-2 | 5 | Change to "utilities, with *additional* contributions." |
| 3-2 | 9 | Change to "remainder of anthropogenic emissions." |
| 3-2 | 10 | Delete "component." |
| 3-2 | 15-16 | Delete "in the pie charts in." |
| 3-2 | 16-17 | Change to "*Results are shown on both a national* basis (contiguous states only) as well as for the eastern." |
| 3-2 | 18 | Delete "For this display." |

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| 3-3 | 1-2 | Delete "Note that emissions from Alaska and Hawaii are not included in any of these charts." |
| 3-3 | 5-6 | (not shown) with the on-road sector *being* the largest contributor, followed by emissions." |
| 3-3 | 8 | Change "overall" to "*national*." |
| 3-3 | 12 | The figure caption needs more information (e.g., that Hawaii and Alaska are excluded in these results). |
| 3-3 | 15 | Change to "*contiguous* states." |
| 3-4 | 3 | Change to "emissions *from* fertilized." |
| 3-4 | 5 | Clarify what is meant by "30%–70%." Does up to 70% of the waste mass be converted to emitted $NH_3$? |
| 3-4 | 9 | Change to "impact from *the input of* total reactive." |
| 3-4 | 10 | Explicitly state what is meant by "This." |
| 3-4 | 12 | Change to "Total *USA* emissions of." |
| 3-5 | 9 | Change to "utilities using *coa*l." |
| 3-5 | 12 | Change to "up to *10 ppm*." |
| 3-5 | 13 | This becomes a little confusing since the figure does not provide emission data from Hawaii and Alaska. Why not include values for these states in these figures. |
| 3-5 | 13-14 | Sulfur is found in other forms besides amino acids in vegetation so change to "Sulfur is a macronutrient (typically being 1 to 2%) and is released as $SO_2$ if vegetation is combusted (Levine and Pinto, 1998)." Also, there can be other forms of gaseous releases of sulfur components from vegetation (as well as soil) including $H_2S$, COS, methyl mercaptan, etc. I am not sure where in the document these biogenic sulfur sources should be included. |
| 3-6 | 1-2 | Delete since information has been included in my suggested modification. |

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| 3-6 | 3-6 | Change to "The *proportion* of $SO_2$ emissions from major source*s* are shown in Figure 3.1-5 both on a national total basis well as for the eastern and western United States." |
| 3-6 | 6 | Why not include Alaska and Hawaii for completeness? |
| 3-6 | 14-16 | The relative importance of this error needs to be stated explicitly. |
| 3-6 | 2-5 | Not sure the term "policy-relevant" is a useful term with respect to defining background concentrations. What is the basis of the actual value being used? Is it based on a specific time period or some calculation that attempts to separate out natural versus anthropogenic emissions? With the general effects of N fertilizers and N atmospheric deposition how can these factors in affecting $NO_2$ flux be evaluated? Is this 100 ppt based on some type of area weighted basis or some other form of calculation? |
| 3-7 | 4 | Delete "policy-relevant?" |
| 3-7 | 25 | Change to "This source distinction and quantification will." |
| 3-8 | 13-14 | Further information should be provided on the source and methods for estimating these deposition values. This information is provided later on page 3-13. This information should be provided earlier in the document. |
| 3-8 | 19 | Not sure what "Great Waters" means. |
| 3-8 | 19-23 | References are needed here. |
| 3-9 | | Figure 3.1-6. Reduce the significant digits in the figure legend to nearest 1/10th. |
| 3-10 | | Figure 3.1-7. Reduce the significant digits in the figure legend to nearest 1/10th. |
| 3-11 | | Figure 3.1-8. Reduce the significant digits in the figure legend to nearest 1/10th. |
| 3-11 | 9-10 | Further details on how these deposition estimates were derived should be provided. This information is provided later on page 3-13. This information should be provided earlier in the document. |

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| 3-12 | Figure 3.1-9. Reduce the significant digits in the figure legend to nearest 1/10th. |
| 3-12-13 | See my previous comments on the placement of this information within the document. |
| 3-12-16 | Isn't much of this information relevant to the overall predictions of N and S deposition? It was not clear why this more extended discussion was focused on the case studies. Was there a different type of analysis done for deposition for the case study regions than for the overall U.S.? |
| 3-16 | Aren't all these abbreviations already provided and hence Table 3.2-1 can be deleted. |
| 3-16 | Are these formulas included to show the conversions from formula compound mass to nitrogen or sulfur mass? If this is there only use then Table 3.2-2 should be deleted. |
| 3-17 11 | Has the western case study been identified? |
| 3-20 | Within each of these regions there have been other measurements made of associated parameters including precipitation amount, atmospheric deposition. Is there any attempt being made in this document to compare these current results with previously published results? |
| 3-23 | There are well established geographic patterns in N and S deposition across the Adirondacks. Shouldn't these be acknowledged? There has also been substantial work on deposition estimates in the Adirondacks using other approaches. For wet only deposition for example see: Ito, M., M.J. Mitchell and C.T. Driscoll. 2002. Spatial patterns of precipitation quantity and chemistry and air temperature in the Adirondack Region of New York. Atmospheric Environment 36:1051-1062. Also, other studies have evaluated the relative contribution of wet and dry deposition. See for example: Mitchell, M.J., C.T. Driscoll, J. Owen, D. Schaefer, R. Michener, and D.J. Raynal. 2001 Nitrogen biogeochemistry of three hardwood forest ecosystems in the Adirondack Mountains. Biogeochemistry 56: 93-133. Would some comparisons with other investigations be warranted? |
| 3-25 | This repeats a previous comment with respect to the inclusion of |

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| | | previous analyses of these regions in these cases studies. |
| 3-26-53 | | There is considerable detailed treatment of the results provided by the CMAQ modeling including temporal and spatial results. With so much emphasis on these model results it would be useful to provide some other confirmation of these deposition values. |
| 3-54 | 14-15 | This is not entirely true. There can be instances were ambient concentrations of $NO_x$ and $SO_x$ are directly deleterious. These conditions, however, have not been generally found in the USA during the period of concern associated with acidic deposition effects. For Europe conditions in the "black-triangle" and other centers of high pollutant concentration there were direct impacts especially associated with $SO_x$. |
| 3-55-70 | | The format and output of the CMAQ modeling runs are provided in some detail. Some further comparison of the CMAQ results, especially, in the areas of test cases, would be helpful in providing some objective measurements of the model results. |
| 3-70-72 | | In evaluating the relationships between $NO_x$ emissions and deposition, including total reduced deposition. The model results need to be verified with actual data that show these relationships. Clearly the model functional relationships and parameterization will affect overall model output. It is important that it be clearly identified when model outputs are producing results that have been verified elsewhere or whether there are currently no empirical and experimental results that provide data to verify these results. Such arguments can also be made for the other N gaseous constituents. |
| 3-70 | 16 | It is suggested that there is "statistical imprecision in the modeling." There are other important sources of model error that need to be considered. |
| 3-82 | 7-9 | Statements such as "Figure 3.2-55 shows that $NO_x$ emissions account for almost all oxidized nitrogen deposition in the Adirondacks Case Study Area, while Figure 3.2-56 shows that NH3 emissions account for almost all reduced nitrogen deposition" have important implications with respect to making recommendations associated with $NO_x$ emission controls. Are these results verifiable and can the emissions also be linked to specific geographical areas? |

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| 3-87 | How are these high emission locations ascertained? Is this a function of the location of specific monitoring locations (e.g., CASTNET sites). |
| 3-112 | This section on uncertainty (3.2.2.5) that has yet to be completed needs to include not only statistical issues, but also uncertainties in the model formulations and associated parameters. |
| 3-113-114 | This is a relatively limited reference list with considerable reliance on EPA documents. This section would be strengthened by inclusion of results from the peer reviewed scientific literature that support the suggested findings. |
| 4-1 | |
| 5-1 | The sections on "ACIDIFICATION" and "NUTRIENT ENRICHMENT" include only outlines, but the suggestion of the importance of providing uniform terminology is encouraging. |
| 6-1-10 | This section gives a useful review of recent findings linking the cycling of S and Hg. This section also includes more references to the referred literature related to this topic |
| 6-10-13 | The section on NITROUS OXIDE provides additional information on this N gas and provides a clear indication of the importance of this gas including its linkage with N deposition and is important role as a "greenhouse" gas. |
| 6-13-18 | The synopsis on CARBON SEQUESTRATION provides a useful overview of important interactions between N deposition, warming and the carbon cycling in terrestrial ecosystems. This is a very large and important subject area. As suggested a detailed analysis is beyond the scope of this review. The term carbon sequestration does not really capture the total content of this section. |
| 6-19-22 | More emphasis in the description of aquatic effects should be placed on changes in the phytoplankton community structure that has been most noted in the western U.S. |
| 7-1 | |
| 7-15 | In its current version this "Synthesis and Integration of Case Study Results" is highly descriptive. This will need to be improved with specific quantitative results that show some clear relationships to the assessment. |

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| 8-1<br>8-30 | | Clearly providing a clear scientific linkage that is policy relearnt between concentrations of $SO_x$ and $NO_x$ and resultant S and N deposition and subsequent translation to ecosystem level effects is a major challenge. Clearly there are good linkages between concentrations of $SO_x$ and $NO_x$ and deposition, but the linkage to ecosystem level effects is more complicated due to inherent variability across the United States of the sensitivity of these systems to acidification, eutrophication and changes in biotic composition. It will be important to formulate an approach that takes into account these geographical patterns of sensitivity. |
| 8-4-5 | | Figure 8.1-1 needs to indicate in some format that these ecological indicators may not be constant over the United States (e.g., N deposition levels with respect to changes in the biotic community structure being more sensitive in the Mountain west than the eastern U.S.). These issues are outlined on lines 1-7, page 8-5. |
| 8-5 | 22 | The problem in not including reduced N chemical species in any formulations associated with N deposition effects is highly problematic. I am not sure it is feasible to focus solely on $_{NOx}$. |
| 8-9 | | There are major difficulties in setting national standards for $SO_x$ and $NO_x$ concentrations without taking into account regional effects. |
| 8-13 | | As suggested the development of these "functional forms" will be critical and may have both spatial as well as temporal components. |
| 8-13 | | Clearly EPA is making heaving reliance on the CMAQ model suggesting the importance of validating results using other approaches and the linkages between gaseous concentrations and specific deposition levels. |
| 8-14-17 | | The issues of deposition velocities are not trivial. For example look at the differences in deposition associated with the estimates at the same site for CAPMoN and CASTNET. Although CAPMoN and CASTNET provide similar concentrations of gases, the deposition velocities are higher for CAPMoN versus CASTNET. |
| 8-23 | | The discussion of uncertainties needs to include the various issues associated with the calculation of deposition velocities. |

Attachments

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| 2, pg 2 5 | | Change "to reflect *most recent* conditions." |
| 2, pg 2 14-15 | | Not sure that there is a relationship between steepness and base cation leaching rates versus the role of slope in affecting the contact of drainage waters to soils and the relative contributions of ground waters to drainage with groundwaters generally being more rich in base cations than waters derived from shallower soil sources. |
| 2, pg 3 | 30 | Isn't a threshold of 400 eq/L or less considered acid sensitive? Would a threshold of 100 eq/L better? Also, note that the correct symbol for liter is "L" not "l." Change for entire document. |
| 2, pg. 5 | | Figure 3.2-1. Give a citation for the Acid Sensitive Waters (USGS). |
| 2, pg. 7 | | Why no selection of a threshold for sulfate deposition? Is this awaiting the determination of a critical load? At a minium the European critical load could be used. |
| 2, pg 8 | | Same comment for total N and S deposition and the assignment of a value as that provided by sulfate. |
| 2, pg. 9 | | The inclusion of high elevation could also be justified by other criteria such as the potential contribution of occult (fog) deposition. Is occult deposition accounted for in the CMAQ estimates? This also relates to rationale provided with respect topographic position (Attachment 2, page 17). |
| 2, pg 9 | 25-28 | This is confusing with respect S and N deposition since it was suggested above that no criteria for S and N deposition were defined. If these top quartiles are used, the actual values need to be supplied that make up these quartiles. Also, the year or years from which these deposition data are derived needs to be given explicitly. |
| 2,pg 10 | 14 | Why "Total nitrogen (Kjeldahl)." Do you mean total dissolved nitrogen or total dissolved Kjeldahl nitrogen? Why exclude nitrate in surface waters? |

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| 2,pg 11 | | Although there are weak linkages between N deposition and N solute concentrations in surface waters, it would be preferable to provide the data layer of actual N solute concentrations in surface waters. |
| 2, pg12 | 18-19 | Provide these nutrient concentrations available from the National Nutrient Database. Provide a citation for this and the other data bases. |
| 2, pg 15 | | These N deposition data layers seem to be redundant from data layers previously described. |
| 2, pg 16 | | More clearly delineate the difference between "content" and "concentration." Content should be reserved for the total amount (mass, molar value) of an element on a per unit area while concentration is the amount (mass, molar value) of an element per unit of mass (sometimes) expressed as %. |
| 2,pg 18 | | The delineation of the location of acidophilic lichens appears to have a strong boundary based upon state borders especially notable in Arizona and an absence in New Mexico. |
| 3, pg 10 | | Here and elsewhere "  M" is not a correct abbreviation. The abbreviation for mole is "mol." |
| 3 pg 12 | | Note that these ANC levels are lower than those suggested to be considered to be of concern in Attachment 2. |
| 3, pg 15 | | There have been other models (e.g., PnET-BGC) that have been applied particularly to the Adirondacks to evaluate especially temporal patterns of acidification. Wouldn't it be helpful to at least do some comparisons using other models besides MAGIC? |
| 3,pg 18 | 8 | Change to "$SO_4^{2-}$." |
| 3 pg 21 | | Clarify how the ratios of wet to dry deposition were derived. |
| 3, pg 22 15 | | Are the total deposition values used in the MAGIC calculations different from deposition estimates used elsewhere in the entire document? |
| 3,pg 22 | 27-29 | Some elaboration on how ASTRAP derived deposition would be helpful and how these estimates confirm or differ with the CMAQ |

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| | estimates. |
| 3, pg 23-26 | There is considerable discussion of the calibration of MAGIC in this section. Some of this could be reduced and inclusion of other ways of comparing model output with other published results including other models (e.g., PnET-BGC) would provide additional perspectives of these results. |
| 3, pg 29 | Similar to previous comments, although the discussion and definition of the F-factor provides background information this is not a novel approach, but rather one that could be cited. More emphasis of the validation and comparisons with actual measurements versus description of the model development would be more helpful. |
| 3, pg 31 | Figure 5.1-1. Trends in LTM monitored lakes in the Adirondacks of New York would be improved if specific lake classes were used. It is not clear from these figures how a general trend for the LTM monitored lakes were obtained. |
| 3, pg 32 | Figure 5.1-2. The modeled values appear to be substantially different that those of the measured values. Doesn't this bring into question to validity of the predictions for 2010? I don't believe the results of this figure are discussed within the document. |
| 3, pg 35 | Clearly the document is incomplete at this stage. |
| 4, pg 1-2 | This seems repetitious of information provided in the main document. |
| 4, pg 5 | "M' is not the correct SI abbreviation for mole. The correct abbreviation is "mol." |
| 4, pg 4-9 | Not sure that this much restating of what has previously been found about Al and Ca relationships is needed. |
| 4, pg 11 | Would it be more efficient to cross reference the description of ecosystem services as provided in the main document? |
| 4, pg 18-20 | Are the details of the history of the Kane Forest needed in this document? |
| 4, pg 21-25 | Similarly to the previous comment, are all these details about |

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| | Hubbard Brook needed for this document? |
| 4, pg 26-47 | This is difficult to follow due to the level of detail. Perhaps a more generalized format in which could be imbedded specific references to the literature or the inclusion of appendices so that the major theme of the discussion is not lost. |
| 4, pg 50 | Not sure of the importance of the inclusion of the section on "Implications for other systems" in this document. |
| 4, pg 51-52 | The uncertainties issues need to be better integrated into the overall document. |
| 4, pg 53 | The "Conclusions" summarize the important issues and these issues should be the focal points of this entire section with a need |
| 6, p 43 | The figures on this page (Figures 5.1-1 5.1-2) appears to have an inappropriate numbers. Shouldn't they be 6.1-1 and 6.1-2? |