Preliminary Comments from Dr. Kathleen Weathers on EPA's Integrated Science Assessment for Oxides of Nitrogen, Oxides of Sulfur, and Particulate Matter – Ecological Criteria (First External Review Draft)

Kathleen Weathers' Preliminary Comments:
Chapter 2:

General comments:

Much of this chapter gets bogged down in details: there is too much detail for some topics and far too little context throughout. What's the big picture? How do these details inform? Why do we care? With what kind of certainty (or uncertainty) do we know? What new data and syntheses have been published since the last ISA and how does this change what we know about sources, chemistry, measurement and modeling of concentration and deposition, as well as uncertainty?

I suggest describing the big picture first, with summary figures and tables, and then assessing the level of detail necessary for each aspect of what we know about sources, chemical transformations (of relevance), measurement and modeling.

While I appreciate geospatial data displays, without additional information on uncertainty or some site-specific data showing temporal trends, they can be misleading (or leave a lot of interpretation to the reader). There are many cases where, in addition to maps, comparative tables or figures are necessary.

I was surprised at the frequent underscoring of the need to consider bi-directionality. It was hard for me to tell when, considering an annual critical load, for example, consideration of bidirectionality for any particular chemical species might truly be important vs when it would make little difference to the net flux to an ecosystem calculation.

The bottom line is that clarity, level of detail, and relevance, in particular need work.

Some detailed comments:

Section 2.1: I suggest adding a table of at least N species including which are considered NOy, NOx, etc.
The language is awkward throughout.

Section 2.2. The language is awkward in this section, too.
Section 2.3.1: I'm not sure that the level of detail in this section is necessary. It seems to

me that it's too much, but I will defer to my atmospheric chemist colleagues. Also, what has been learned that is new and relevant since the last ISA?

Page 2-14. Wouldn't wind be a factor here?

Section 2.3.2: Page 2-15, lines 19+: make sure not to conflate concentration and flux here.

Section 2.3.3: I agree completely with the first sentence (more complete description of the composition of rainwater). I suggest that you add a figure or a table that shows, for a handful of sites, the top 3-4 (in equivalents) anions and cations in rainwater for some characteristic regions of the US, just as an illustration of what is predominant (and what is not—organic acids likely not being predominant).

Figure 2-3: Again, I will defer to my atmospheric chemist colleagues about its inclusion and importance; it's not immediately clear to me what the main point of this figure is (vis-à-vis this ISA).

Section 2.5: CASTNET consists of 261 sites? Is this number correct?
I suggest summarizing the details of the networks that make measurements relevant for deposition in a table and including here.

Section 2.5.1: when was the FRM accepted (i.e. is this something new)? If data from EMEP are not being used here, why do we need to know the details?

Primary references for descriptions of the CMAQ, GEOS-chem etc. models should be included when they are first introduced.

Page 2-25: odd N species?
Section 2.5.4: Need a summary statement at the end of this section, and all sections. What's the take home. For this section is it that the data are not very comparable?

Section 2.5.6: How is the CSN similar or different than CASTNET?
How high are extraction efficiencies? Is there a difference between the two coatings used in IMPROVE or CSN? A table showing similarities and differences between methods and the extent to which they've been compared, results of those comparisons (even qualitative results).

Section 2.6. Should be clarified. The figure legend should be written to reflect the map. To my understanding this is not really a distribution—the data are modeled concentrations based on CMAQ. Give spatial resolution. The names of some of the urban centers on map would be helpful.

I suggest you add the chemical notation in the headers along with the names

Section 2.6.1, lines13-15: "not much confidence.." how about there is such significant uncertainty. And, "due to limitations in the satellite retrievals" …for what?

Section 2.6.4: Highest annual? What time period?

Section 2.6.5: Add at the end of the paragraph: "And we interpret this to mean…"

Sections 2.6.6 and 2.6.7. Include a summary graphic or table of the temporal trend changes in particulate sulfate.

I would be curious to see regressions of the SO4 and SO2 concentration data for CASTNET sites.

Section 2.7: Here are additional references that may be useful for clarifying this section.

Weathers, K.C., G.E. Likens, and T.J. Butler. 2007. Acid rain. Pp. 1507-1522. *In:* W. Rom (ed.). Environmental and Occupational Medicine, 4th edition. Lippincott-Raven and Wilkins Publishers, Philadelphia*;*

Weathers, K.C., and A. Ponette-González. 2011. Atmospheric Deposition. *In:* D.F. Levia, D.E. Carlyle-Moses, and T. Tanaka (eds.). Forest Hydrology and Biogeochemistry: Synthesis of Past Research and Future Directions. Ecological Studies Series, No. 216, Springer-Verlag, Heidelberg, Germany.

Weathers, K.C., S.M. Simkin*, G.M. Lovett, and S.E. Lindberg. 2006. Empirical modeling of atmospheric deposition in mountainous landscapes. *Ecological Applications* 16:1590-1607.

Weathers, K.C. 1999. The importance of cloud and fog to the maintenance of ecosystems. *Trends in Evolution and Ecology* 14:214-215.

Kimball, K.D., R. Jagels, G. Gordon, K.C. Weathers, and J. Carlisle. 1988. New England coastal fog and mountain cloud water chemistry. *Water, Air and Soil Pollution* 39:383-393.

Weathers, K.C., G.E. Likens, F.H. Bormann, S.H. Bicknell, B.T. Bormann, B.C. Daube, Jr., J.S. Eaton, J.N. Galloway, W.C. Keene, K.D. Kimball, W.H. McDowell*, T.G. Siccama, D. Smiley, and R. Tarrant. 1988. Cloud water chemistry from ten sites in North America. *Environmental Science and Technology* 22:1018-1026.

Figure 2-17 shows scavenging processes but does not succeed in showing important mechanisms for transferring atmosphere to the surface (vis-à-vis deposition).

Section 2.7.2: This section is significantly lost in the details. How is dry deposition currently estimated, what are the uncertainties, and what advances have been made since the last ISA?

2.7.2: Inferential methods should be invoked in the first paragraph.

Page 2-51: Absent is any discussion of scale differences. For example, name the grid cell size in line 21. Further the significance of the paragraph from line 7-21 should be made clear.

As noted in the phrase on line 24, it is the net flux that is of interest for mass balance. How much does bidirectional exchange of gases matter? I cannot tell beyond the claim that "serious errors" can result.

I am confused by Table 2-5: wet deposition velocities for SO2?

Page 2-56 and top of page 2-57. This is the kind of (useful, semi-big picture) information that needs to be summarized and compared in tables, for example. What are the most sensitive variables in dry deposition models, and how do they compare? While there is no 'known' to which to compare (i.e., because it is not possible to measure dry deposition directly, we don't have a true answer), comparison across model results is useful in honing estimates and begin to unravel what drives the differences. While I agree that the estimation of dry deposition remains uncertain, what is certain is that in some locations, dry deposition can be a large component of total deposition, and must be considered as part of the mass balance. Uncertainties are likely to be large now, but will get smaller over time.

2.7.3: It is important to note that for many montane regions in northeastern north America, while deposition can be quite high where cloud deposition occurs, the spatial extent of regions of high deposition as a result of cloud or fog can be quite low.

Once again, the end of this section seems to trail off into the weeds; I'm not sure what the point is.

2.7.4: Start this section with why we want to know about throughfall vis-à-vis deposition. Where, how, and why is it useful and where is it not. What's new?

Figure 2-20: Can arrow size be adjusted to importance/magnitude of pathways relative to overall flux?

2.8: The Schwede and Lear method of estimating deposition to the US is one of the biggest advances using network-based data since the last ISA. It is not a panacea, but it is a major advance. What have been the major advances since the last ISA with CMAQ, and GEOS-chem, or other modeling efforts that estimate deposition?

Sentences such as "Note, however that he sampling artifacts mentioned in Section 2.5 should be considered," should be moved to a different, "uncertainties," section and then described (briefly!)? What should be considered and how might it affect the model output?
Lines 26 and 27: achieve greater consistency…with what? What's the problem that is being solved with wet deposition? How do NADP data fit into this?
Lines 32-33: Wasn't a comparison of CMAQ and monitoring data part of the last ISA? And isn't it regularly done as improvements are made to the CMAQ model? If not, these things should be done.

Lines 20-22: Shown but not compared with other estimates?!

Figure legends for 2-22 and 2-23 should remind what's included in reduced and oxidized N in these maps.

2.8.2: has there been a corresponding decrease in precipitation in the areas where dry deposition is increasing?

2-70, line 14: "increased markedly." Here's another case where having some quantitative data—a table showing actual and percent increases for selected locations or regions is desirable.

2-71, top of page. Absolutely, but they to be identified and even if a qualitative indicator is used, they need to be (qualitatively) quantified.

2-71 line 9: improvement in the acidity of rainwater.

2-72 lines 5-7: ????

2-72 lines 8-18: Why not compare changes in NADP site data rather than interpolate and reinterpolate, with a high degree of uncertainty, maps?

2-75: Please spell out the logic and/or equations for calculating acid loading/expressed as hydrogen ion equivalents for S and N wet dep.

2.8.4: How do these estimates compare? And, !! lines 16-19. So, what might that mean? What's the difference between older algorithms and new ones, even qualitatively? I agree that the fact that maps can be produced is promising, BUT, where are the comparisons? Kudos for producing an uncertainty map, but on what are those uncertainties based? Under what circumstances should satellite or GEOS-Chem models be used vs total deposition estimates based on monitoring data (I now see that some of this is addressed, somewhat indirectly, in section 2.9).

2-80 line 22: "could be given?"

2-80 line 28: dry deposition is modeled, not measured, at CASTNET sites.

2-81 lines 28-31: Yes!

Figure 2-34. I assume it is the 1:1 line that is shown. It should be noted.

2-83: line 12: status of N limitation (to ocean?)

Figure 2-35 legend should read: Estimated or modeled contributions to... based on GEOS-chem.

The Summary is very good (the absence of uncertainty summary notwithstanding). In fact, if the rest of the chapter could be structured around the summary, this chapter could be very much improved. Another possibility is to put the summary as the first section of this chapter.

Chapter 3:

Chapter 3 characterizes scientific evidence on the effects of gas-phase SO_2, NO_2, NO, peroxyacetyl nitrate (PAN), and HNO_3 on vegetation. Please comment on the characterization of these effects and the integration of new information into the long history of evidence on this topic.

EPA has put together a generally informative introduction. However, throughout the introduction summaries from past ISAs should be pointed out as distinct from new information. One way to do this is to use subheaders, for example "new information since the last review" (e.g., beginning with line 30, page 3-5).

There is little that is new—or that has been new for many years, so there is not much to say about this brief chapter. I am curious about what EPA thinks about the kinds of data/studies that would be useful to have conducted before the next ISA? Would they would include whether and how ecosystem productivity is affected by these gases, or what interacting effects of gases and other pollutants and nutrients are on vegetation, or long-term effects of low concentrations of gases on plant physiology? A few sentences on what the gaps in knowledge are would be helpful. There are a few interspersed in the text (e.g., page 3-13, lines 25-27)

I did not notice that Geiser and Neitlich 2007 was cited.

Appendices

The Appendices are dense and, taken alone (which is not the intent), not very clear.

Appendix A should include maps as well as figures showing changes. I'm a fan of geographical representations, but temporal trends, for example, are very hard to discern. It would be useful to include 3 (ish) main bottom lines for each of the figures. What is it that they are intended to illustrate?

Appendix B:

Appendices B and D are important to include as they recognize complex interactions among gases, particles, and interacting element cycles.

Please consider using subheaders.

Say what the ELA is the first time it is referred to rather than in section B.4.1.

Appendix C:

I suggest standardizing the initial information given for each of the case study ecosystems: e.g., study area, slope and elevation gain, climate, distance from the coast, percent forest cover by type (northern forest, etc.). They are quite uneven now.

How near to Class I areas is Hubbard Brook?

Both ACAD and Hubbard Brook are included in this relevant paper using the DayCent-chem model to examine the effects of N dep as well as warming on ecosystem functions:

Hartmann, M.D., J.S. Baron, H.A. Ewing, and K.C. Weathers. 2014. Combined effects of warming and atmospheric nitrogen deposition on net ecosystem production, greenhouse gas flux and water quality in nine US mountain ecosystems. *Biogeochemistry*. 119:85–108.

I don't understand the reliability ratings in Table C-7. How is a peer reviewed paper not expert judgment?