Compilation of Comments from Members of the CASAC Secondary NAAQS Review Panel for Oxides of Nitrogen and Sulfur on the Panel’s Draft (8-4-17) Report

(As of 8-25-17)

The following comments from Panel members are grouped according to each section of the Panel’s draft (8-4-17) report. The compilation contains comments received from Panel members as of 8-25-17.
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General Comments

Edith Allen -- The CASAC NOx-SOx Review Draft Report of 8-4-17 is a clear synthesis of many diverse Panel comments and recommendations. I have a few editorial comments related to the Chapter Summaries in the cover letter, and for the Consensus Response to Charge Questions for Chapters 1, 3, 6, 14 and Appendix C.

Praveen Amar - I do not think CASAC panel should recommend a stand-alone NAAQS for ammonia during the REA and PA phase, as we currently do, but should recommend a NAAQS for total reactive nitrogen (page 2 of draft letter, comments on Chapter 3).

Laurie Chestnut -- I have reviewed the draft report and it all looks okay to me.

Ivan Fernandez -- The panel has made clear by general comments, highlights, and other comments, a priority of issues. However, to further highlight the importance of material included in the sections titled “key findings and recommendations,” it may be useful to move the text in the sections titled “other comments” into an appendix. The appendix could be titled “Other Comments and Technical Corrections.”

Frank Gilliam – This draft report reads quite clearly, and so I would have little to add. Although I have looked over the entire document, I have read most closely the chapters I was directly involved with during the evaluation process. This document appears to represent our collective deliberations in Durham, NC, extremely well. Authors are to be commended for the assembly of the document and the conciseness and clarity of the summary remarks.

Hans Paerl – I have read through the entire document and find it very well prepared, comprehensive and very useful for the preparation of the final document. Only one minor change I'd like to see, on p. 25 line 20 change “limitation” to “co-limitation”.

Introductory Text

p. 1 lines 24-25 (James Galloway) I suggest nitrate be mentioned in lines 24-26, just as sulfate is mentioned on lines 26-27. Note that the charge on sulfate is shown, but throughout most of the document it is not shown

Executive Summary and Chapter 1 (Response to Charge Question 1)

p. 2 line 10 (Ivan Fernandez) After the word “lengthy” insert “(1,412 pages).”

p. 2 line 19 (Ivan Fernandez) Insert “in the scientific literature” after “uncertainties.”
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p. 2 line 20 (Ivan Fernandez) At the end of the sentence add “…and concepts (e.g., recovery).”

p. 2 line 25 (Daven Henze) I wonder if it would be worthwhile to state more explicitly which aspects of specific models we were thinking of here; I recall modeling source apportionment and treatment of NH3, in particular; there are likely others.

p. 2 line 28 (Praveen Amar) Revise the sentence as follows: “The CASAC also recommends that the EPA clearly identify science topics that are not thoroughly reviewed in the ISA because they are to be addressed in the REA and the policy-oriented phases of the review. The EPA should also explain why these topics are to be best addressed in the policy phase of the review.”

p. 2 line 37 (Ivan Fernandez) Change the word “rest” to “length.”

p. 2 line 39 (Praveen Amar) Change “5” to “five.”

p. 3 line 11 (Praveen Amar) Revise the sentence as follows: “A presentation of the NAAQS associated with this review, the requirements/components of the NAAQS (form, indicator, level, and averaging time), and a discussion of how these requirements/components may impact the formulation of a standard should be included in the introduction.”

p. 3 line 25 (Praveen Amar) In the following text I think the first three terms are described well, but the last term “surprises” is not explained. Maybe, give an example, perhaps? “The following two new sections should be included near the beginning of Chapter 1: (1) a section on uncertainties, and (2) a section on “Concepts, Connections, Changes, and Surprises.” This second section should provide readers with a clearer view of: what is to be presented (e.g., important concepts such as deposition, critical load, and biodiversity); major conceptual changes in the science since the last review (i.e., Changes); how different aspects of the Draft ISA fit together (i.e., Connections); and specific aspects of the review of the science that might be most informative (i.e., Changes, Concepts, and Surprises).”

p. 3 line 34. (Edith Allen) I am not sure what the following sentence means. Should it be explained here? “In this regard, it is important to: (1) differentiate between what may be called a “scientist’s critical load” and a “policy-maker’s critical load...”

p. 3 line 36 (Praveen Amar) Revise the sentence as follows: “In addition, the CASAC recommends that the discussion of critical load indicate that critical loads may vary in time due to past cumulative deposition.”

p. 3 line 42 (Praveen Amar) Revise the sentence as follows: “In particular, this would apply to the expanded scientific assessment of the approaches used in the prior review and the section on “Concepts, Connections, Changes and Surprises.”
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p. 3 line 44 (Ivan Fernandez) Should be “Concepts, Connections, Changes, and Surprises.”

Chapter 2 (Response to Charge Question 2)

p. 4 line 30 (Praveen Amar) Revise the sentence as follows: “Structural uncertainty (e.g., arising from the use of transference ratios) should be distinguished from quantitative uncertainty (e.g., associated with measurement of concentrations and deposition or with estimates of emission rates).”

p. 5 line 22 (Praveen Amar) Revise the sentence as follows: “Further, wet deposition is also sparsely measured, and dry deposition is mostly modeled.”

p. 5 line 23 (Praveen Amar) Does the following sentence mean “attainment decision process” for SECONDARY STANDARDS? “These points should be taken into account when considering the potential use of models in the attainment decision process.”

p. 5 line 30 (Praveen Amar) The report states that “It is important to note the increasing role of transportation as a source of NH3. The role of soil NOx should also be discussed. It is not clear whether this is included in biogenic emissions or agriculture.” I think it is our understanding of the role of transportation NH3 emissions is increasing. In any case, we must note that the fraction of ammonia emissions from agriculture sector including animal CAFÉ sources is much larger than transportation sources ammonia.

p. 6 line 1 (James Galloway) ‘rainwater’ should be changed to ‘precipitation’.

p. 6 lines 3-4 (James Galloway) The word ‘notes’ is not very strong. Can there be an additional sentence that recommends a more stable funding base?

p. 6 lines 5-7 (Mark Fenn) In summary here, the report should point out the limitations of the current monitoring networks for effects evaluation—a point that seems to often be overlooked. Notwithstanding the accomplishments of the NADP/NTN, particularly successes in documenting long term directional trends in deposition (but not trends in total deposition), both CASTNET and the wet deposition network (NTN) are quite limited in their ability “to support the current review of ecological effects”. CASTNET has been shown to poorly model actual dry deposition inputs, often with large underestimates of dry deposition when compared to empirical data. And because wet deposition fluxes are often many-fold lower than total deposition, dry deposition is often the largest component of total deposition, especially in more polluted sites. Thus, where dry deposition is elevated, sometimes NADP deposition maps present such sites as moderate or even with very low deposition—which is highly misleading and limits the usefulness of NADP/NTN data for effects evaluations. And of course this is ignoring the need for more spatially intensive monitoring, which is unlikely to improve because of funding limitations.
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p. 6 line 13 (James Galloway) the term ‘reactive N’ is used a few times in the document. In the context of this report it does not include nitrous oxide, but in most N-related literature, it does include nitrous oxide. Therefore, I recommend that ‘reactive N’ be defined.

p. 6 line 43 (James Galloway) the citation should be Galloway et al. (1982).

Chapter 3 (Response to Charge Question 3)

p. 7 lines 26-27 (Edith Allen) The report states that “In Chapter 3, the EPA should note that there is a need for research on NH₃ fumigation effects on plants to fill a key knowledge gap.” I agree with this statement. However, there are a few citations (below) from Europe on NH₃ fumigation that can be cited in Chapter 3.

Cape et al. recommend a CL of 1 ug/m³ NH₃ for lichens and 3 ug/m³ NH₃ for higher plants.


Sheppard et al. observed growth decline of a heathland shrub (Calluna vulgaris) and death of a lichen (Cladonia) at ammonia critical levels of 8 ug/m³ NH₃:


Chapter 4 (Response to Charge Question 4)

p. 8 line 16 (Ivan Fernandez) Change ‘time” to “time(s)”

Chapter 5 (Response to Charge Question 5)

p. 11 lines 7-12 (Mark Fenn) The comments in this paragraph provide a critique of using soil Ca:Al ratio as an indicator of vegetation effects as described by Cronan and Grigal (1995) and others, but it does so citing a 2007 review paper on Ca:Al ratios in roots, assuming that Ca:Al ratio in soil and fine roots are equivalent indicators. Seems that the jump to criticized soil Ca:Al
ratios based on root studies might be an apples and oranges comparison. Is further information
needed here?

p. 11 line 9 (James Galloway) The Vangelova paper is a decade old. Are there more recent
papers that support this conclusion?

p. 11 lines 26-28 (Mark Fenn) Clarify why this study by Whitfield et al (2010) isn’t really
pertinent to include in the report.

p. 11 line 30 (James Galloway) whenever ‘kg/ha/yr’ is used, the chemical species should be
given (e.g., kgN/ha/yr).

Chapter 6 (Response to Charge Question 6)

p. 12 lines 11-15 (Edith Allen) The report states that “CASAC recommends that the following
references be cited to address the possible differential ecological effects of oxidized versus
reduced forms of N: Van den Berg et al. 2005; Kleijn et al. 2008; Stevens et al. 2011; Verhoeven
Some of these could also be cited in Chapter 3 to emphasize the importance of ammonia
deposition.

p. 14 lines 11-15 (Edith Allen) The report states that “The CASAC notes that in the table entry
on page 6-162 (Table 6-28) for “Mediterranean California,” the old N critical load values are for
NO3 leaching in streams from montane forested watersheds (17 kg/ha/yr) and tree mortality in
mixed conifer forests (39 kg/ha/yr), but the new critical load given is for coastal sage scrub
vegetation type. The old and new critical loads are for very different vegetation types that occur
in different elevations/habitats.” Pardo et al. 2011 cite 7.8 to 10 kg/ha/yr as low to high CL for
coastal sage scrub diversity loss. These values can be used here as well, and are consistent with
the more recently published CL for mycorrhizal diversity loss. The values of 17 to 39 are for
different vegetation types and for different ecosystem responses to elevated N. I suggest delete
the values 17 and 39, replace with 7.8 and 10 kg/ha/yr.

p. 15, line 6 (James Galloway) The EPA report and the CASAC assessment relies heavily on
published literature. Given this, citing ‘unpublished’ information is not a good idea, in my view.

p. 15 lines 36-40 (Edith Allen) The report states that “In the second entry in Table 6-28 on page
6-161 (coastal sage scrub), the correct values for the low and high critical loads for conversion to
exotic grasslands are 7.8 and 10 kg N/ha/yr (see Table 13.3 in Pardo et al. 2011). The current low
value of 6 kg N/ha/yr is actually for serpentine grasslands (see Table 13.4) and the current high
value of 33 kg N/ha/yr given in Table 6-28 is for biodiversity of forest understory in the San
Bernardino Mountains (see Table 13.5 in Pardo et al. 2011).” Same as comment above, the
values of 6 to 33 reported in Table 6-28 are for different vegetation types and different
ecosystem responses. I suggest delete the values 6 and 33, replace with 7.8 and 10 kg/ha/yr.
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Chapter 10 (Response to Charge Question 10)

p. 22 line 4 (James Galloway) Has ‘NHy’ been defined?

p. 22 line 29 (James Galloway) The word ‘the’ is misspelled.

p. 25, line 3 (James Galloway) The charge on nitrate should be a superscript.

P. 25 line 20 (Hans Paerl) change “limitation” to “co-limitation”.

Chapter 13 (Response to Charge Question 13)

General comment (Praveen Amar) – My major comment is on CASAC recommendation on Chapter 13 that addresses climate modification of ecosystem responses to nitrogen and sulfur deposition. As currently formulated (page 3, lines 44-45), it states, “However, the CASAC suggests that EPA consider whether the discussion of climate change effects should be integrated into other chapters of the Draft ISA OR organized at the end of the document as a separate chapter.” CASAC draft letter goes on to say that “by including a separate chapter on climate change at the end of the document, the EPA may convey the incorrect message that this is a minor issue to be considered in the NAAQS process.”

My sense (as a non-expert) is that the emerging literature on the effect of climate change on certain ecosystem effects (Chapters 3 to 12) is important enough and robust enough that we should recommend that EPA discuss such climate effects in the body of each of the effects chapters itself. We should also recommend that the current chapter 13 be retained with focus on integrating/synthesizing the climate effects included in all of the previous chapters.

p. 28 lines 9-10 (James Galloway) Of course, both could be done (that is including a separate chapter on climate change as well as and integrating the issue into chapters of the ISA.

p. 28 lines 17 - 18 (Erik Nelson) Do we want to specifically refer to the Climate Science Special Report (https://assets.documentcloud.org/documents/3914777/Third-draft-of-the-Climate-Science-Special-Report.pdf)? I know that a draft copy has been published.

Chapter 14 (Response to Charge Question 14)

p. 29 line 34 - 40 (James Boyd) The report reproduces a sentence the review team included in its Chapter 14 report. I would like to revisit and change/amend the sentence. The sentence is:

“For the purposes of the Draft ISA the authors could declare that they consider any ecological process that humans value or utilize, and is affected by NOx, SOx, or PM emissions and/or related deposition, as an ecosystem service of relevance.”
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Upon reflection, I find this statement to be a source of potential confusion. The problem is that “any ecological process that humans value or utilize” could be misinterpreted, since in principle any/all ecological processes can be viewed as undergirding human welfare. The reviewers want the authors to move away from broad, vague definitions and toward more specific definitions and quantification – as we make clear in the rest of the comments.

Proposed solution. Change the sentences (starting with the sentence quoted above) to read:

“For the purposes of the Draft ISA the authors could declare that they consider any ecological process that humans value or utilize, and is affected by NOx, SOx, or PM emissions and/or related deposition, as an ecosystem service of relevance. However, that is not our recommendation. We recommend that the report’s approach to ecosystem services focus on the outcomes of ecological processes (biophysical or social) – and, specifically, focus on outcomes that facilitate social and economic interpretation. A reference that makes that terminological point is Boyd and Banzhaf (2007).”

p. 30 lines 2-4 (Erik Nelson) Revise the sentence as follows: “Analysis that translates ambient and deposition outcomes into biological, and physical, or monetary metrics that give people insight into the public welfare impacts of ambient and deposition outcomes that facilitate social evaluation and public understanding of loads’ effects on public welfare.”

p. 31 lines 5-6 (Edith Allen) add the following citations from Ecosphere publications on societal ecosystem services:


p. 32 lines 17-23 (Erik Nelson) Is this recommendation redundant? Wouldn’t the table recommended here be part of the table recommended in lines 10 – 25 of page 31?
Appendices (Response to Charge Question 15)

p. 35 line 21 (Praveen Amar) Revise the sentence as follows: “There is a large amount of information included in the case studies.”


Letter to the Administrator

p. 1 line 31 (Daven Henze) Perhaps this is a bit obvious, but it might be worthwhile to state here that the content in these pages is just the main highlights, and that the detailed comments include topics that are not represented here in the highlights.

p. 1 line 35 (James Galloway) The word “also” is not needed.

p. 1 line 35 (Praveen Amar) The report states that “The CASAC also recommends that the following topics be discussed in Chapter 1: (1) key scientific uncertainties; (2) requirements for National Ambient Air Quality Standards (NAAQS)…” I think we mean Components of NAAQS (form, level, indicator, averaging time)

p. 2 line 1 (Praveen Amar) Revise the sentence as follows: “In addition, the CASAC recommends that: (1) the Chapter 2 summary (Section 2.11) be moved to the front of the chapter; (2) a comprehensive discussion of the major atmospheric transport and flux modeling platforms be included in Chapter 2; (3) the process used to obtain emissions estimates (including estimates of ammonia emissions) in Chapter 2 be better documented; (3) text be added to Chapter 2 to discuss the usefulness of data from various monitoring networks to estimate deposition; and (4) more definitive conclusions be presented about the usefulness of transference ratios for linking ambient air quality to total deposition.”

p. 2 line 10 (Praveen Amar) Revise the sentence as follows: “The CASAC notes that the ecological effects of reduced nitrogen compounds are important for understanding the effects of atmospheric total reactive nitrogen deposition and should be emphasized included in Chapter 3 and other chapters of the Draft ISA.”

p. 2 line 12 (Praveen Amar) The report states that “Ammonia emissions are increasing in many regions of the U.S. and the CASAC recommends that EPA consider the need for developing National Ambient Air Quality Standards for reduced forms of nitrogen during the Risk and
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Exposure Assessment and Policy Analysis phases of this NAAQS review.” Not sure if we want
to say that NAAQS for reduced forms of nitrogen need to be developed on stand-alone basis. I
think the focus should be the development of NAAQS or critical loads for total reactive nitrogen
and total sulfur deposition.

p. 2 lines 12-15 (Edith Allen) The draft letter states that “Ammonia emissions are increasing in
many regions of the U.S. and the CASAC recommends that EPA consider the need for
developing National Ambient Air Quality Standards for reduced forms of nitrogen during the
Risk and Exposure Assessment and Policy Analysis phases of this NAAQS review.” Can we
state this more strongly? For instance, wording such as: CLs are developed based on total N
deposition, but only half or less of the total N deposition in the U.S. is regulated based on the
NO2 standard. I realize the rationale for regulating ammonium is stated in the more detailed
Panel responses below, but it would be helpful to give a rationale here in the cover letter.

p. 2 line 17 (Praveen Amar) the report states that “Chapter 4 of the Draft ISA summarizes soil
biogeochemical responses to atmospheric sulfur and nitrogen deposition. The chapter is
generally complete with respect to the discussion of indicators, processes, models, monitoring,
and national sensitivity.” The reference to national sensitivity is not clear.

p. 2 line 20 (Edith Allen) The language in the report “(and other effects chapters in the Draft
ISA)” is not clear. Is there a word missing or incomplete phrase?

p. 2 line 23 (Praveen Amar) The report states that “It would also be useful to present maps in
Chapter 4 to show the relative contribution of sulfate, nitrate, and reduced nitrogen driving
acidification and eutrophication effects, particularly for areas of the U.S. where acidification and
nitrogen critical loads are exceeded.” Should we say nitrate or oxidized nitrogen?

p. 2 line 29 (Praveen Amar) The report states that “In addition, the CASAC finds that much of
the literature reviewed in Chapter 5 concerns natural variability in soil pH, calcium
concentration, and base saturation impacts on plant health and microbial composition (rather
than effects of nitrogen or sulfur deposition on acidification). These studies are useful for
understanding basic physiological principles of acidification effects on biota, but the Draft ISA
should clearly identify studies that are not based on elevated levels of nitrogen or sulfur
deposition.” Not clear what we are emphasizing.

p. 2 line 34 (Praveen Amar) The report states that “The CASAC also recommends that the
chapter discuss uncertainty in the base cations to aluminum ratio considered to be protective for
various species.” The last sentence needs to be written more clearly. The uncertainty of the B/Al
ratio is NOT considered to be protective, but the ratio is.

p. 2 line 42 (Praveen Amar) Should be “conclusions”
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p. 3 line 4 (Praveen Amar) The report states that “The CASAC also recommends that: (1) the section on dissolved organic carbon be rewritten to address specific concerns identified in the enclosed report, and (2) a framework be provided in Chapters 4, 5, 7, and 8 to define recovery.” Should we say “recovery of ecosystems” or “chemical and biological recovery of ecosystems”?

p. 3 line 9 (Praveen Amar) The report states that “The CASAC recommends that Section 8.4.6 of the chapter discuss the different mitigation responses of direct application of lime on water and whole watershed liming.” Not sure if this recommendation about studying the effects of adding lime on mitigation responses is important enough to be included here.

p. 3 line 23 (Praveen Amar) The report states that “The CASAC notes that reduced forms of atmospheric nitrogen are playing an increasingly important role in estuarine and coastal eutrophication, and in harmful algal bloom dynamics.” Do we mean to say that “reduced forms of atmospheric nitrogen are PLAYING….” Or should we say “that reduced forms of atmospheric nitrogen PLAY” an important role. The way it is written implies that reduced nitrogen forms are PLAYING an important role now, but they did not in the past.

p. 3 line 41 (Praveen Amar) Revise the sentence as follows: “The chapter should indicate that the changing character of atmospheric temperatures, precipitation and increasing carbon dioxide can modify processes discussed in the other chapters (Chapters 3 to 12) of the ISA.”

p. 3 line 44 (Praveen Amar) The report states “However, the CASAC suggests that the EPA consider whether the discussion of climate change effects should be integrated into the other chapters of the Draft ISA or organized at the end of the document as a separate chapter.” My suggestion is that EPA do both. Each effects chapter (Chapters 3 to 12) should have a section on how climate modification is expected to impact the effect discussed in that chapter, and then Chapter 13 brings together all the effects of climate change in an integrated/combined manner.

p. 4 line 5 (Praveen Amar) The report states that “Recommendations to improve Chapter 14 include: (1) incorporating a better definition of ecosystem services …” Do we need to use the term “ecosystem services VALUATION” as in “ecosystem services analysis and VALUATION”? 

p. 4 line 19 (Praveen Amar) Revise the sentence as follows: “…2) a section be added to Appendix C to compare various aspects of the five case studies…”

Panel Roster

p. ii (Daven Henze) Dr. Henze’s affiliation should be Associate Professor and Johnson faculty fellow.
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Individual Comments from Panel Members

p. A-49 lines 5 - 45 (James Galloway) There is some text duplication on this page.

p. A-53 line 41 through A-57, line 24 (Daven Henze) Replace Dr. Henze’s individual comments on Chapter 2 with the following edited version that was previously submitted.

2. Atmospheric Chemistry

Major comments for discussion:

Table 2-1: This table strikes me as overly precise; as mentioned in the supporting discussion, there are considerable uncertainties associated with many of these values. At the very least, it should specifically state the year and version of the NEI being used. At best, some estimates or uncertainties, ranges, or notes on levels of certainty (high, medium, low) could be included. Another question about Table 2-1 is the apportionment of soil NOx – about 10% of soil NOx emissions are owing to fertilizer applications (e.g., Hudman et al., 2012). Why are these then not classified as agricultural emissions, as is the emissions of NH3 owing to fertilizer?

2-6, 15-24: NH3 emissions are quite uncertain. This should be discussed, including references to the relevant literature. NH3 emissions from transportation may also be underestimated in the NEI by x2 (e.g., Sun et al., ES&T, 2017).

2-17, 27 through 2-21, 14: This very generic section on atmospheric transport feels a bit dated, and somewhat out of place. While some review of basic terminology relevant to transport discussions seems warranted, the content here seems a step or two removed from the aspects of transport critical for discussion of N and S. It seems like boilerplate description of transport associated with the Bermuda high copied from another report. This is but one mode of transport of interested to S and N deposition. Boundary layer dynamics and mixing, mountain up-slope effects (e.g., transport into Rocky Mt National Park from ag and urban areas to the east), continental-scale (e.g., transport to Rocky Mt. NP from CA) and transcontinental-scale long-range transport all come to mind.

Section 2.5: This section focuses on (some) ground network measurement techniques and remote sensing techniques. What about measurements from aircraft (e.g., work from John Nowak on NH3), mobile platforms (Mark Zondlo, also NH3), or other research-grade techniques? Or does the title of the section just need to be renamed?

2-29, Section 2.5.4.1: This section unfortunately does not include the best remote sensing measurements available for NH3 – those from CrIS. These measurements essentially combine the accuracy of TES with the spatial coverage of IASI. See details in Shephard and Cady-Pereira, AMT, 2015, and Dammers et al., AMTD, 2017.
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2-30, 1-4: This is a strangely incomplete and out of place overview of the use of remote sensing to constrain NH3 emissions. It doesn’t even mention the most relevant works targeting US emissions. Regardless, discussion of top-down emissions estimates should be the subject of another sections (e.g., end of Section 2.2) and should be kept separate from a description of the observations here.

2-39, Fig 2-11: Later, in Section 3 (3-12, 37; 3-13, 8-10), it is noted that exposure to HNO3 may have been a driver of declines in lichen in the LA basin, and may be continuing to have an impact. I believe this is the only direct gas-phase impact of S or N species referenced to believed to be occurring in ambient conditions in recent times in all of section 3. The cited studies of HNO3 impacts on lichen were referenced to daily peak exposure. Would it be useful then to present estimates of daily peak HNO3 values, rather than / in addition to annual averages?

Section 2, figures: Scanning across the figures of species distributions in this section leads to a misleading first impression as to the spatial variability of different compounds owing to some seemingly arbitrary choices of color-bar scales. For example, the distribution of SO2 concentrations looks quite flat, as the max value plotted is ~6 ug/m3 on a scale up to 20. In contrast, nitrogen oxide concentrations are plotted on a much better scale (Fig 2-9), such that gradients associated with particular sources (cities, power plants, and roads) are visible. Can all of the figures in this section be adjusted so that the max color-bar scale matches the max value on the map?

2-43, Section 2.6.6: Would it be worth providing information on current levels of 3hr max SO2 concentrations, as that is what the NAAQS are based on, instead of / in addition to the 3 year annual mean?

2-66, 3: The statement that N dep is overall in reduced form – is that based on the total budget of reduced N dep to oxidized N dep? Or does that mean there is a greater area where the former dominates? Is organic N accounted for in this statement? I thought (e.g., Zhang et al., ACP, 2012) that oxidized N dep was larger than reduced N dep in the US under current conditions, although that is expected to change in the near future as NOx emissions decrease and NH3 remain the same or increase (e.g., Ellis et al., ACP, 2013; Paulot et al., ES&T, 2013).

2-74: Fig 2-29, 2-30 and 2-31 -- any comment on the changes at the tip of Florida?

Section 2.9, 2-80…: This section delves into the details of a study by Koo et al. (2012) documenting variability across two models for transference ratios. Transference ratios are the inverse of atmospheric lifetimes, weighted by the ratio of the volume over which the concentration is calculated to the area over which the flux is calculated, ie the height of the system. Presumably since the latter isn’t a variable quantity across studies, variability in transference ratios is just variability in the lifetime of NOy, SOx, or NHx. And then I tend to think that variability in lifetime has been more widely studied across models.
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Section 2.10: I think there should be an additional paragraph or two in Section 2.10 (or an entire section) devoted to source-receptor estimates for N dep within the US. Lee et al. (ACP, 2016) evaluated the sources driving deposition of reactive N in several Federal Class I areas in the US. This study quantified the natural contribution, as well as provided maps of the footprints for each location. Locations such as Big Bend had substantial contributions from soil NOx (~20%) as well as foreign emissions. Another important point of this work was the domestic, yet long-range influence of some anthropogenic emissions on N deposition in remote parks such as Rocky Mountain, which was impacted by NOx and NH3 emissions from CA. This type of long-range influence (on deposition in RMNP in particular) was also previously identified in works such as Paulot et al. (ES&T, 2013), Benedict et al. (JGR, 2013) Malm et al. (JAWM, 2013), and Thompson et al. (JGR, 2015).

2-42, Section 2.6.4: Increasing trends in NH3 are mentioned in several other locations throughout the report (e.g., 2-87, 1) – have these trends been detected with measurements from AMoN or SEARCH? There are papers examining trends in NH3 from remote sensing instruments.

General: It struck me as a bit odd that there is a lot of discussion of CLs, but for the section on the distributions and trends in actual deposition values, the latter were presented with little reference to their magnitudes in relationship to CLs. Works such as Ellis et al. (ACP, 2013) and Lee et al. (ACP, 2016) broadly examine drivers of deposition above CLs, as well as others. Should this be discussed?

General: There were some pretty strong statements about the importance of bidirectional exchange, in a few locations. However, no results were shown for how bidirectional exchange impacts model estimates of concentrations or deposition. Could/should these be included?

Minor comments:

2-5, line 4-18: Evidence of NOx emissions overestimates also in Texas during the GoMACCS campaign (Yu, S. C., et al. (2012), Comparative evaluation of the impact of WRF-NMM and WRF-ARW meteorology on CMAQ simulations for O3 and related species during the 2006 TexAQS/GoMACCS campaign, Atmospheric Pollution Research, 3(2), 149-162.) And note the paper by Travis is now published.

2-6, 35: Some top-down studies don’t use CTMs, such as the constraints on NOx and SOx emissions derived from OMI (e.g., Fioletov et al., GRL, 2011).

2-14: What about uptake of HNO3 by coarse-mode dust?
2-17, 15: The lifetime of NOx is typically a few hrs, owing to rapid dry deposition. It seems odd to pit this against the lifetime of HNO3, which is also short, but I’m not sure that the latter is significantly shorter.

2-23, 33: sometimes models are used here as well

2-33, 3: Shouldn’t discussion of NO2 have been in the earlier section on OMI NO2?

Section 2.5.4: What about the techniques used in the SEARCH network?

Section 2.5.5.1: Should probably also include discussion of SO2 work from Fioletov et al., GRL, 2013, and others from NASA Goddard. There should also be a distinction between generating and using retrievals of SO2 columns, which aren’t intrinsically tied to GEOS-Chem, and extrapolating these column concentrations to estimates of surface concentrations, as done in Lee et al. (2011) and Nowlan et al. (2014).

2-40, 7-12: I believe that paper only identifies a 5% /yr trend in December. Another possible reason discussed was increasing domestic NH3.

2-54, 35: I wasn’t sure about the claim that aerosol nitrate is found “mainly” in the coarse mode in the Eastern US. Could this be clarified, and referenced? Are they referring to nitrate uptake on dust, or sea salt?

2-55, Fig 2-18: The axis labels are nearly illegible – could these be remade?

2-57, 4-6: Well, yes, that is bound to happen since the unidirectional model doesn’t include the upward flux.

2-59, 31: Is there any large-scale evidence of this type of NO2 emission from e.g. SOAS/SEACRS? As found in the BEARPEX study mentioned on the next page, much of this may be converted to other species before it exists the canopy.

2-70, 9: This result from Krotkov 2016 was just over the Eastern US (see their Fig. 1). But the decreases in NO2 are generally supported by evidence from remote sensing, reported in Krotkov as well as several other studies cited therein (Duncan et al., 2013; Lamsal et al., 2015; Lu et al., 2015; Russell et al., 2012; Tong et al., 2015).

2-70, 16: This is also owing to reduced levels of sulfate, and hence less NHx present as ammonium sulfate.

2-72, 27-32: Didn’t they also suggest increasing NH3 may play a role?
Comments from individual members of the Clean Air Scientific Advisory Committee (CASAC) Secondary NAAQS Review Panel for Oxides of Nitrogen and Sulfur to assist meeting deliberations. These comments do not represent consensus CASAC advice or EPA policy. DO NOT CITE OR QUOTE

1 2-75, 1-2: A little redundant with the text at the end of page 2-72.
2
3 2-90, 5-17: It may be good to also reiterate the results of Kim (2015) here as well, in terms of interactions between organics and inorganics impacting aerosol neutralization.
4
5 2-87, 18-26: As with the main body, the summary should be updated to reflect uncertainties in NH3 emissions.
6
7 Editorial:
8 2-17, 19-26: Perhaps this is just a writing style issue, but the text seems to make a point about NOx lifetime vs that of HNO3, and then backs this up (for example) by reference to the lifetime of NH3 and SO2, which doesn’t flow (logically). Perhaps all of these are just examples of the broader topic of this paragraph (different species have different lifetimes), which should be clarified.
9 2-20 16: Strangely worded, as emissions cannot be transported (emitted species can be though).
10 2-24, 26: which has shown \(\rightarrow\) have shown
11 2-52, 19: remove comma: models, typically
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