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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Sources of nitrogen deposition in Federal Class I areas in the US" *by* H.-M. Lee et al.

Anonymous Referee #2

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General comments:

This is an interesting manuscript that uses the GEOS-Chem adjoint model to investigate the contribution of various emissions source categories to the magnitude and spatial extent of critical load exceedances in Federal Class I areas. The authors first compare deposition estimates from the model against deposition values from observations and other modeling efforts. After demonstrating the ability of the model to provide reasonable values of deposition, the authors next explore the sensitivity of the nitrogen deposition to NOx and NH3 emissions using several different cost functions. Finally, the authors explore the sensitivity of the modeled deposition values to the emissions inventory used.

The scope of the paper as written is quite broad and I feel that the reducing the scope and enhancing the analysis of results from the adjoint model would improve the paper. The longest subsection in the Results section is devoted to the comparison of the model results with measurement data and other models while that is not the stated intent of the manuscript. Similarly, the comparison of the model results from using different emissions inventories is also lengthy and again detracts from the intent of the paper. Overall, I suggest reducing section 3.1 or maybe moving some of the information to the supplemental information and deleting section 3.3.

Additionally, given the importance of NH3 in the results, it would seem that a discussion of the potential impact of neglecting bidirectional exchange in the modeling should be included. There is a brief mention of bidirectional exchange in the future work but it is buried in the Discussion and Conclusions section.

Specific comments:

Pg 23090, Line 8 – I suggest replacing "measurements" with "deposition values". This gets around the need to explain (as you do later) that CASTNET values are not measurements.

Pg 23092, Line 21 – Do your "net emissions" here represent some sort of bidirectional exchange?

Pg 23093, Line 6 – I suggest rewording to "source contributions to the deposition at the collection of all"

Pg 23093, Line 13 – please define better "these specific areas" – maybe "These 8 focus areas" would help.

Pg 23094, Lines 10-11 – please correct grammatical mistakes

Pg 23094, Line 14 – ozone is measured hourly at CASTNET sites

Pg 23094, Lines 13-22 – How appropriate is the use of MLM for these areas? Late in

the paper, you do discuss the issues surrounding data in clearings (e.g. Hicks paper). This can definitely be an issue. Are the CASTNET sites representative of the Class I areas in terms of the mix of species, etc?

Pg 23094, Line 23 – it would be clearer to name the section "GEOS-Chem model description"

Pg 23096, Lines 1-29 – The potential impact of the large size of the grid cells is not noted until the last paragraph of the paper. I think there should be some discussion in the section. Also, it should be noted that bidirectional flux is not considered.

Section 2.3 – I don't with the order of presentation of the cost functions. It would seem to make more sense to discuss Jp first since it is the most basic definition and is also the first one discussed in the results. The explanations of the cost functions should have the same level of detail.

Page 23097, Line 1 – respect is missing a p

Page 23097, Line20 – it might be useful to insert "user defined" in front of "cost function"

Page 23098, Line 2 – maybe substitute chemical species for components?

Page 23098, Line 3 – clarify what "full-chemistry" simulation means?

Page 23098, Line 5 – I have trouble with "efficiency of impact" and "emission efficiency" (used later). I think a better explanation is required to understand why this calculation results in a determination of efficiency.

Page 23098, Line 23 – suggest "Total Nr deposition consists of all chemical species"

Page 23099, Lines 1-2 – "reduced" and "oxidized" are a bit vague without "Nr deposition" after them

Page 23099, Lines 9-13 – This section is a bit confusing since Du et al focused on wet deposition and your model estimates include wet and dry

Page 23099, Line23 – overestimated compared to?

Page 23099, Line 25 – HNO3 concentration or deposition?

Page 23099, Lines 24-27 – the sentence should be broken into multiple sentences

Page 23100, Line 5-6 – the phrase "owing to" is overused in the paper. Consider other wordings that might be clearer as to the relationship between the factor and the result.

Page 23100, Lines 7 – 28 – The notation in this section is very confusing between the "model" value and "Jp".

Page 23100, Line 12 – clarify this to be "wet deposition of" and "dry deposition of" these compounds.

Page 23100, Line 13 – what is the correlation between?

Page 23100, Lines 15-17 – I didn't find the later discussion of the winter deposition which could have been quite interesting depending on the level of sophistication of the treatment of deposition to snow for many of these areas.

Page 23100, Lines 21-22 – fix the grammar, please. Also, what model estimate are you referring to – Nr or a single species?

Page 23100, Lines 22-23 – It would be important to understand if the differences in HNO3 flux estimates from CASTNET and GEOS-Chem are due to differences in concentration or differences in the deposition velocity. I suspect, that for CASTNET sites where the meteorological measurements are taken in a clearing, the wind speed and stability are quite different than that for the grid model or than would be measured above the canopy. This has a great influence on the deposition velocity. You do cite the Hicks paper, but the discussion is limited.

Page 23101, Line 6 – Is dry deposition of NH3 14% of the total at all sites for all seasons?

Page 23101, Lines 9-11 – Clarify “organics” – maybe organic N?

Page 23101, Lines 11-12 – please fix the grammar

Page 23101, Lines 14 – 23 – This paragraph is a bit of a jumble of ideas and jumps back to figure 3.

Page 23101, Line20 - it would help to insert the word “deposition” after “Nr”

Page 23102, Lines 18-25 – The point of this paragraph is unclear as it really doesn’t present any new information.

Page 23102, Line 20 – add “deposition” after Nr

Page 23102, Line 22 – is versus the correct word?

Page 23102, Line 23 - add “deposition” after Nr

Page 23103, Lines 14-15 – the discussion of efficiency is unclear. Is it the efficiency of the impacts or the transport efficiency?

Page 23103, Lines 26-28 and Page 23104, Lines 1-3 – This discussion is not explained well and in some places makes little sense.

Page 23105, Line 1 – please be more specific than “more to the former, but less to the latter”

Section 3.3 – it is not clear what new information is gained from this section over previous sensitivities. You should clarify that or consider deleting this section to allow a more complete discussion of other sections.

Page 23106, Line 27 – do you mean HNO3 wet deposition at the end of the line?

Page 23107, Lines 8-9 – there appears to be a typographical or grammatical error

Page 23107, Lines 10-13 – this sentence seems misplaced as it does not follow from the previous sentence.

Page 23107. Lines 24- 29 – We do expect NH3 emissions to increase in the future, but Nox emissions to decrease. How does this impact your efficiency analysis?

Page 23108, Lines 8-16 – This section is not very clear. Line 11 contains a type (should be “one”?). It would be helpful to define better the two approaches.

Figure 5 – I would suggest changing the text on the first line to “in each Class I area”. If this is a sensitivity, should the units be deposition per kg emission? If that is not the case, then a better explanation is needed. Are only the footprint values scales or are the cost function values also scaled?

Figure 8 – Is this Jp in the figure?

Figure 11 – I suggest adding text to the caption to remind readers what Ja and Jc are.

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 23089, 2015.