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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.

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General comments:

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

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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.

> We appreciate the suggestions. We have reduced the extent of section 3.1 (now it is actually extended as explanations on model uncertainty are added). We have kept section 3.3, but have added additional text to the introduction to clarify the scope of the study. We feel the uncertainty in NH3 sources is an important aspect of understanding our results, as pointed out by the reviewer's comment regarding the lack of treatment of bidirectional exchange of NH3. We have also included more discussion on bidirectional exchange.

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.

> Fixed as suggested.

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

> No, this version does not include bidirectional exchange. We have changed "net" to "total" in this location to avoid confusion regarding this aspect, as here we meant to refer to the total emissions from the entire US.

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

> Revised as follows:

"To accomplish this goal, we evaluate source contributions to the deposition at the

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collection of all Class I areas as well as eight..."

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

> Revised as suggested.

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

> Revised as follows:

"For GT, we use the average of Yellowstone and Pinedale, WY, measurements because there are no wet deposition measurements made in 2010 in GT."

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

> As ozone is not used in this study, O3 is removed here.

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?

> There is a non-physical component (vertical level dependent leaf boundary layer resistance) in the MLM formulation as implemented in CASTNET (Saylor et al., 2014). The non-physical component makes HNO3 overestimated at some of the CASTNET sites by about 10 - 32%. On the other hand, Hicks (2006) suggests underestimation of CASTNET dry deposition derived from clearing data, which needs to be about twice as large to be consistent with the forest measurement. It is indeed unlikely the CASTNET sites are representative of the mix of conditions present throughout the Class I areas, or those present throughout the extent of the model grid cells containing these areas.

Now the text has revised in line 274:

"Lastly, comparison to dry deposition measurements warrants some additional consid-

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erations. The MLM model used for deriving the CASTNET dry deposition values is subject to uncertainty in estimating dry deposition velocities (Schwede et al., 2011) because of a height dependent non- physical component that can lead to overestimate of HNO3 deposition by 10-30% (Saylor et al., 2014). Additionally, Hicks (2006) found that measurements of HNO3 dry deposition in a clearing, such as the CASTNET sites in SM and SD from which dry deposition measurements are derived, are lower than measurements of dry deposition to the surrounding forest canopy. Thus, measured Nr deposition in Class I areas that have large forested areas (such as SM, SD, RM, GT, and SQ, see Fig. 1) is likely underestimated."

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

> Revised as suggested.

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.

> The issue of model resolution is now recognized when considering the comparison to measurements (line 270):

“Additionally, our model grid-cell size (∼3350 km2) is larger than the largest Class I area (BB, 2866 km2). Representational error may thus also contribute to the discrepancy between the model and the measurement for regions with large emissions within grid cells containing the Class I area (e.g., SM and SD).”

Also, it should be noted that bidirectional flux is not considered.

>Bidirectional flux is now discussed in the introduction (line 98):

“Another consideration is that the air-surface exchange of NH3 emissions is actually bidirectional (Nemitz et al., 2001; Sutton et al., 2007), an aspect that has recently begun to be implemented to air quality models (Cooter et al., 2010; Bash et al., 2013; Pleim

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et al., 2013; Zhu et al., 2015). Zhu et al. (2015) found increased net NH3 emissions in July (5.9%) and decreased net NH3 emission in April (23.3%) and October (13.9%) over the US when including the bi-directional flux of NH3 in the GEOS-Chem model. As bi-directional flux of NH3 is not considered in our present work, this provides additional motivation for studying the response of Nr source attribution to uncertainties in NH3 emissions."

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.

> The order of the cost functions has been updated and their explanations expanded.

Page 23097, Line 1 – respect is missing a p

> Fixed.

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

> Revised as suggested.

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

> Revised as suggested.

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

> Revised as follows:

"full-chemistry" simulation, which considers NOx-Ox-HC-aerosol chemistry.

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.

> Use of the phrase efficiency has now been clarified as (line 209):

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"Non-normalized sensitivities quantify the change in the cost function per change in kg emission. We thus refer to this type of sensitivity as an efficiency in that large non-normalized sensitivities indicate areas where reducing Nr emissions would have a very strong impact on Nr deposition in terms of the response of Nr deposition achieved per amount of emissions reduced (as opposed to locations where reducing emissions would have little effect on Nr deposition in the areas of interest, or locations where Nr emissions are just large in magnitude). These are defined as ..."

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

> Revised as suggested.

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

> Nr deposition is added after both.

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

> This discussion has been clarified to more clearly refer to wet deposition alone.

Page 23099, Line23 – overestimated compared to?

> Revised: overestimated compared to observation

Page 23099, Line 25 – HNO3 concentration or deposition?

> Revised: HNO3 deposition

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

> Revised as suggested now in line 248:

“Further, comparison of modeled to measured HNO3 in Zhang et al. (2012) required consideration of sub-grid concentration gradients near the surface. Simulated ambient HNO3 concentrations are also overestimated (Heald et al., 2012), possibly owing to

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excessive N2O5 hydrolysis. This suggests that oxidized Nr may be overestimated in our study."

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.

> Changed to "because of"

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

> We have clarified in several places in this paragraph that Jp is the modeled deposition including non-measured species.

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

> Revised as suggested.

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

> As stated in the Figure caption, the R2 values refer to the correlation of the measured and modeled (same set of species) deposition. This is now also clarified in the text as:

"The squared correlation coefficient (R2) of measured and modeled Nr is shown in each plot."

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.

> We have removed "which will be further discussed..."

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

> This refers to the modeled Nr species that can be compared with the measured Nr

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species (as opposed to Jp, which includes non-measured species). This text is revised as (line 269): “Modeled Nr deposition is...”

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.

> It is discussed in a previous comment.

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

> Fixed: Dry deposition of NH3 accounts for 14 % of contiguous US total annual Nr deposition.

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

> Revised to “Organics (PANs and alkyl nitrate)”

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

> Revised as (line 301):

“... we expect organics to be underestimated in our model because . . .”

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

> This paragraph has been moved up one paragraph and revised to more clearly focus on discussion of annual Nr deposition values and their relation to annual critical loads.

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

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> Revised as suggested.

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

>The point of this paragraph is to highlight aspects of the source-attribution for RMNP, a subject of several other studies, which are now cited in the first sentence of this paragraph (which itself we have moved to the end of this section, lines 337 – 344).

Page 23102, Line 20 – add "deposition" after Nr

> Revised as suggested.

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

> This has been rephrased now in line 341 as:

"... the spatial distributions of the emissions of NH3 compared to those of NOx surrounding the park."

Page 23102, Line 23 - add "deposition" after Nr

> Revised as suggested.

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

> It is the efficiency of emissions changes to impact Nr deposition through transport. This is perhaps now clarified given our expanded explanation of the term efficiency on section 2.4.

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

> This discussion (now lines 355) has been revised for clarity.

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

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latter”

> This text has been revised as (line 384):

“Nr originating from Idaho, Utah, Washington, and Arizona contribute more to reduce the extent of Class I areas in CL exceedance but less to the amount of excessive Nr in Class I areas, as the Nr deposition in these regions is not as excessive as it is in other regions, as shown in Fig. 10.”

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.

> We appreciate that the motivation for section 3.3 was not clear. We have thus added more explanations about uncertainties in NH3 emissions in the introduction of the papers so that this section is better connected to entire paper (line 88):

“The secondary purpose is to evaluate the impact of uncertainties in NH3 emissions on source attribution of Nr deposition. NH3 emissions are known to have uncertainties of more than a factor of two in total US emissions in some seasons (e.g., Henze et al., 2009; Paulot et al., 2014). Thus, NH3 emissions inventories are often updated through top-down approaches, using constraints provided through inverse modeling of wet deposition measurements (e.g., Gilliland et al., 2003, 2006; Zhang et al., 2012; Paulot et al., 2014) or, more recently, remote sending observations (Zhu et al., 2013). Here we consider constraints on NH3 emissions throughout the US from Zhu et al. (2013) that were derived from 4D variational assimilation of NH3 remote sensing observations from the Thermal Emissions Spectrometer (TES) aboard the Aura satellite (Shephard et al., 2011). We investigate the impacts of these adjustments to NH3 emissions, relative to those from a national emissions inventory, on source attribution of Nr deposition in 3 Class I areas (VY, SD, and RM).”

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

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> Yes, thanks. This has been fixed.

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

> Corrected.

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

> This sentence has been moved to the beginning of the paragraph on line 428.

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?

> To address this question we have added discussion on line 457:

"As it is expected (e.g., Ellis et al., 2013) that NH3 emissions will increase and NOx emissions will decrease in the US in the coming decades, the formation of ammonium nitrate will increasingly be limited by NOx. This will cause the sensitivities of deposition that contains considerable contributions from ammonium nitrate (e.g., VY, GT) to be increasingly sensitivity to perturbations in remote NOx emissions, even though NH3 emissions will make larger contributions to total Nr deposition."

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.

> Fixed to "one." The two approaches are defined as:

"emissions contributing to the extent of the total area of Class I areas that are in CL exceedance (Ja) and emissions contributing to the magnitude of the excessive Nr deposition above CLs (Jc)."

These qualitative definitions map to the definition of Jc and Ja, which are now referenced for further guidance.

Figure 5 – I would suggest changing the text on the first line to "in each Class I area".

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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?

> Revised as suggested: “in each Class I area”. We now direct readers to Eq. 5 where the sensitivity is defined.

Figure 8 – Is this Jp in the figure?

> Yes, J is revised to Jp.

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

> Revised as suggested.

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/15/C10098/2015/acpd-15-C10098-2015-supplement.pdf

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