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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.

H.-M. Lee et al.

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Response to referee #1

General comments:

I find that the presentation of results is confusing in many areas due to the cost functions. It would be much clearer and easier to follow if the cost function was described in the main text instead of using the Jx abbreviation. It also seems like the main goal of doing the cost functions are missing until you get to the discussion and conclusion section where they seem to be clearly explained.

> We apologize for the confusion caused by notation and brief explanation. To help make it clearer and easier to follow, we changed the order of the definition of the cost

functions in the methods section to be consistent with the order in which they are presented in the results, expanded and revised the explanations in the methods section, and refer to cost functions by their meaningful name repeatedly throughout the text, rather than by their mathematical abbreviation alone.

Specific comments:

Line 184: what does “efficiency of emissions” mean?

> We have now clarified what we mean by efficiency as follows the first time this phrase is used (line 209):

“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 asâĂĺ...”

Line 217: How do this line and the first line of the paragraph reconcile if oxidized Nr deposition was 24% less in the current study and wet HNO3/NO3 and dry NO3 were similar but are the majority components of oxidized Nr deposition?

> The oxidized N deposition in Zhang et al. (2012) is 31% higher than that in our simulation, consistent with the NOx emissions in Zhang et al. (2012) being 27% higher than those used in ours.

Line 219: how much smaller?

> As suggested by Reviewer #2, we have revised the manuscript to focus less on inter-model comparisons and have thus removed the detailed description of comparison to Zhang et al. (2014) in this location. Regardless, to answer the reviewer’s question, dry NO2 is 0.64 (Zhang et al., 2014) and 0.21 (this study). Dry N2O5 deposition is 0.18

ACPD
15, C10084–C10097, 2015

(Zhang et al., 2014) vs. 0.083 (this study). Units are Tg N/yr.

Line 220: It is unclear if the discussion of HNO3 in the Zhang et al (2012) paper is about wet, dry or both types of deposition.

> It is both. “wet and dry” is added now in line 246.

Line 223: I don’t understand the second part of the sentence and how it fits with the first part “and ambient HNO3 concentrations are overestimated” -where are they overestimated?

> This has been clarified as following now in line 245:

“Zhang et al. (2012), using the same model we use but with the different emissions, found that HNO3 deposition is overestimated compared to observations when the model’s isoprene nitrate is treated as HNO3, as in our simulation, rather than being treated separately as organic nitrate.”

Line 231: What are the differences models to get higher dry deposition of reduced N? Is it strange the difference is occurring only in 1 region?

>As suggested by Reviewer #2, we have revised the manuscript to focus less on inter-model comparisons and have thus removed the comparison to Schwede and Lear (2014) at this point (although it is still mentioned a few paragraphs later). Regardless, the differences are likely owing to the use of different emissions inventories as well as the different deposition schemes and resolutions used in the GEOS-Chem vs CMAQ simulations. Comparison of our results to those of Schwede and Lear (2014) is now revised as follows (line 251):

“Schwede and Lear (2014) generated maps of Nr deposition for multiple years, including 2010. These maps display localized hotspots in parts of Colorado and Idaho that are not evident in our results. The high Nr deposition in these regions is attributed to dry deposition of reduced nitrogen (Schwede and Lear, 2014), whereas in our result the contribution of reduced nitrogen deposition is generally less than that of oxidized

ACPD
15, C10084–C10097, 2015

nitrogen deposition (Fig. 2), possibly owing to the aforementioned overestimation of HNO3."

Line 236: I understand the tracking of gas phase species in wet deposition to understand the source of the deposition but in Figure 4 I would like to see HNO3 and NO3- (and NH3 and NH4+) wet grouped together or at least plotted next to each other.

> The figure has been updated as suggested.

Line 237: The correlation of what? You have three variables plotted.

> 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 (line 262) as:

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

Line 239: The low correlation of JT is not in the next paragraph. I can't find a discussion of the winter deposition at JT in the manuscript.

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

Line 242 and 243: Are both of these monthly values actually 1.3?

> Yes. The max value for measured Nr deposition in Voyageurs in July is 1.3 and the max value for modeled Nr deposition in Smoky Mountain in July is 1.3. The min/max range is not evident from Fig 3 alone (which just shows the standard deviation within the season), which is why we provide the value in the text.

Line 243: It would be much easier to read and follow the discussion if Jp wasn't used. I suggest either no abbreviation or one that makes sense (you think of simulated Nr deposition) Maybe measured Nr, modeled Nr, and modeled Nr+

> We now define at the top of this paragraph on line 258 that "total modeled Nr depo-

ACPD

15, C10084–C10097, 2015

sition ... (Jp, which includes non-measured species)" and reiterate this definition later on line 268 as "Jp (modeled Nr including non-measured species)".

Line 245-248: You already said this at the end of the previous paragraph (line 230). Can you add their data to Figure 3 to show the difference?

> As mentioned in response to a previous comment, this section discussing the results of Schwede and Lear (2014) has been consolidated.

Line 254: What organics?

> It is now stated on line 298 "Organics (PANs and alkyl nitrate)"

Line 261: Can you somehow identify which are in exceedance annually? Some of the sites are easy to see how this would be the case but for GT it might not be.

> The text has been clarified as follows on line 285:

"Class I areas considered to be in CL exceedance on an annual basis based on simulated values are VY, SM, SD, RM, GT, and SQ and those in exceedance based on measurement are VY, SM, SD, RM, and SQ".

Line 273: The maps in Figure 5 are interesting but you don't really discuss them until you talk about Figure 7 and then you don't even discuss them directly. Are they necessary? I think they are interesting and an important component of the results and discussion. Is there a way to combine Figures 5 and 6? Color scale from oxidized/blue to reduced/red (or something)...

> Figure 5 and 6 are based on sensitivities to different cost functions, so they cannot be combined. We have moved discussion of the maps and spatial distribution using Fig.7 up one paragraph and moved the description of Figure 6 to the end of the section. Thus the figure order and number have changed.

Line 286: The focus on RM seems to take away from the other sites more than it adds to the discussion.

ACPD
15, C10084–C10097, 2015

> Given the prevalence of other studies, both measurement-based and modeling, on source attribution of Nr deposition in RMNP, including the recently published paper by Thompson et al. (2015), we feel a more in depth consideration for this park is warranted.

Line 308: Remove efficiency from the section title. It doesn't make sense without more explanation.

> We have further clarified how this form of sensitivity analysis relates to an "efficiency" in Section 2.4, thus we retain the use of this word in the section title.

Line 311: I'm not sure I understand how these results were calculated. Were the emissions actually changed?

> Calculation of adjoint sensitivities does not entail changing any of the emissions, although the results can be used to estimate the impact of such changes. The methods used here have been further explained on line 346 as:

"For Class I area, we also calculate the non-normalized adjoint sensitivities defined with Eq. 4 using the cost function from Eq. 1 (Jp). These provide estimates of the response of Nr deposition (Jp) in each park per kg emissions of NH3-N, NOx-N, and SO2-S in each month."

Line 315: I don't see the northern signal at JT in the summer.

> The text refers to sensitivities to the NW and SE of JT in the summer. While there is indeed a significant spread in the sensitivities in the W-E axis, that sensitivities are evident from as far north as San Francisco, CA, to as far south as Punta Chivato in Baja shows the distribution along the N-S axis.

Line 322 – 325: The logic here doesn't make sense or I'm misunderstanding the analysis. There is limited NH4NO3 because there is less NH3 in the winter but NH4NO3 has a longer lifetime than NH3 and HNO3 and Nr deposits far beyond the park.

ACPD
15, C10084–C10097, 2015

> We have attempted to explain this more carefully as follows now in line 358:

“These negative sensitivities occur because NH4NO3 formation is limited by NH3 in the winter in SD. In these conditions, emissions of NH3 promote formation of NH4NO3. Since NH4NO3 has a longer lifetime in the atmosphere than gas-phase NH3 or HNO3, formation of NH4NO3 causes Nr to be transported further away, and thus less Nr deposits in the park. Thus, the deposition of Nr in the park has a negative sensitivity with respect to NH3 emissions”.

Line 348: I suggest adding a reference to Figure 11a here to make it clear which of the figures you are talking about. And a few lines later add one for Figure 11b.

> Thank you for the suggestion. The text has been revised as:

“Decreasing Nr emissions in California and regions surrounding RM and SM would be useful for reducing both the extent of Class I areas in CL exceedance (Fig. 11(a)) and the amount of excessive Nr in Class I areas (Fig. 11(b)).”

Line 374: Showing the difference between the Optimized NEI2005 and 2005 might more clearly make your point and the maps would be larger and easier to see.

> As we make use of the maps of the emissions themselves for understanding features of the sensitivities, we hesitate to show only a difference plot. We could add a difference plot as another column, but that would make the current plots even smaller.

Line 375: Please redefine Jp here.

> The definition of Jp has been reiterated on line as:

“sensitivities of Jp, total modeled Nr deposition in individual Class I areas,...”

Line 394: What is wet nitric acid?

> Nitric acid dissolved in wet convective mass fluxes and precipitation fluxes. This has been clarified now on line 434 as: “from wet HNO3 deposition”

ACPD
15, C10084–C10097, 2015

Line 421: What are effective regions? Regions where emissions reductions would be effective in reducing N dep? Please clarify.

> We have now defined this term on line 465 as:

"... effective regions, where emissions from the region would contribute to more than $\sim\pm 1.0\times 10-8$ kg N/ha/yr per kg N emission or $\sim\pm 1.0\times 10-9$ kg N/ha/yr per kg S emission, are ..."

Line 430-433: Is there a place and benefit to discussing the importance of local versus long range transport at the different class 1 areas?

> The importance of local vs long-range transport for different Class I areas is discussed on lines 444-446:

"Quantifying the contribution of local versus long-range transport and the contribution of different sectors to Nr deposition may serve as a guide for devising locally-tailored strategies to reduce Nr deposition in different Class I areas"

Figure 3: The legend is confusing. Is there a way to make it clearer that the model is the same as measurements while cost function includes extras. Model+? I know the difference is explained in the caption.

> As the legend appeared to be confusing, we removed it so that readers will focus more on the explanation in the caption.

Figure 4. Can you combine figures 3 and 4? Height of bar is the blue diamond and you can use a light colored symbol for the model and measured. And put other deposition pathways that aren't in measured/modeled on the top of the bar?

> An interesting suggestion. To do so it would make most sense to stack the bars such that the measured quantities are all at the bottom. But this would conflict with the desire to lump by oxidized vs reduced species, as suggested in a previous comment.

Figure 5. Can you look in on the region of interest/extent of significant deposition

ACPD

15, C10084–C10097, 2015

(east or west half of US)? In the caption last sentence, Footprint values are scaled for visibility with scaling factor in parenthesis.

> We considered this as well. It did make it easier to see the details in each region, but the collective view made it harder to recognize the area in question (the continental US being a more readily identified shape) and also harder to compare the extent of the footprint across regions, such as the footprint for JT and SQ being much smaller than that for other parks. Given these factors, and that the images in vector form can be arbitrarily and manually zoomed by the reader, we decided to stick with the current format.

The caption text has been modified as suggested.

Figure 6. Please indicate the park location. This would also be interesting for GT. Could you do for all with reds/blues in Figure 5.

> The location for Rocky Mountain National Park has been added in Fig 7 (previously Fig 6). Unfortunately we don't understand the second comment. All park locations are already indicated in Fig. 5 with black circles.

Figure 8. Can you add column titles to more clearly indicate summer and winter?

> JJA and DJF are indicated at the top of each column.

Figure 9. Can you inset the wind roses in Figure 8 maps? This would complement Figure 8 and make the explanation of results easier to follow.

> The figure has revised as suggested.

Technical corrections:

Line 160: with respect > Fixed.

Line 244: Our model estimate - no "s" > Fixed.

Line 316: owing ? > line 320. owing → due

ACPD

15, C10084–C10097, 2015

Line 429: one instead of on > line 433. Fixed.

Line 452: lower case n > line 456. Fixed.

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/15/C10084/2015/acpd-15-C10084-2015-supplement.pdf

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

ACPD
15, C10084–C10097, 2015

Interactive Comment

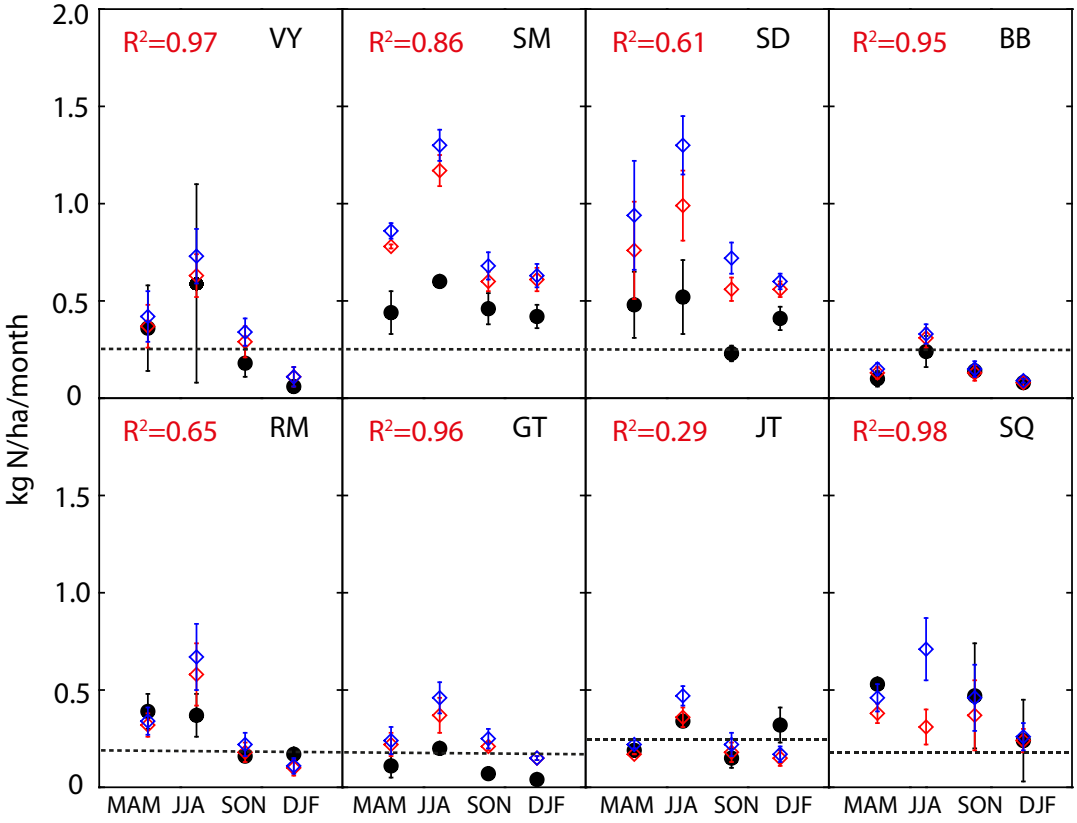

Fig. 1. Revised Fig.3

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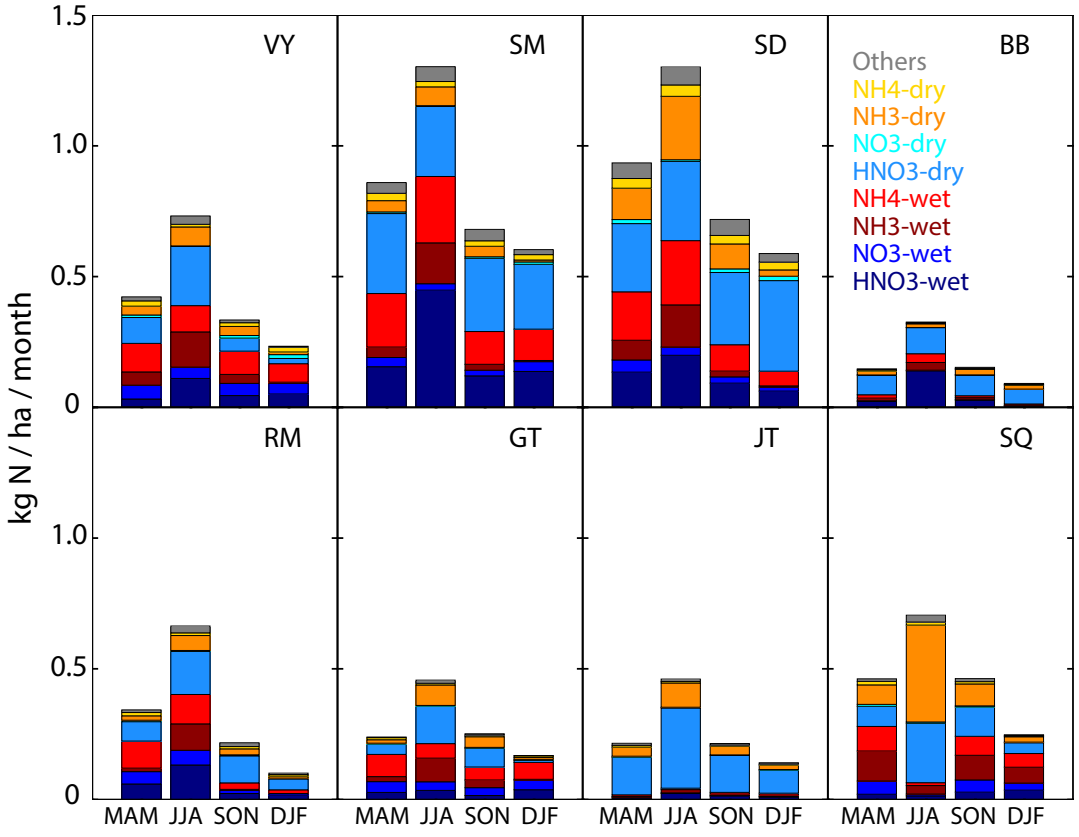

Fig. 2. Revised Fig.4

ACPD
15, C10084–C10097, 2015

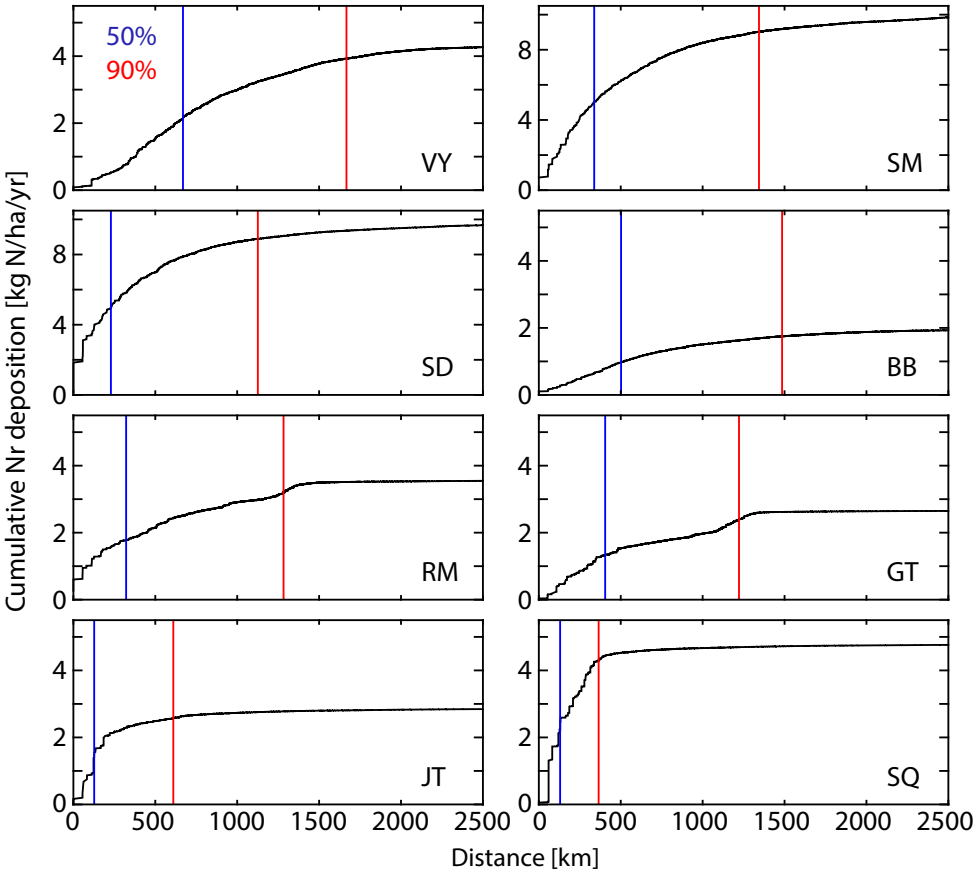

Fig. 3. Revised Fig.6

ACPD
15, C10084–C10097, 2015

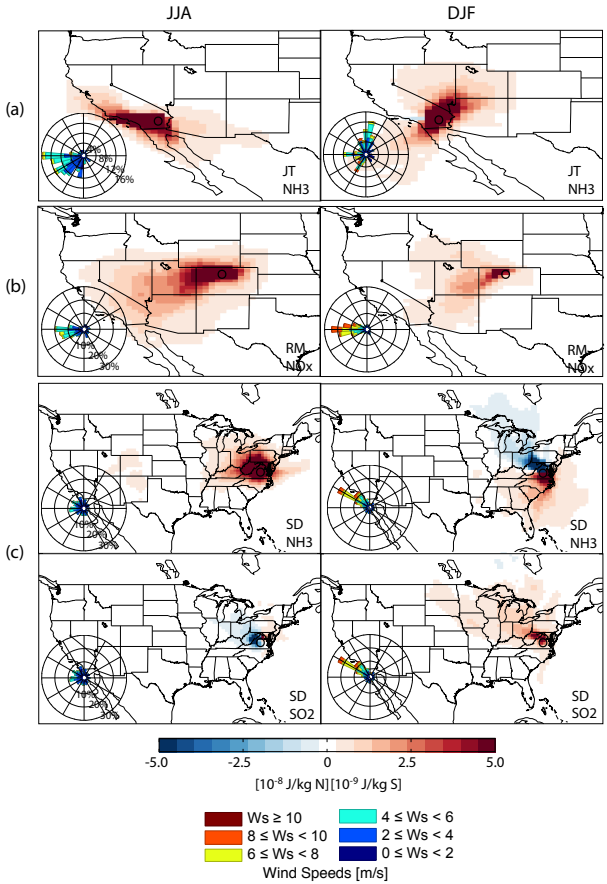

Fig. 4. Revised Fig.8

ACPD
15, C10084–C10097, 2015