

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 as ...”

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 NO_x 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 (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 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 (R^2) 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 deposition ... (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.

> 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 (J_p). These provide estimates of the response of Nr deposition (J_p) in each park per kg emissions of $\text{NH}_3\text{-N}$, $\text{NO}_x\text{-N}$, and $\text{SO}_2\text{-S}$ 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 NH_4NO_3 because there is less NH_3 in the winter but NH_4NO_3 has a longer lifetime than NH_3 and HNO_3 and Nr deposits far beyond the park.

*> We have attempted to explain this more carefully as follows now in line 358:
"These negative sensitivities occur because NH_4NO_3 formation is limited by NH_3 in the winter in SD. In these conditions, emissions of NH_3 promote formation of NH_4NO_3 . Since NH_4NO_3 has a longer lifetime in the atmosphere than gas-phase NH_3 or HNO_3 , formation of NH_4NO_3 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”

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 (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*

Line 429: one instead of on

> *line 433. Fixed.*

Line 452: lower case n

> *line 456. Fixed.*

Response to referee #2

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 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 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 considerations. 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 ($\sim 3350 \text{ km}^2$) is larger than the largest Class I area (BB, 2866 km^2). 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 NH_3 emissions is actually bi-directional (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 et al., 2013; Zhu et al., 2015). Zhu et al. (2015) found increased net NH_3 emissions in July (5.9%) and decreased net NH_3 emission in April (23.3%) and October (13.9%) over the US when including the bi-directional flux of NH_3 in the GEOS-Chem model. As bi-directional flux of NH_3 is not considered in our present work, this provides additional motivation for studying the response of Nr source attribution to uncertainties in NH_3 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 NO_x-O_x-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):*

“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, Line 23 – 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 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 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”

> *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 NO_x 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 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 sensing 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?

> 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 NO_x 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 NO_x emissions will decrease in the US in the coming decades, the formation of ammonium nitrate will increasingly be limited by NO_x. 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 NO_x 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". 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 J_p in the figure?

> *Yes, J is revised to J_p .*

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

> *Revised as suggested.*