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Interactive comment on "Source regions contributing to excess reactive nitrogen deposition in the Greater Yellowstone Area (GYA) of the United States" *by* Rui Zhang et al.

Anonymous Referee #2

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The manuscript by Zhang et al. considers the sources of reactive nitrogen deposition in the Greater Yellowstone Area (GYA). The topic is timely and of relevance to this journal. The paper is in general clearly organized, well written, and is easy to read; the figures and tables are descriptive and appropriate. In terms of findings, the authors do a thorough job of first evaluating their modeling results compared to available measurements and other modeling studies in the literature. An issue is that they find very significant overestimation of HNO3 and underestimating of NH3. They then present source attribution results. Overall, findings of sources being from oxidized vs reduced nitrogen, different sectors, and different source reasons are interesting and seem sensible. They also consider a sensitivity study to try to address some of the modeling

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Discussion paper

shortcomings. My major criticism in this regard though is that such analysis or consideration of model biases is not reflected in the reporting of results elsewhere in the manuscript nor the abstract âĂŤ given the rather significant model biases it seems results should be presented much more cautiously throughout. It would be useful if the authors could estimate some uncertainty ranges to their source attribution results âĂŤ for example do they think they are accurate to within 1%? 10% an order of magnitude? Detailed comments along this line as well as a few other minor points are described in detail below. Addressing these would amount to minor revisions.

Specific comments:

Abstract: The model biases for NH3 and HNO3 are significant. Suggest adding some material to the abstract to address how modeled SA results should be interpreted, given these biases. Suggest referring to SA results as they pertain to the model (i.e., "largest source contributions in the model..."), unless this disconnect between measured and modeled values is resolved.

Abstract: importance of boundary conditions is not clear without having stated where these boundaries are. Nor is it clear that influence across the boundary would be international in origin (as opposed to natural oceanic emissions, recirculated domestic Nr, etc).

1.26: I thought is was already established that Nr deposition is already in excess (see first sentence of the abstract), thus it is odd here to say that the "results suggest that Nr deposition ...was above critical loads".

2.17: Worth indicating that these numbers are approximate and perhaps specific to a particular time period given trends in emissions from these sectors.

2.20: Missing some references here, e.g. work from Zondlo's group.

3.14: for zero-out –> using zero-out

3.17: "found the importance of emissions from California" is a bit vague. Were these

found to be more important than local sources? Or more important than otherwise expected?

3.19: This paragraph feels rather tangential and could be removed from the introduction or significantly shortened so only the content as it relates to understanding Nr dep in GYA.

4.13 - 20: several studies in the past year have identified an overestimation of mobile NOx emissions in the NEI2011 inventory. How were these addressed in the present work?

4.13 - 20: Does the inventory here contain the amount of NH3 from mobile sources mentioned in the introduction, or is if felt that this inventory under-represents this source?

4.13 - 20: It would be very useful for answering these questions and others if the emissions totals by sector and species for the different tagged regions could be included in the supporting information and summarized in the text (as opposed to the summaries mentioned in the introduction, which reflect values in the literature but do not specifically refer to the values used in the modeling for this work).

5.14: As anthropogenic SO2 emissions have declined in the US, the role of NOx and NH3 in forming ammonium nitrate aerosol has increased. How would PSAT account for the influence of the EGU sector via SO2 on deposition of PNH4 and PNO3, or is this not accounted for?

6.9: Could the authors clarify what constituted questionable data, such that their results could be more reproducible?

6.22: Does the mechanism for formation of N2O5 in CAMx match that in GEOS-Chem? If not, it's not clear how the reference to Heald et al. (2012) is relevant here.

7.2: Is a unidirectional NH3 emission model expected to lead to larger NH3 concentrations in this region of the US than a bidirectional flux model?

7.2: I would suspect that another possible factor leading to poor correlation and underestimation for NH3 is the overestimation of HNO3, which would promote excessive partitioning of NH3 to the particle phase. Did the authors consider evaluating NHx, or HNO3+PNO3, to get around the issues of partitioning (and thus hone in on issues related to sources and sinks)?

7.7: Are the performance metrics referenced here relevant for a study focusing on Nr source attribution? I could imagine if a studies goal was to forecast total PM2.5 concentrations, then opposing large biases in e.g. NH3 vs HNO3 would be of little concern; here, these issues seem much more considerable in terms of their impact on the final conclusions. Overall, I think the authors need to do more work in this regards to convince the readers of the merits of the application of the model so SA in the presence of such errors and biases.

Fig 3: I find it interesting that the measurements at each site show a distinct reduction in NH3 dry dep in September, whereas CAMx shows a maximum in September for Driggs and Grand Targhee. Can this authors comment on this?

12: How much did reducing the NH3 dry deposition change the total NH3 deposition amounts and their underestimation compared to observations mentioned in previous sections?

13.1: It seems like earlier their were several possible reasons for this, such as overestimated HNO3 concentrations, and yet here only precipitation biases are considered?