Comments from anonymous referee #1

General comments
This manuscript reports on a modelling study, whereby the source sectors and regions of reactive nitrogen (Nr) are determined for the Greater Yellowstone Area in the United States. The model was evaluated thoroughly, and then used for quantifying source contributions to Nr deposition via a tagged model method. Agriculture from the Snake River Valley was determined to be the largest source. They took model error into account by doing a sensitivity study to give approximate uncertainties on the source contributions. This study represents new work as there is a lack of source attribution studies for Nr deposition for this region, however, I feel that they could emphasize further how their study is new, different, and important compared to previous studies.

Response:

Thanks for the recognition of the value of this modeling study and providing the opportunity for us to revise the manuscript accordingly. In order to emphasize the importance and new findings compared with previous modeling studies targeting nitrogen deposition in remote areas of the United States, we follow the suggestions of the reviewer to add a few sentences to emphasize how our study stands out compared with previous similar source apportionments. The detailed changes can be seen in the "track changes" version of the revised manuscript as well as in the responses to the specific comments below.

Specific and technical comments below.
Specific comments
p2, line 21: state where the 40% of NH3 emissions from mobile applies? U.S. urban areas? A national average?

Response:

The sentence: "Mobile sources are also an important source of NH3 and can be the primary emitter in urban areas. A recent study found the increasing importance of on-road emissions of NH3, which at 40% exceed agricultural emissions (Fenn et al., 2018)." Was modified to:

"Mobile sources are also an important source of NH3 and can be the primary emitter in urban areas (Sun et al., 2014; Sun et al., 2017). Emissions from this sector have large uncertainties and a recent study suggests that on-road NH3 emissions in the 2011 National Emissions Inventory (NEI) were underestimated by a factor of 2.9 (Fenn et al., 2018)."

p4, first paragraph: can you emphasize more what's new from your study? It simply says that it "add to a growing body of Nr modeling source apportionment studies"? For example; is your study more detailed than that of Zhang et al (2012) and Lee et al (2016)?

Does yours use a different technique (e.g., tagged model vs. zero-out scenario and adjoint model)? Is your study at higher resolution or does your model contain more detailed processes than GEOS-Chem? Etc. Emphasize why it was important to do this particular work despite the previous publications. Please also add to Section 6 to emphasize the importance of what's new in this study.

Response:

Based on the reviewer's suggestion, we revised this paragraph to the following:

"In this work, we add to the growing body of Nr modeling source apportionment studies by conducting a detailed analysis using the Particulate Source Apportionment Technology (PSAT) module within the CAMx (Comprehensive Air Quality Model with extensions) (Ramboll Environ, 2014) CTM to quantify the seasonal contributions from different source regions and source sectors to Nr throughout the GYA. Compared with previous Nr deposition simulation studies in United States, this work uses tagged reactive tracers to attribute the contributions from four designated emission sectors and 27 designated emission regions to Nr deposition in the GYA with a much higher horizontal grid resolution (12 km) and an up-to-date emission inventory instead of using a zero-out approach (e.g., Zhang et al., 2012) or an adjoint model (e.g., Lee et al., 2016). The model simulation of Nr and its constituents were first evaluated against routine measured data as well as the unique data measured during the GrandTReNDS campaign period (Benedict et al., 2013a; Prenni et al., 2014). Nr deposition from CAMx simulations was also compared with total deposition maps (TDEP), which were developed for deposition trend analysis and ecological impact assessment (Schwede and Lear, 2014). The detailed source apportionment results are presented here, focusing on seasonal variations and the relative importance to CL exceedance in sensitive ecosystems within the GYA. The discussion of identified model bias and uncertainties to source apportionment results interpretation, including the model lateral boundary conditions, the impact of model precipitation to wet deposition simulation, and the impact of ammonium dry deposition velocity to dry deposition are also presented."

Also, in section 6, the first paragraph, we added a sentence to emphasize the uniqueness or the importance of our modeling work here:

"Nevertheless, this N_r source apportionment work is the first thorough analysis of the origin of inorganic Nr in the GYA using a regional air quality modeling platform. The detailed source sector and source region configurations in PSAT enabled quantitative, though uncertain, estimates of their relative importance. This is needed information by stakeholder and regulator groups to understand the causes of excess Nr deposition in the GYA, monitor changes in Nr deposition and develop possible future mitigation strategies" *p4, line5: The sensitivity tests you did are an important part of this paper. I suggest emphasizing this more here in the introduction that this was done, given the large model biases.*

Response:

We changed the sentence from "The final source apportionment results are then interpreted within the context of the identified model bias and uncertainties" to "The discussion of identified model bias and uncertainties to source apportionment results interpretation, including the model lateral boundary conditions, the impact of model precipitation to wet deposition simulation, and the impact of ammonium dry deposition velocity to dry deposition are also presented"

p7, line 8: Comparing Table 1 in this paper to Figures 8, 11, and 12 in Simon et al, (2012), and it seems like CAMx model performance is within the range reported in Simon et al. However, just because it is within the range of what other models do, it doesn't necessarily follow that the model results are "adequate". Also the Simon et al. (2012) paper summarizes results published between 2006-2012, whereas model publications 2013-2017 may have improvements. Can you please add a few more recent references which have similar model biases as yours, and add some further justification to what is meant by "adequate"?

Response:

We do not explicitly use the word "adequate" in the description of the base model performance from CAMx in 2011. As requested, we added additional citations from the model publications from 2013 to 2017 with similar model biases to justify that the modeling platform we were working with has the capability to capture the general spatial and temporal variations of the reactive nitrogen in the atmosphere and that the model performance is in line with the peer modeling results applied for the continental United States using regional photochemical models (e.g., CMAQ and CAMx). Also, we provided Table S3 in the supplementary material to summarize model performance of series simulations with nitrogen-deposition-related species.

We deleted the sentence referring only to the Simon et al. (2012) study and added the new description at the end of this section as follows:

"Table S3 provides a comparison of regional air quality model, N- related species performance, evaluated by observations over the United States from peer-reviewed studies in recent years (e.g., Simon et al., 2012; Bash et al., 2013; Zhang et al., 2013; Yu et al., 2014; Thompson et al., 2015; Li et al., 2017), and it shows that our results are comparable, with some similar model biases such as overestimation of HNO3 and underestimation of NH3. Overall, the CAMx results provide a reasonable platform for evaluation of the contribution of sources to Nr deposition throughout the GYA."

Table S3. Summary of regional air quality model nitrogen related species performance in terms of normalized mean bias (NMB) evaluated by observations over the continental United States

Species	Photochemical model	Duration	Model resolution	Region evaluated	NMB value	Reference
NH3	CAMx	2011 full year	12km	GYA	-65%	this study
	CMAQ,CAMx	Jan, Jul 2002	4km	Southwest US	[-23% -79%]	Zhang et al. (2013)
	CAMx	2009 full year	36km/12km	Colorado	-55%	Thompson et al. (2015)
	CAMx	Summer 2011	4km	Colorado	[-83% 46%]	Li et al. (2017)
HNO3	CAMx	2011 full year	12km	GYA	108%	this study
	CAMx	2009 full year	36km/12km	Colorado	23%	Thompson et al. (2015)
	CMAQ,CAMx	Jan, Jul 2002	4km	Southwest US	[-17% 45%]	Zhang et al. (2013)
PM25 nitrate	CAMx	2011 full year	12km	GYA	[37% 58%]	this study
	CAMx,CMAQ, WRF-Chem (n=34)	varies, case study to full year simulation	9-45km	varies, states to CONUS	[-49% 11%]	Simon et al. (2012)
	CMAQ,CAMx	Jan, Jul 2002	4km	Southwest US	[-92% -103%]	Zhang et al. (2013)
	CAMx	2009 full year	36km/12km	Colorado	57%	Thompson et al. (2015)
	CMAQ	2002 full year	12km	CONUS	[-24% 45%]	Bash et al. (2013)
	CMAQ	1990-2010	108km	CONUS	[-41% 106%]	Xing et al. (2015)
	WRF-CMAQ	Aug, Sep 2006	12/4km	CONUS/Texas	[-82% 83%]	Yu et al. (2014)
PM25 ammonia	CAMx	2011 full year	12km	GYA	3%	this study
	CMAQ,CAMx	Jan, Jul 2002	4km	Southwest US	[-57% 61%]	Zhang et al. (2013)
	CAMx,CMAQ, WRF-Chem (n=31)	varies, case study to full year simulation	9-45km	varies, states to CONUS	[-17% 7%]	Simon et al. (2012)
	CAMx	2009 full year	36km/12km	Colorado	-31%	Thompson et al. (2015)
	CMAQ	1990-2010	108km	CONUS	[-54% 23%]	Xing et al. (2015)
	WRF-CMAQ	Aug, Sep 2006	12/4km	CONUS/Texas	[-75% 48]	Yu et al. (2014)
NOx wet deposition	CAMx	2011 full year	12km	GYA	31%	this study
	CAMx,CMAQ, WRF-Chem (n=16)	varies, case study to full year simulation	9-45km	varies, states to CONUS	[-45% 19%]	Simon et al. (2012)
	CMAQ,CAMx	Jan, Jul 2002	4km	Southwest US	[-94% 52%]	Zhang et al. (2013)
NH4 wet deposition	CAMx	2011 full year	12km	GYA	49%	this study
	CAMx,CMAQ, WRF-Chem (n=16)	varies, case study to full year simulation	36km/12km	varies, states to CONUS	[-33% 28%]	Simon et al. (2012)
	CMAQ,CAMx	Jan, Jul 2002	4km	Southwest US	[-51% 19%]	Zhang et al. (2013)
	CMAQ	2002 full year	12km	CONUS	[-16% 18%]	Bash et al. (2013)

We also add the following citations to the reference list:

Bash, J.O., Cooter, E.J., Dennis, R.L., Walker, J.T., and Pleim, J.E. (2013), Evaluation of a regional air-quality model with bidirectional NH3 exchange coupled to an agroecosystem model, Biogeoscience, 10, 1635-1645, doi:10.5194/bg-10-1635-2013.

Li, Y., Thompson, T.M., Damme, M.V., Chen, X., Benedict, K.B., Shao, Y., Day, D., Boris, A., Sullivan, A.P., Ham, J. and Whitburn, S.: Temporal and spatial variability of ammonia in urban and agricultural regions of northern Colorado, United States, Atmos. Chem. Phys., 17(10), 6197-6213, 2017.

Thompson, T.M., Rodriguez, M.A., Barna, M.G., Gebhart, K.A., Hand, J.L., Day, D.E., Malm, W.C., Benedict, K.B., Collett, J.L. and Schichtel, B.A.: Rocky Mountain National Park reduced nitrogen source apportionment, J. Geophys. Res., 120(9), 4370-4384, 2015.

Xing, J., Mathur, R., Pleim, J., Hogrefe, C., Gan, C.M., Wong, D.C., Wei, C., Gilliam, R. and Pouliot, G., Observations and modeling of air quality trends over 1990–2010 across the Northern Hemisphere: China, the United States and Europe. Atmos. Chem. Phys., 15, 2723-2747, 2015.

Yu, S., Mathur, R., Pleim, J., Wong, D., Gilliam, R., Alapaty, K., Zhao, C. and Liu, X., Aerosol indirect effect on the grid-scale clouds in the two-way coupled WRF-CMAQ: model description, development, evaluation and regional analysis, 14, 11247-11285, Atmos. Chem. Phys., 2014.

Zhang, Y., Olsen, K.M. and Wang, K., Fine scale modeling of agricultural air quality over the southeastern United States using two air quality models. Part I. Application and evaluation. Aerosol Air Qual. Res., 13(4), 1231-1252, 2013.

p10, line 12: it is mentioned above this that NH3 from agriculture is emitted into the first model layer and therefore doesn't get transported as far. Can you please also discuss the fire emissions – specifically how high they get put into the model? It is described a bit on p4, lines 19-20, but can you mention here approximately how high the fires spread in the vertical, and thus how it would affect deposition at some distance downwind?

Response:

We used the fire emissions developed from the Particulate Matter Deterministic and Empirical Tagging and Assessment of Impacts on Levels (PMDETAIL) study (Moore et al., 2012). The emissions for fire activities include prescribed fires and wildfires. In the PMDETAIL fire plume rise methodology (Mavko and Morris, 2013), three parameters were defined to provide the release heights of fire smoke emissions as hourly inputs to CAMx, namely (1) height above ground of plume top (P_{top}), (2) height above ground of plume bottom (P_{bot}), and (3) the fraction of emissions emitted near the ground (f_{Lay1}). When allocating the fire emissions to different vertical layers according to the CAMx vertical layer setting, the PMDETAIL methodology included the WRF estimated hourly planetary boundary layer (PBL) in the grid cell containing the fire emissions and injected the fire emissions near the surface between the CAMx model layer 1 and the maximum of P_{bot} and PBL values:

Fire emission (f_{Lay1}) = ground to max.(P_{bot}, PBL)

For the elevated fire emissions, the PMDETAIL methodology released the emissions in layers between P_{bot} and the maximum of P_{top} and PBL value for the hour and grid cell of the fire:

Fire emission (1-f_{Lay1}) = P_{bot} to max.(P_{top}, PBL)

We did not have the detailed information for those three parameters for each fire accounted for in the PMDETAIL and used in the 2011 CAMx modeling. However, looking at the attached figure below, we can deduce that those three fire plumes in summer within the GYA were injected into the vertical layer between P_{bot} and the model PBL height so that it may be mostly mixed within the PBL and has the dominant impact to adjunct grids where the fire emission occurs. It has little chance to disperse higher and impact N deposition at a longer distance downwind.

We changed page 4, lines 19–20, from "PMDETAIL developed 2011 fire emissions using satellite data, ground detects, and burn scar and estimated the plume rise depending on fire size and type (Mavko and Morris, 2013)." to "PMDETAIL developed 2011 fire emissions using satellite data, ground detects, and burn scar and estimated the plume rise, depending on fire size and type. The hourly, nonsurface fire emissions were allocated to the proper CAMx vertical layers based on the model-predicted planetary boundary layer (PBL) height and the spanning of the plume top and bottom above the ground (Mavko and Morris, 2013)."

We added Figure S4 to the supplementary file to show that the fires occurring during summer 2011 near the GYA predominantly impacted the adjacent grids. Now the sentences on page 12, line 12 that describe the fire emission impact to seasonal N deposition in the GYA read as "The footprint of fire emission impacts depends on the simulated injection height of the fire plumes. The emissions from fires that occurred within the GYA during the summer and fall likely remained within the mixed layer and had less chance to be transported far downwind to impact more distance areas (Figure S4)."

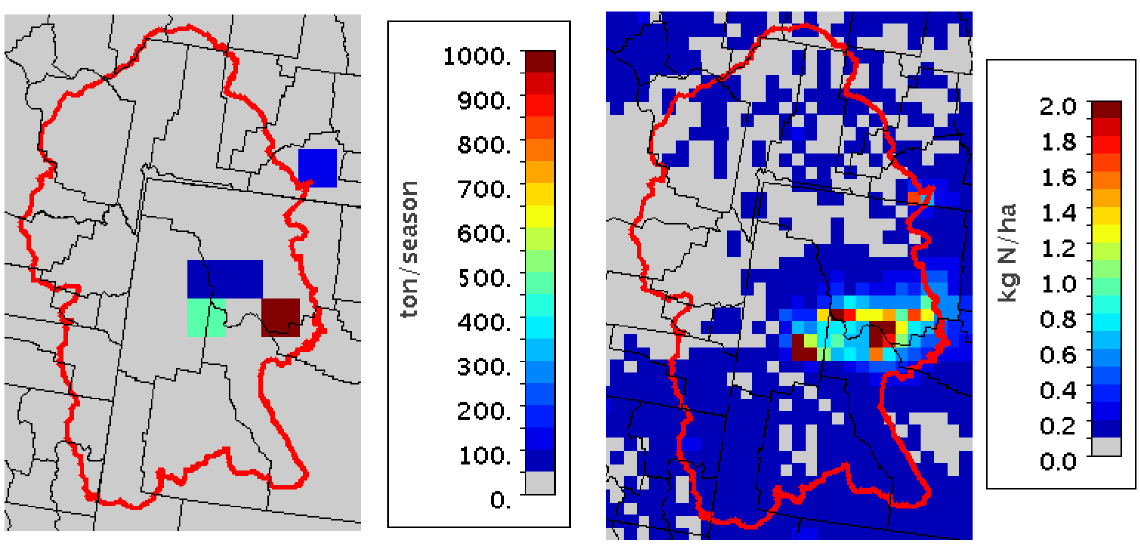

Figure S4. (left) Spatial pattern of total NOx emission from Fire emission sectors during summer (June, July, August) 2011 near the Greater Yellowstone Area (GYA). (right) the Spatial pattern of total N deposition attributed to Fire emission during summer 2011.

References:

Mavko, M. and Morris, R., DEASCO3 project updates to the fire plume rise methodology to model smoke dispersions. Air Science Inc. Portland, Oregon and ENVIRON International Corporation, Novato, California. December 3, 2013. http://wraptools.org/pdf/DEASCO3_Plume_Rise_Memo_20131210.pdf

Moore, C.T., Randall, D., Mavko, M., Morris, R., Koo, B., Fitch, M., George, M., Barna, M., Vimont, J., Anderson, B. and Acheson A., Deterministic and empirical assessment of smoke's contribution to ozone (DEASCO3), final report, 2012, Joint Fire Science, Program Project #11-1-6-6, https://www.firescience.gov/projects/11-1-6-6/project/11-1-6-6_final_report.pdf.

Technical corrections

p2, line 18: particulate nitrate (NO3), and other...

Response:

To be consistent with the notation in other places in the manuscript, such as page 5, line 14, and Table 1, we changed the sentence from "Atmospheric reactions of NOx result in nitric acid (HNO3), particulate nitrate, and other compounds." to "Atmospheric reactions of NOx result in nitric acid (HNO3), particulate nitrate (PNO3), and other compounds."

p6, line 22: may be related with the high: : :

Response:

Changed from "The poor NH3 results may related with the high …" to "The poor NH3 results may be related to the high …".

p10, line 19: There is no "Table S4" in the supplement document. The table on the last page of the supplement has no label, and doesn't seem to be what you're talking about here. I think you may mean Table S3.

Response:

We corrected the sentence to "Most (74%) of the Nr from this region was from the AG source sector and was composed of reduced N (Table S3)." The last table in the supplemental material belongs with the supplementary File S1 in the section "regional evaluation of CAMx nitrogen deposition in 2011" and is therefore not assigned a label.

p24, line 4: (caption to Fig 1) National Trend Network: typo in National

Response:

Corrected the typo from "Natiaonl" to "National".

p5, line 4: I expected to see the 24 tagged regions in Fig 1 given the text here, but actually that map is Fig S2. Text should be clarified. And I feel that knowing where those tagged regions are is important enough to be included in the main paper, rather than the supplemental material.

Response:

We followed the suggestion to move the Figure S2, including the 27 tagged regions, from supplemental material into the main content. The caption in old Figure 1 (now Figure 2) has been changed to clarify that the source region partition for the CAMx PSAT simulation shown here is only for the 12-km inner modeling domain. The number of the figures in main document and supplemental material has changed accordingly.

p.14, line 9-10: It wasn't measured HNO_3 concentrations were overestimated by 108%. Modelled HNO_3 was overestimated.

Response:

Changed the sentence from "However, the model simulation underestimated available measured NH_3 concentrations by 65% on average, and measured HNO_3 concentrations were overestimated by 108%." to "However, the model simulation underestimated the measured NH_3 concentrations by 65% on average and overestimated the measured HNO_3 by 108%."

Fig 9: the Oil and Gas pattern is difficult to see in the legend – looks very similar to the Other pattern in the legend, and doesn't seem to be as dark as in the pies. In the pies, the Oil and Gas is (I think) the gray, but the legend looks much lighter. This doesn't seem to be a problem in Figs. 6 and 10 which has the same system.

Response:

We double-checked Figure 10 (previously Figure 9) and made sure the legend, color map setting, as well as notation are consistent with Figure 7 (previously Figure 6) and Figure 11 (previously Figure 10). The updated Figure 9 is attached here for reference.

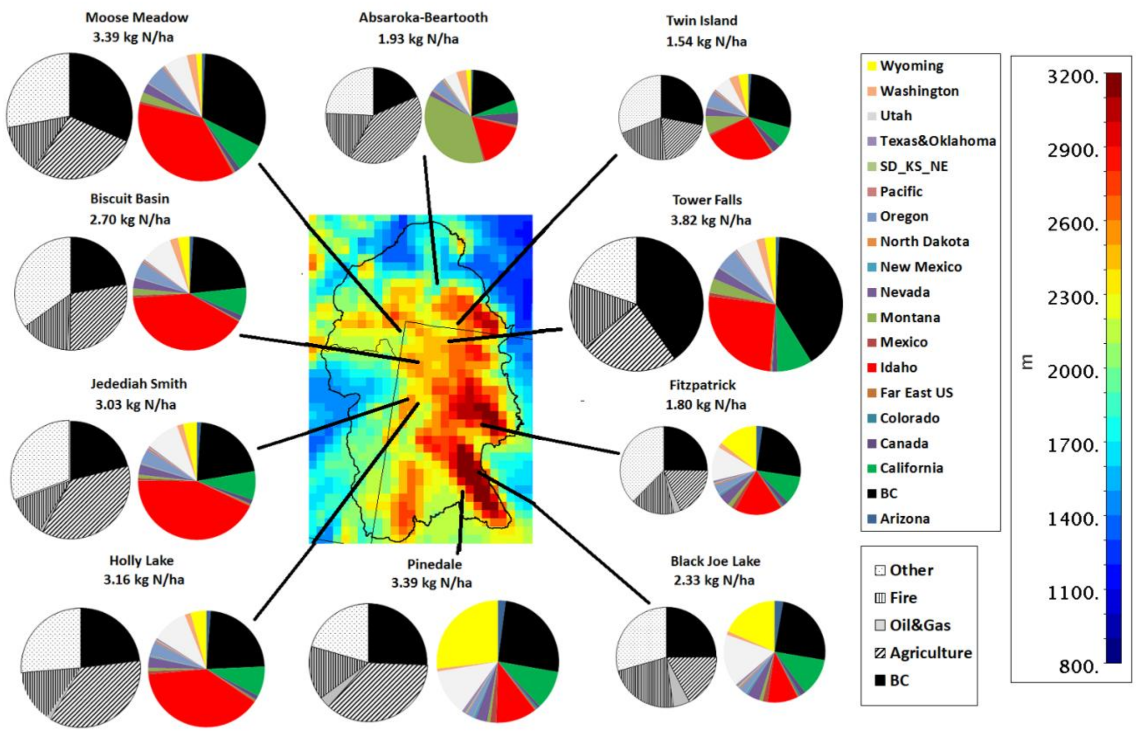

Fig 11: I think the legend at the bottom should be removed because seeing MOZART/IMRPOVE next to the red square with the line through it is confusing and doesn't really make sense. It's not needed since in the text we know that the BC came from MOZART, and from the caption we know that the simulation was sampled at IMPROVE sites.

<u>Response:</u>

We accepted this suggestion to revise the caption for Figure 12 (previously Figure 11) as: "Figure 12. Ratio of simulated versus measured particulate nitrate (PNO3) concentrations against the boundary contributions to simulated PNO3 at IMPROVE sites over a 12-km domain."

The revised figure attached below.