Comments from anonymous referee #2

General comments

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 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 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 at 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.

Response:

We appreciate the favorable overall sentiment and the opportunity to revise our manuscript in response to those comments. We have addressed each comment and suggestion as described below. Note that we do not know the uncertainties in the source attribution (SA) results, but suspect that they are large based on the model performance evaluation. This is why the results are discussed in more general and semi-quantitative terms in section 5. However, in response to the comment we have made a greater effort to convey the uncertainties and potential biases where appropriate. For example, in the abstract we included the sentences: "These uncertainties appear to result in an overestimation of distant source regions including California and BC and an underestimation of closer agricultural source regions including the Snake River valley. Due to these large uncertainties the relative contributions from the modelled sources and their general patterns are the most reliable results."

Also, the discussions on the change of deposition velocity of NH3 in CAMx to SA results showed that less than 10% change of the contributions for each source sectors/regions for the conducted 2 month sensitivity simulations (Figure 11). Also, the SA results due to different boundary conditions usage didn't change much (less than 10%, see Figure S8). The detailed comment below further address this issue.

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.

Response:

We agree with the reviewer and added the following sentences to the abstract:

"These uncertainties appear to result in an overestimation of distant source regions including California and BC and an underestimation of closer agricultural source regions including the Snake River valley. Due to these large uncertainties the relative contributions from the modelled sources and their general patterns are the most reliable results."

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).

Response:

The following sentence was added to the abstract: "The BC were outside the conterminous United States and thought to represent international anthropogenic and natural contributions."

1.26: I thought it 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".

Response:

We deleted this sentence as suggested.

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

Response:

Based on the suggestion, this sentence now read as:

"These compounds arise from a variety of sources, with inorganic oxidized N primarily emitted as nitrogen oxides (NOx) from fossil fuel combustion, with approximately 25% from power plants, 50% from automobiles, and 10% from other mobile sources on annual based county level estimation (EPA, 2015)."

2.20: Missing some references here, e.g. work from Zondlo's group.
Response:
We added two highly cited references from Zondlo's group regarding the on-road NH3 emissions (Sun et al., 2014; Sun et al., 2017). The sentence now read as
"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)."

References:

Sun, K., Tao, L., Miller, D.J., Khan, M.A. and Zondlo, M.A.: On-road ammonia emissions characterized by mobile, open-path measurements. Environ. Sci. Tech., 48(7), 3943-3950, 2014.

Sun, K., Tao, L., Miller, D.J., Pan, D., Golston, L.M., Zondlo, M.A., Griffin, R.J., Wallace, H.W., Leong, Y.J., Yang, M.M. and Zhang, Y. Vehicle emissions as an important urban ammonia source in the United States and China. Environ. Sci. Tech., 51(4), 2472-2481, 2017.

3.14: for zero-out –> using zero-out
Response:
Changed.

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?
Response:
Lee et al. (2016) used the adjoint of GOES-Chem to investigate the spatial and sectoral distribution of annual Nr deposition contributed by different sources. As expected, NH3 emissions from livestock and NOx emissions from mobile sources are the major contributors to Nr deposition in nearly all selected Class I areas in the United States. Nr deposition in the mountain regions in the western U.S (Grand Teton and Rocky Mountain NPs) are ~50% from nearby sources (<400 km) and the rest from sources as far away as California (~1300 km). To avoid the ambiguity, we rewrote this sentence as:

"Lee et al. (2016) used the adjoint version of GEOS-Chem to quantify the sources of Nr deposition in eight selected federal Class I areas in 2010 and found a nonnegligible footprint (>20%) of Nr deposition in western United States, including GTNP and Rocky Mountain National Park (RMNP), attributed to long-range transport from sources in California, especially during summer time."

Reference:

Lee, H. M., Paulot, F., Henze, D. K., Travis, K., Jacob, D. J., Pardo, L. H., and Schichtel, B. A.: Sources of nitrogen deposition in Federal Class I areas in the US, Atmos. Chem. Phys., 16(2), 2016.

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.
Response:
We significantly shortened this paragraph into one sentence and combined it with the previous paragraph to show the similarity of source apportionment modeling studies' focus on Rocky Mountain to the GYA area. Now the new sentence read as:
"Similar modeling studies focusing on RMNP also suggested the important contributions of distant sources including those from California and other counties and the fact that the contributions from source of reduced Nr were larger than those from sources of oxidized Nr (Thompson et al., 2015; Malm et al., 2016)."

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?
Response:
The mobile emissions we used in this modeling study were from the NEI 2011 inventory, which used MOVES2010 to generate emission inventories or emission rate lookup tables for on-road mobile sources (UNC-Chapel Hill and ENVIRON, 2014). We notice there are reports commenting that the NEI may overestimate the mobile NOx emission. For example, Anderson et al. (2014) estimated the NEI may overestimate mobile NOx emissions by 51–70%, based on the observed molar CO/NOx emission ratios from the DISCOVER-AQ campaign data. They argue that "the NEI overestimate of NOx emissions could indicate that engines produce less NOx and catalytic converters degrade more slowly than assumed by MOVES2010. MOVES2010 likely fails to capture dependence of NOx emissions on vehicle age accurately." We didn't explicitly explore the uncertainty of mobile NOx emission to the source apportionment results.

References:
UNC-Chapel Hill and ENVIRON International Corporation, Three-State Air Quality Modeling Study (3SAQS) – Final modeling protocol: 2011 emissions & air quality modeling platform,
http://vibe.cira.colostate.edu/wiki/Attachments/Modeling/3SAQS_2011_WRF_MPE_v8_draft_Aug04_2014.pdf
Anderson, D.C., Loughner, C.P., Diskin, G., Weinheimer, A., Canty, T.P., Salawitch, R.J., Worden, H.M., Fried, A., Mikoviny, T., Wisthaler, A. and Dickerson, R.R.,

Measured and modeled CO and NOy in DISCOVER-AQ: An evaluation of emissions and chemistry over the eastern US. Atmos. Environ., 96, 78-87, 2014.

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?

Response:

As mentioned in the previous response, the on-road mobile source is provided by MOVE2010, and it does account for the NH3 emissions from the mobile sources; see the attached picture below. However, these emissions are likely underestimated since recent work by Fenn et al., (2018), which was discussed in the manuscript, estimates that the 2011 NEI underestimates mobile NH3 emissions by a factor of 2.9.
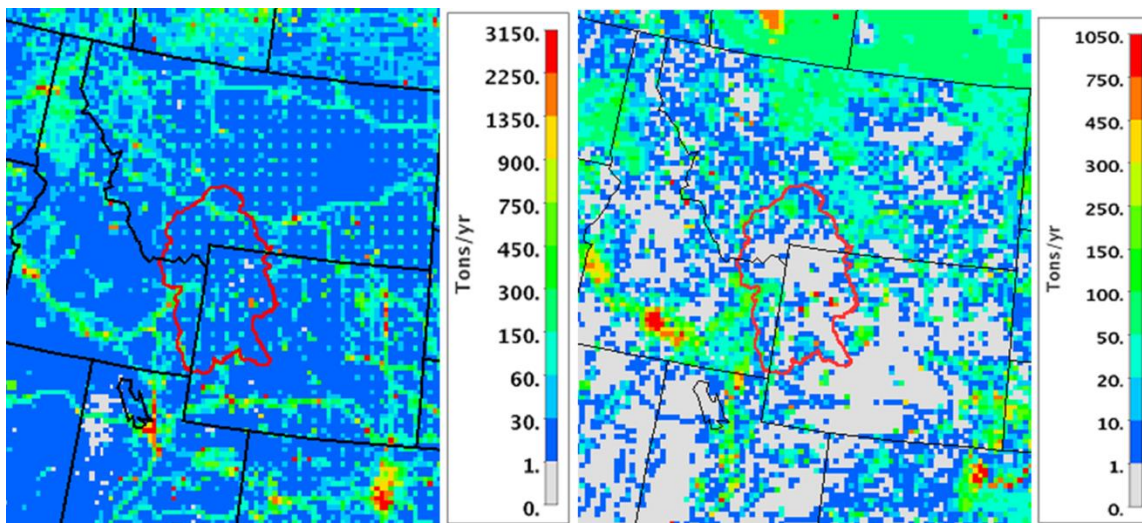
Reference:

Fenn, M.E., Bytnerowicz, A., Schilling, S.L., Vallano, D.M., Zavaleta, E.S., Weiss, S.B., Morozumi, C., Geiser, L.H. and Hanks, K.: On-road emissions of ammonia: An underappreciated source of atmospheric nitrogen deposition, Sci. Total Environ., 625, 909-919, 2018.

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).

Response:

For this work, we used the 2011 NEI version 2 inventory from the EPA and updated the oil and gas sector at western U.S. based on the local survey data. As requested, we

provided the designated table (Table S2) in the supplemental material to provide the summary of 27 tagged regions in CAMx PSAT in this study and annual emissions for NH3 and NOx. The table is attached for reference.

Table S2. Summary of 27 tagged regions in CAMx PSAT in this study and their corresponding annual emissions for NH3 and NOx with agriculture (AG), oil and gas OG), wildfires and prescribed fires (fire), and remaining emission source sectors (Other). The items in the parentheses are aggregate regions based on prevailing wind patterns over the GYA for the source apportionment results reported in Figures 9–11.

| Tagged region | Total emission for nitrogen species (tons/yr) | | | | | | | | | |
| | NH3 | | | | | NOx | | | | |
	AG	OG	Fire	Other	total	AG	OG	Fire	Other	total
NW Colorado (Southwest)	4,900	0	55	418	5,373	0	12,046	564	54,827	67,437
NE Colorado (Southwest)	37,041	0	415	3,157	40,613	0	16,002	749	72,830	89,581
SE Colorado (Southwest)	20,281	0	227	1,728	22,237	0	20,869	976	94,980	116,825
SW Colorado (Southwest)	6,672	0	75	569	7,315	0	5,504	258	25,051	30,812
Upper Green River, Wyoming	2,358	0	525	110	2,993	0	11,412	3,016	43,523	57,952
Jackson, Wyoming	2,375	0	529	111	3,015	0	477	126	1,817	2,420
Eastern Wyoming (Other WY)	7,298	0	1,625	342	9,265	0	3,013	796	11,490	15,299
Western Wyoming (Other WY)	18,046	0	4,018	845	22,910	0	10,925	2,887	41,662	55,474
Yellowstone (Other WY)	1,511	0	336	71	1,918	0	761	201	2,902	3,864
Northern Idaho (Northwest)	16,887	0	2,193	910	19,991	0	669	6,906	47,036	54,612
Snake River Valley, Idaho	43,696	0	5,674	2,356	51,726	0	682	7,030	47,882	55,594
Northern Utah	12,946	0	69	2,163	15,178	0	10,235	200	92,312	102,747
Southern Utah (Southwest)	10,083	0	54	1,685	11,822	0	8,907	174	80,338	89,419
Nevada	5,569	0	825	2,533	8,926	0	189	2,725	107,900	110,814
Montana	54,343	0	7,531	1,313	63,187	0	13,806	11,510	153,220	178,537
Washington (Northwest)	44,118	3	825	7,400	52,345	0	467	2,458	268,831	271,757
Oregon (Northwest)	43,626	0	8,858	5,164	57,649	0	925	28,231	146,062	175,218
California	203,204	155	3,056	111,240	317,655	0	8,806	9,457	669,421	687,684
Mexico (Non U.S.)					246,344					782,600
New Mexico (Southwest)	35,327	0	4,374	2,673	42,374	0	71,863	15,197	170,550	257,609
Arizona (Southwest)	33,247	0	9,041	8,520	50,808	0	1,489	26,817	250,201	278,506
Texas&Oklahoma (Southwest)	364,835	44	24,481	39,179	428,539	0	410,736	35,635	1,450,095	1,896,465
Canada (Non U.S.)					421,830					934,900
North Dakota (Eastern U.S. + Great Plains)	93,163	0	952	6,995	101,110	0	8,408	1,407	171,869	181,683
Pacific (Non U.S.)					292					251,698
Far East U.S. (Eastern U.S. + Great Plains)					2,627,200					9,296,000
SD_KS_NE (Eastern U.S. + Great Plains)	480,670	4	6,245	9,439	496,359	0	96,945	25,572	666,950	789,467
Total:					5,128,972					16,834,975

Also, we added a summary in the text about the emissions we used in this modeling study: "Table S2 provides the annual NH3 and NOx emissions used in this modeling study with a breakdown by tagged source regions and source sectors. Figure 2 provides the annual emissions of NH3 in the inner 12-km domain as well as the monitoring sites or receptor areas used for the model evaluation and analysis. For NH3 emissions, the AG sector contributed 84.1% of the total emissions within 12-km domain, while the OG, Fire, and Other sectors contributed 0.1%, 4.5%, and 11.4%, respectively (Table S2). In the Snake River valley, the AG sector emissions dominate the emission budget. For NOx emissions, the contribution rankings from the four tagged emission sources are Other (83.8%), OG (12.8%), Fire (3.2%), and AG (0%)."

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?

Response:

We are not completely clear as to exactly what the reviewer is asking in this question. However, CAMx contains relatively complete chemical and thermodynamic mechanisms for inorganic sulfur and nitrogen gases and particles. Therefore, the interplay between SO_2 - NO_x - NH_3 is accounted for in the model. For example, with the decreases in SO_2 emissions there should be more NH_3 available to neutralize HNO_3 forming particulate ammonium nitrate. The CAMx chemical and thermodynamic mechanism can account for these and other shifts and their impact on nitrogen deposition and be reflected in the PSAT source attribution results.

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

Response:

Questionable data refers to the measurements used to evaluate the model. There are certain protocols used by the measurement community to report their data and the associated credentials. For instance, for the wet deposition data reported by the NTN, a series of codes are assigned to samples that are considered invalid by the NTN for the purposes of computing weighted-mean concentrations, depositions, and data completeness estimates. The common reasons are contaminated samples, inadequate volume collected in the bucket for analysis, and lab error, for example. To make this statement clear, we changed the sentence from "All data flagged as questionable were removed from the analysis" to "All measurement data flagged as questionable, either due to maloperation or due to insufficient samples to calculate representative values, were excluded from the analysis. In Table 1, we also reported the percentage of validate measurements used for statistical analysis during evaluation time. For most of the nitrogen species, the percentage of validate samples are more than 80%."

We also added the percentages of measurement data completeness in the model performance evaluation table (Table 1) for reference.

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.

Response:

Thanks for pointing this out. The reference here is not proper. In GEOS-Chem, the inorganic chemistry mechanism used to model the pollutants' evolution from surface to

the stratopause is called the "tropchem" mechanism and is based on the NASA/JPL publication 10-6 for chemical kinetics and photochemical data for use in atmospheric studies. In total, 236 reactions were included in this mechanism, and reaction #225 has the parameterization of heterogeneous N_2O_5 reaction to form HNO_3 based on the ambient aerosol type, relative humidity, and temperature (Evans and Jacob, 2005). In CAMx, we used the CB6r2 mechanism, and it also includes consideration of this heterogeneous HNO_3 formation with the initial parameterization protocol as in Evans and Jacob (2005) but with revisions (Foley et al., 2010). However, since GEOS-Chem is a global photochemical model and the "tropchem" is different from a carbon bond mechanism, it is unfair to quote the evaluation statements regarding GEOS-Chem to the CAMx simulation results here. Therefore, we deleted this statement. Instead, we added two additional citations for reporting the same HNO_3 overestimation problem using regional air quality models (e.g., CMAQ, CAMx). Now this sentence read as:

"The overestimation of HNO_3 has also been reported in other regional-scale modeling simulations over the United States (e.g., Barker and Scheff. 2007, Foley et al., 2010; Thompson et al., 2015) with the carbon bond mechanism used in this study. The possible reason for the overestimation of HNO_3 may be due to the uncertainty for the N_2O_5 uptake coefficient setting for heterogeneous reactions (Foley et al., 2010)."

References:
    Baker, K. and Scheff, P.: Photochemical model performance for PM2. 5 sulfate, nitrate, ammonium, and precursor species SO2, HNO3, and NH3 at background monitor locations in the central and eastern United States, Atmos. Environ., 41, 6185-6195, 2007.
    Foley, K.M., Roselle, S.J., Appel, K.W., Bhave, P.V., Pleim, J.E., Otte, T.L., Mathur, R., Sarwar, G., Young, J.O., Gilliam, R.C. and Nolte, C.G., Incremental testing of the Community Multiscale Air Quality (CMAQ) modeling system version 4.7. Geosci. Model Dev., 3(1), 205-226, 2010.
    Evans, M.J. and Jacob, D.J., 2005. Impact of new laboratory studies of N2O5 hydrolysis on global model budgets of tropospheric nitrogen oxides, ozone, and OH. Geophysical Research Letters, 32(9).

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?
Response:
Currently, there is no bidirectional flux model for NH3 implemented in CAMx. The bidirectional flux model calculates the compensation point of NH3 between canopy and land-surface terrain and allows a portion of deposited NH3 to be emitted back into the atmosphere based on the emission potential of the soil NH3 pool. Conceptually, given the occurrence of re-emittance of certain amounts of NH3 into the atmosphere, the NH3

ground concentrations at the surrounding modeling grids (especially downwind grids) should be increased. The GYA area is adjacent and downwind of the Snake River valley and northern Utah, both of which have significant portions of agricultural sources (see Table S2). Therefore, it is a logical expectation that if the bidirectional NH3 model was implemented in CAMx, the bias in the simulated NH3 concentrations in this region would be decreased. Furthermore, in section 5, we discussed the potential benefit of including NH3 bidirectional parameterization into the CAMx model and the difficulties for implementation. To specifically address the reviewer's comment, we added the following statement:

"The poor NH3 results may be related to the high uncertainty in the NH3 emission inventory (Clarisse et al., 2009) and important missing physical mechanisms in the model, including the lack of bidirectional NH3 deposition (Zhang et al., 2010; Bash et al., 2013; Zhu et al., 2015). The GYA area is located downwind of the major agriculture sources in the Snake River valley and northern Utah (Table S2). The incorporation of the bidirectional NH3 flux mechanism in the model should increase ambient NH3 concentrations in the GYA and thus decrease the large model underestimation of NH3 concentrations."

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)?
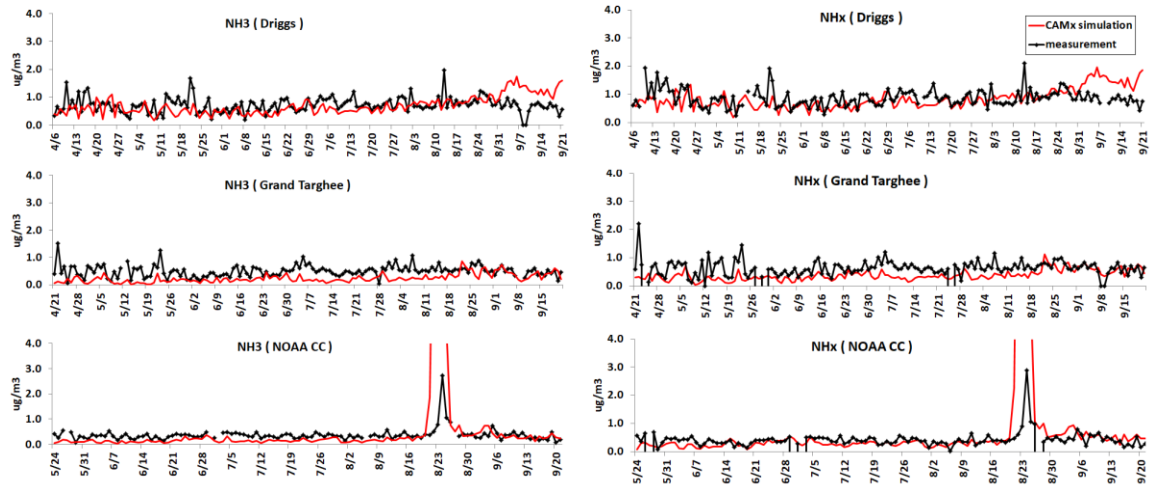
Response:

It is possible that the poor model performance for NH3 may relate to the overestimation of HNO3 in the model, which would push excessive partitioning of NH3 into the particle phase. CAMx uses ISORROPIA to calculate the inorganic gas–particle thermodynamic equilibrium. From the old Table 1, we also see a slight overestimation of PNH4 in conjunction with the large underestimation of NH3 at CASTNET sites within the GYA. Therefore, we followed the suggestion of the reviewer to evaluate NHx to try to get around the possible bias in gas-particle partitioning. However, only a few locations existed within the GYA where a network has concurrent measurements of nitrogen gas and particulate species. We added the statistics for NH3, PNH4, and NHx model performance during the GrandTReNDS campaign at the three sites in the updated Table 1 (attached below).

Table 1. CAMx model performance for nitrogen species concentrations as well as nitrogen dry/wet depositions evaluated at sites in AMoN, CASTNet, IMPROVE, NTN

networks as well as the 3 sites during GrandTRENDS campaign over the GYA region (see Figure 1 for site locations) in 2011.

Species		Network	Duration	OBS[a]	SIM[b]	#Site[c]	N[d] (% completeness)	R[e]	NMB[f]	NME[g]	FB[h]	FE[i]
concentration	NH₃ (ppb)	AMoN[1]	Sep 22-Dec 12	0.49	0.30	1	7 (100%)	0.20	-65%	67%	-52%	53%
		GrandTReNDS[2]	Apr 5-Sep 21	0.55	0.46	3	434(97.7%)	0.30	-16%	57%	-42%	63%
	HNO₃ (ppb)	CASTNet[3]	Jan 4-Dec 27	0.23	0.47	2	83(98.8%)	0.72	108%	117%	60%	71%
		GrandTReNDS[2]	Apr 5-Sep 21	0.28	0.54	3	435(97.9%)	0.60	106%	109%	63%	68%
	PNO₃ (µg m⁻³)	CASTNet[3]	Jan 4-Dec 27	0.19	0.25	2	83(98.8%)	0.42	37%	76%	26%	64%
		IMPROVE[4]	Jan 3-Dec 29	0.14	0.22	4	332(68.5%)	0.35	58%	108%	51%	80%
		GrandTReNDS[2]	Apr 5-Sep 21	0.13	0.15	3	435(97.9%)	0.45	15%	71%	14%	60%
	PNH₄ (µg m⁻³)	CASTNet[3]	Jan 4-Dec 27	0.17	0.18	2	83(98.8%)	0.28	3%	39%	7%	41%
		GrandTReNDS[2]	Apr 5-Sep 21	0.14	0.17	3	433(97.7%)	0.12	23%	64%	34%	61%
	NHₓ (µg m⁻³)[4]	GrandTReNDS[2]	Apr 5-Sep 21	0.68	0.63	3	427(96.2%)	0.26	-7%	48%	-22%	46%
N Deposition	HNO₃ dry (kg N ha⁻¹)	CASTNet[3]	Jan 4-Dec 27	0.071	0.187	2	83(98.8%)	0.81	153%	156%	77%	82%
		GrandTReNDS[2]	Apr 5-Sep 21	0.016	0.049	3	435(97.9%)	0.66	204%	209%	101%	104%
	PNO3 dry (kg N ha⁻¹)	CASTNet[3]	Jan 4-Dec 27	0.012	0.023	2	83(98.8%)	0.14	96%	148%	48%	97%
		GrandTReNDS[2]	Apr 5-Sep 21	0.010	0.011	3	435(97.9%)	0.61	8%	58%	1%	65%
	PNH₄ dry (kg N ha⁻¹)	CASTNet[3]	Jan 4-Dec 27	0.018	0.019	2	83(98.8%)	0.1	7%	57%	22%	61%
		GrandTReNDS[2]	Apr 5-Sep 21	0.006	0.004	3	433(97.7%)	0.1	-33%	46%	-28%	53%
	NO3⁻ wet (kg N ha⁻¹)	NTN[5]	Jan 4-Dec 27	0.079	0.097	5	214(82.3%)	0.34	31%	126%	12%	100%
		GrandTReNDS[2]	Apr 5-Sep 21	0.051	0.083	3	427(96.2%)	0.15	60%	94%	42%	71%
	NH₄⁺ wet (kg N ha⁻¹)	NTN[5]	Jan 4-Dec 27	0.088	0.126	5	214(82.3%)	0.32	49%	142%	19%	106%
		GrandTReNDS[2]	Apr 5-Sep 21	0.103	0.147	3	427(96.2%)	0.48	42%	72%	30%	64%
	Precipitation (cm)	NTN[5]	Jan 4-Dec 27	0.77	2.34	5	214(82.3%)	0.54	215%	242%	64%	118%
		GrandTReNDS[2]	Apr 5-Sep 21	0.33	0.95	3	427(96.2%)	0.42	187%	207%	69%	94%

The time series plots with the daily mean concentration comparisons are also given below. The CAMx model still underestimates the NH3 concentration (NMB = -16%) and overestimates PNH4 concentration (NMB =23%) at the three sites, but if we evaluate NHx, the model bias is smaller (NMB = -7%).

Also, we added a sentence in the first paragraph of section 3.2 as:
"The underestimation of NH3 concentration still existed (NMB = -16%), and one of the possible reasons may be due to the overestimation of HNO3 in the model pushing excessive partitioning of NH3 into the particle phase, which can be shown by the better model performance for NHx simulation (NMB = -7%) without splitting the gas-particle partition bias."

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.

Response:

The performance metrics referenced here from Simon et al. (2012) are the compilation of 69 peer-reviewed articles published between 2006 and 2012 focusing on regional air quality model performance evaluation for total PM2.5, speciated PM2.5, and wet deposition of sulfate, nitrate, and ammonium over the United States and Canada. None of the simulations compiled by the authors focus on the N_r source attribution. Reviewer #1 also has suggestions on this sentence. In here we just want to demonstrate that our CAMx base case modeling performance is in line with the peer modeling results and provides a good platform for further source attribution analysis. We provided Table S3 in the supplemental material to summarize the collected recent model performance evaluations for nitrogen species and revised this sentence to:

"Table S3 provides a comparison of regional CTM performance evaluations against measured N- containing species 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). The model performance results in this study are comparable to these past studies including the overestimation of HNO3 and underestimation of NH3. Resolution of these biases requires additional research and these biases need to be taken into account when interpreting the source attribution of Nr deposition within the GYA."
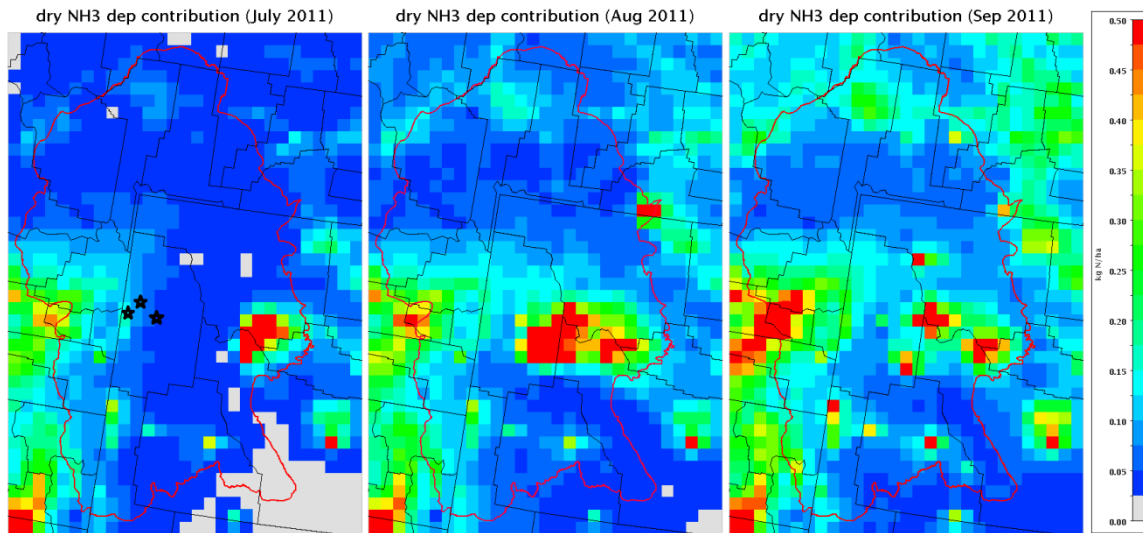
Fig 3: I find it interesting that the measurements at each site show a distinct reduction in NH3 dry dep in September, whereas CAMx shows a maximum in September for Driggs and Grand Targhee. Can authors comment on this?
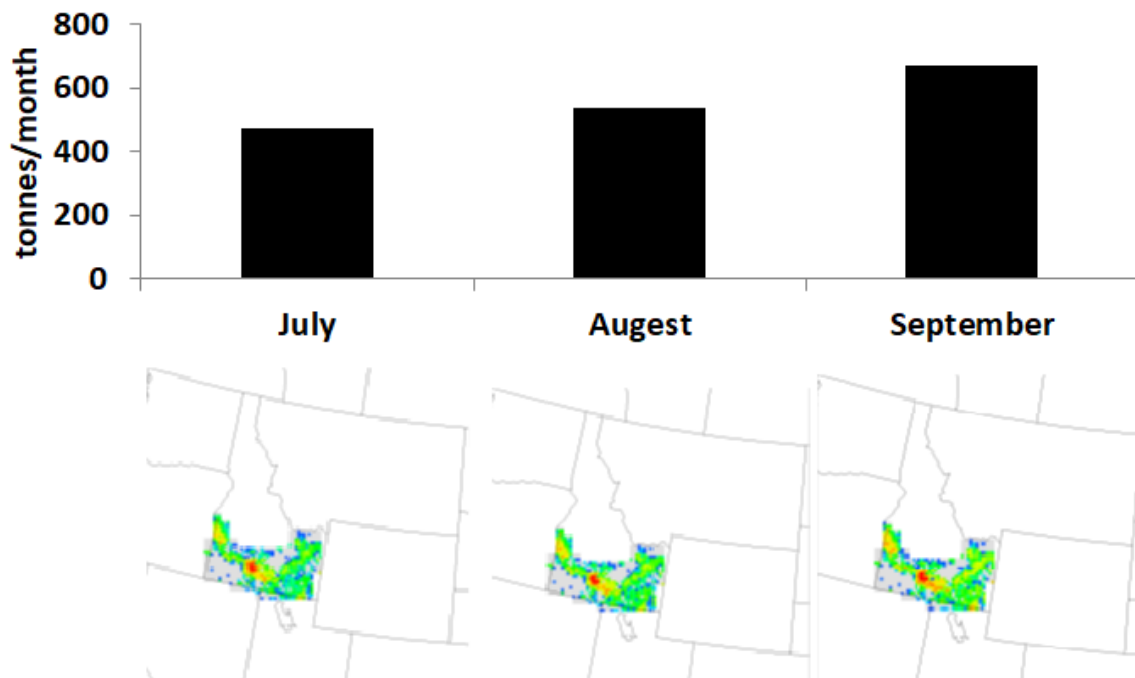
Response:

The monthly dry NH3 deposition values at the three sites associated with Figure 3 (now Figure 4) are attached below as a Table for clarification. It is true that the NH3 dry deposition (light blue in the figure) in September at each site shows a distinct reduction compared with the previous month (0.094 versus 0.209 in Driggs, 0.074 versus 0.147 in Grand Targhee, and 0.049 versus 0.113 in NOAA), but the corresponding CAMx results have the opposite trend for the Driggs and Grand Targhee sites.

		GrandTReNDS	CAMx
		(kg N/ha)	(kg N/ha)
Driggs	Apr	0.114	0.142
	May	0.158	0.104
	Jun	0.156	0.104
	Jul	0.194	0.101
	Aug	0.209	0.134
	Sep	0.094	0.194
Grand Targhee	Jul	0.018	0.071
	Aug	0.147	0.101
	Sep	0.074	0.119
NOAA CC	May	0.018	0.043
	Jun	0.076	0.050
	Jul	0.085	0.049
	Aug	0.113	0.102
	Sep	0.049	0.088

Back trajectory analysis shows that during the GrandTReNDS campaign period, the dominant source origins impacting the Nr in the GYA are from Snake River valley and northern Utah (Prenni et al., 2015). The high NH3 deposition at the three sites in September in the CAMx simulation results is also verified with the spatial plots attached below. The high deposition is associated with the high NH3 emission rates in September from the Snake River valley.

dry NH3 dep contribution (July 2011)   dry NH3 dep contribution (Aug 2011)   dry NH3 dep contribution (Sep 2011)

More importantly, if we compare the monthly mean dry deposition velocities used to calculate the measured NH3 dry deposition with the corresponding CAMx values, we find that there is a steep jump from August to September from the GrandTReNDS calculations, while the deposition velocity values from the models keep steady. Therefore, we believe this discrepancy is mainly due to the different variation trend of dry deposition velocity between the measurements and the model.
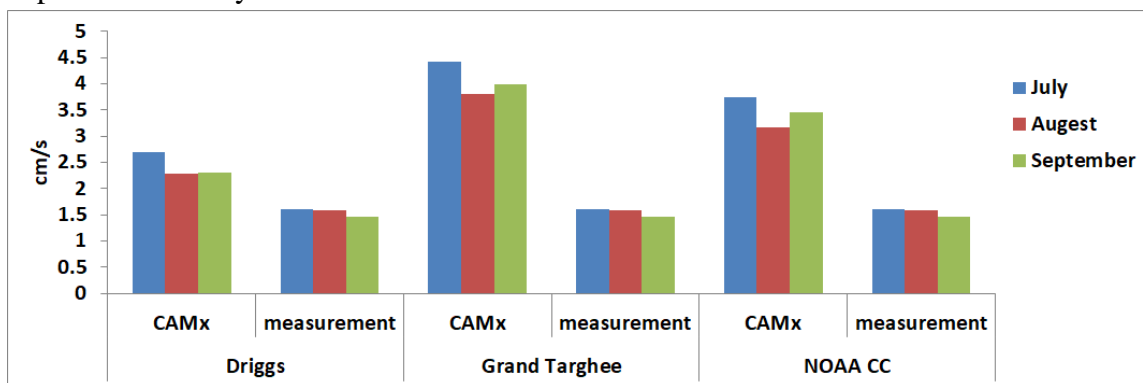
We revised the corresponding sentences in section 3.2 as:

"As shown, the simulation does a poor job of reproducing the total Nr deposition rates both in the month-to-month variation as well as across the sites. The difference in the dry NH3 deposition monthly variation between measurements and simulation is mainly due to the difference in associated dry deposition velocity used for calculation. However, consistent with the observations, the simulation shows that wet deposition is larger than dry and that the contribution from reduced N deposition was larger than from the

oxidized N deposition at all three sites, although the observed range of 70–80% reduced N was more than the 55–68% simulated in CAMx."

Reference:

Prenni, A.J., Levin, E.J.T., Benedict, K.B., Sullivan, A.P., Schurman, M.I., Gebhart, K.A., Day, D.E., Carrico, C.M., Malm, W.C., Schichtel, B.A. and Collett, J.L., Gas-phase reactive nitrogen near Grand Teton National Park: Impacts of transport, anthropogenic emissions, and biomass burning. Atmos. Environ., 89, 749-756, 2014.

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?

Response:

In the supplemental material, Figure S6, we updated the change of spatial patterns of the simulated total NH3 deposition over the GYA during July–August 2011 due to the change of NH3 deposition velocity in CAMx (the middle panel in the attached figure below).
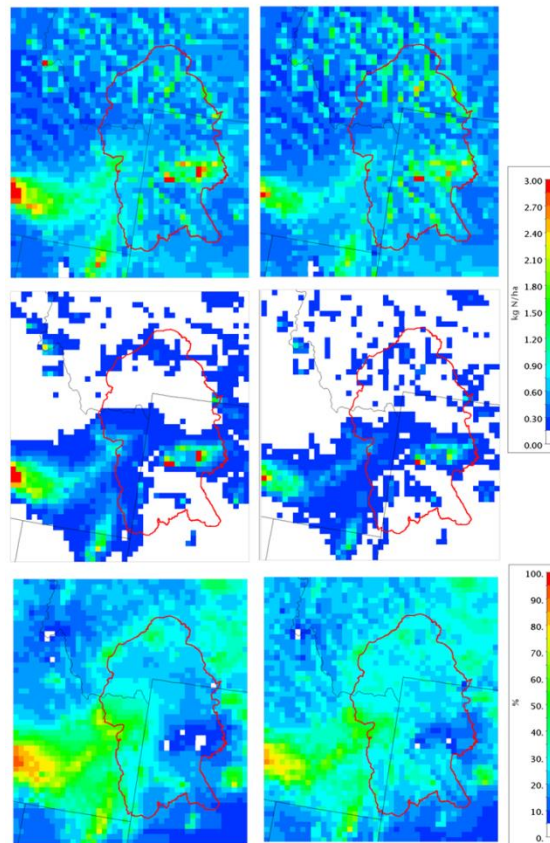

Figure S6. Change of spatial patterns of the simulated total Nr deposition (top panel), total NH3 deposition (middle panel) as well as contributions from agricultural emissions

sector to total Nr deposition budget (bottom panel) over the Greater Yellowstone Area (GYA) during July–August 2011 due to the change of NH3 deposition velocity in CAMx.

Attached table shows the dry and wet nitrogen deposition change at the GYA due to changing NH3 deposition velocity in CAMx during July-August 2011. Decreasing the NH3 deposition velocity will increase the NH3 surface concentration and improve the model bias for underestimation (see Figure S5). Still, the total NH3 dry deposition in the GYA will decrease by 3%. However, the NH3 wet deposition in the GYA is significantly increased (73%) due to longer NH3 lifetime since emit and further deposition into the GYA during precipitation events. On average, a 31% increase for total Nr deposition from the agriculture source sector (which is dominated by NH3 emissions) can be seen by decreasing the NH3 dry deposition velocity.

	base (kg N/ha)			DV_0.1 (kg N/ha)			difference(%)		
	Dry	Wet	Total	Dry	Wet	Total	Dry	Wet	Total
BC	0.033	0.040	0.073	0.029	0.045	0.074	-12.5%	14.3%	2.1%
Agriculture	0.038	0.030	0.069	0.037	0.052	0.090	-2.7%	73.1%	30.8%
Oil&Gas	0.004	0.001	0.005	0.004	0.002	0.005	-1.2%	14.1%	3.2%
Other+Fire	0.149	0.056	0.206	0.130	0.070	0.200	-13.2%	25.6%	-2.7%
Total	0.224	0.128	0.352	0.199	0.170	0.369	-11.2%	33.2%	4.9%

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

Response:

Due to the limited amount of computational resources, we didn't conduct the HNO3 sensitivity study or quantify its impact to source apportionment results. It is true that the overestimation of HNO3 concentration is a major uncertainty for the simulated nitrogen deposition budgets (see Figure 3 and Figure 4). Heald et al. (2012) used GOES-Chem to simulate inorganic aerosol loading and NH3 concentrations over the United States. They also reported significant overestimation of HNO3 concentrations and found that by reducing HNO3 concentrations to 75% of their simulated values, the model can correct the bias in nitrate as well as in ammonium simulation. They didn't pinpoint the mechanism underneath this model performance improvement but provided a general statement that it may be due to "a combination of errors in chemistry, deposition and sub-grid near-surface gradients." However, the findings from Heald et al. (2012) using GEOS-Chem are hard to refer here to justify the similar impact from CAMx given the differences of those two photochemical models in terms of implementation scales (regional versus global) and chemical mechanism (carbon bond versus tropchem). We expect the decrease of deposition of oxidized nitrogen in the GYA by decreasing the HNO3 concentrations in the model and we suspect the impact from further source regions with high NOx emissions will become smaller to the GYA.

We added a sentence at the section 5 as:

"The overestimation of HNO3 concentrations in the GYA is another reason for the wet Nr deposition overestimation. However, its impact on source apportionment results was not conducted here due to unclear reasons for the model bias (emission, chemistry, meteorology, deposition scheme) and limited computational resources."

Reference:

Heald, C.L., Collett Jr, J.L., Lee, T., Benedict, K.B., Schwandner, F.M., Li, Y., Clarisse, L., Hurtmans, D.R., Van Damme, M., Clerbaux, C. and Coheur, P.F., Atmospheric ammonia and particulate inorganic nitrogen over the United States. Atmos. Chem. Phys., 12(21), 10295-10312, doi:10.5194/acp-12-10295-2012, 2012.