

RESPONSE TO REVIEWS

ACPD article "Atmospheric ammonia and particulate inorganic nitrogen over the United States"

We thank both reviewers for their comments and suggestions. Responses are noted below with original reviewer text given in italics/blue. A "track changes" version will also be submitted to the editor. We also note an additional change to the manuscript. Since original submission, we learned that the PBL heights in the GEOS-5 product are artificially low at night which can lead to biased nighttime PM concentrations in the surface layer. This impacts comparisons with 24-hr average surface data presented in the manuscript, although not the IASI comparisons (daytime retrievals only). A fix for this PBL error was proposed by Sajeev Philips and Randall Martin (Dalhousie University) and all simulations were repeated with this fix. This update does not modify the conclusions of the study, however the specific values reported in the text and plots do differ slightly and have been updated in the final version submitted to ACP for publication (along with a description of the fix). We have added Philips and Martin as co-authors for their contributions in resolving this issue.

Response to Referee #1

The manuscript, 'Atmospheric ammonia and particulate inorganic nitrogen over the United States' by Heald et al. uses observations from ground sites and satellites along with a 3-D chemical transport model to study inorganic aerosol levels and ammonia concentrations in the United States. Sulfate simulations were in good agreement with observations while nitrate simulations had a positive bias throughout most of the US except in California where nitrate was underestimated. Comparisons of ammonia simulations with satellite observations suggest that current ammonia emissions are likely underestimated in the Midwest and California. The manuscript is well, written, clearly structured, and precise. It is of interest to ACP readers. I recommend its publication and only have a few minor corrections/clarifications and suggestions that are listed below.

The manuscript discusses a 'New NH₃ Seasonality' regarding NH₃ emissions and points out the influence of agricultural practices (page 19467, line 25). In addition to seasonality, e.g. when fields are fertilized, economic factors, i.e. type of fertilizer used, influence agricultural practices and, in turn, emissions. Livestock, in particular cattle, are a large source of NH₃ to the atmosphere. The review by Hristov, et al., 2011 and work by Liu et al., 2012, for example, show that diet composition, such as, percentage of crude protein in cattle feed, affects NH₃ emissions. Market factors (e.g., feed cost versus sought yield) and regional availability largely determine feed composition. Though difficult to assess (and alluded to here page 19486, lines 16-19), these issues will also have to be taken into account when analyzing seasonal and yearly variability of long-term regional data sets such as those from satellites. Hristov, et al., 2011, Review: Ammonia emissions from dairy farms and beef feedlots, Can. J. Anim. Sci., 91: 1-35 Liu, et al., 2012, Gas Emissions from Dairy Cows Fed Typical Diets of Midwest, South, and West Regions of the United States, J. Environ. Qual. 41 doi:10.2134/jeq2011.0435

The reviewer makes an excellent point that there are other factors (not limited to the one example given) that could contribute to different ammonia emissions seasonality. We have expanded the text to also include the cattle feed composition reference mentioned by the reviewer.

Page 19468, line 21. Can a reference be provided for the cattle inventory in Weld County?

We could only find grey literature references to Weld County being the largest cattle inventory in the US, thus we have modified our text to read: “one of the largest inventory of cattle in the United States (Weld County).”

Page 19473, lines 1-15. The authors note that time-resolved gas-particle vertical profile measurements through the boundary layer are required to investigate vertical gradients in NH₄NO₃ formation. Such observations have been discussed from a previous aircraft study by Neuman, J. A., et al. (2003), Variability in ammonium nitrate formation and nitric acid depletion with altitude and location over California, J. Geophys. Res., 108, 4557, doi:10.1029/2003JD003616. Section 3 does not describe the vertical spacing of the model so it is not clear if the study describe in this paper is applicable.

We are aware of the Neuman et al. study, however the aircraft in that study covered an extensive horizontal range and flew regularly up to 8km. While they did investigate ammonium nitrate formation in plumes, they do not provide a systematic investigation of ammonium nitrate formation within the boundary layer at a fixed location, as we are suggesting is required to investigate vertical sub-grid processes near the surface.

Figure 2 & 3. The superscript on sulfate is cut off and mostly unreadable. The unit label for the color scale is ambiguous. Adding a unit label (as in Fig. 6) to the right of each, e.g. after 8.0 and 4.0, makes it clear. I also suggest adding a dashed line or a hash mark on the x-axis of the maps as a reference point for 100 W related to the scatter plots for those not familiar with the United States geography.

Figure 2 and 3: The unit labels apply to both panels. Repeating these would limit the space occupied by the colour bar and thus reduce legibility of the figure. We have added gridlines which are annotated in Figure 2 to help define the 100W longitude as suggested.

Figure 4. There is no unit label on the color scale for the first column of plots. For clarity, I suggest adding ‘# retrievals’ on the color scale even if it is repeating the column label. Also, similar to Figs. 2 and 3, I suggest repeating the color scale unit label for the column and column difference color scales for clarity.

Figure 4: No unit is required for the first column (it is ‘number’) and as noted by the reviewer this information is given in the plot label. To eliminate confusion with the units, labeling has now been placed below the colour bars for this figure.

Figure 5. The format used for mass concentrations on the y-axis here is different than used in previous figures, i.e. no negative superscript. Here, and in Fig. 9, error bars present the standard deviation of the

monthly average observation. Assuming that the emissions and meteorology are correct, is there an estimate or sense of range for uncertainty in the chemical reactions and partitioning in the model?

Figure 8. Change in mass concentration format, as in Fig. 5.

Figure 9. Change in mass concentration format, as in Fig. 5.

Figure 10. Change in mass concentration format, as in Fig. 5.

Figure 5, 8, 9, 10: unit corrected.

Figure 5 and 9: Estimating the error associated with the uncertainty in chemical reactions and partitioning is a significant scientific endeavor and is beyond the scope of this work. There is a study in preparation: Ridley et al. "Using a Monte-Carlo approach to determine the critical reaction rates and initial conditions driving uncertainty in trans-Atlantic pollution plume modeling" that investigates how the uncertainties with reaction rates impacts transpacific transport of pollution. I am not aware of any similar study for inorganic aerosol formation.

Figure 6. Defining the four sensitivity simulations in the figure caption makes it easier for the reader to go between the text and the figure.

Figure 6: Caption has been expanded as suggested.

Figure 7. Since the second column is showing the updated GEOS-Chem simulation, I suggest adding '(updated simulation)' under the GEOS-Chem heading as done in Fig. 4 for GEOS-Chem (retrieved).

Figure 7: Label has been modified as suggested.

Figure 11. It is hard to see the symbols representing the sites on the figure. One suggestion is to make the outline of the circles thicker. Another suggestion is to list the sites according to the corresponding season in the caption. Also, it would be helpful to state that no SO₂ was measured at Big Bend N.P. in the caption. I found it confusing that Big Bend was missing in some panels and then had to go back to section 2.2 to found out why.

Figure 11: Symbols have been modified as suggested, and text on Big Bend added to caption.

Response to Referee #2

The manuscript entitled "Atmospheric ammonia and particulate inorganic nitrogen over the United States" examined atmospheric ammonia and particulate nitrate formation over the US using surface and satellite measurements interpreted with the GEOSChem model of atmospheric chemistry. Satellite measurements of ammonia column and in situ aerosol measurements were used to evaluate the model performance. It showed ammonia emissions in the model were biased low over California and the Midwest in spring. It also showed significant model overestimates of nitrate concentrations over the northeast and Midwest. Sensitivity simulations were conducted to test possible causes for the nitrate overestimates. It further examined the seasonal and spatial variability of gas-particle partitioning over the US. The study is comprehensively performed, and the manuscript is well written. I recommend publish on ACP after the following minor comments been addressed.

1. Page 19462, Line 10: Can you include here the typical values of DOF for the ammonia retrievals?

We have added the following to the text: "(typically < 0.3 over the continental United States)"

2. Page 19463, Line 19-20: Are the emissions in the model fixed for the three-year simulation?

No they are not. We have added text to clarify: "Anthropogenic emissions are scaled to the model year following national energy use statistics as described by van Donkelaar et al. (2008)."

3. Page 19465, Line 13-15: To my understanding, the regression slope calculated from the reduced-major-axis regression fit does not reflect the overall model bias. The value of the slope equals the ratio of standard deviations of the two variables, so it does not reflect the model bias. I suggest also report the normalized mean difference.

The slope of the RMA line is the slope of an ordinary least squares regression divided by the correlation (and thus identical at high correlation). However, given some of the low correlation values in winter/fall, we agree that additional statistics are helpful and have provided normalized mean bias values.

4. Page 19468, Line 6: Suggest change "retrieved concentrations" to "retrieved model concentrations" to avoid confusion with satellite retrieved values.

We are in fact referring to the satellite retrievals (which are underestimated) in this sentence. To clarify we have modified to "IASI concentrations".

5. Page 19471, Line 27: Does the artificially decrease in nitric acid concentrations only go to change the gas-aerosol thermodynamic partitioning? Would it change other gas phase chemistry in the model? Please clarify.

Yes, this change only impacts the gas-aerosol thermodynamic partitioning. We have added this text to clarify: "(as an input to the thermodynamic gas-particle partitioning only)".

6. Page 19472, Line 15: Should "Fig.3" be "Fig. 5" that shows ammonia concentrations were biased low in winter?

Yes, corrected in text.

7. Page 19488, Fig. 4: Please state in the figure caption that this GEOS-Chem simulation is the baseline simulation. Is it possible to use a different color (e.g. grey) to show areas with number of observations in gridcells smaller than 4, so that in the 5th column white color only shows areas with small differences.

We have added the qualifier "baseline" to the caption text describing GEOS-Chem. We attempted to modify the figure as suggested by the reviewer, but found that it reduced legibility of the figure (given the multiple panels and small gridboxes). Given that most of the white regions (representing less than 4 retrievals in a grid box) in the previous columns are over ocean, we think that the reader can distinguish these two conditions in the figure as provided.

8. Page 19491, Fig. 5: Missing a sentence ": :.(solid)..." in the figure caption.

We are not sure what the reviewer is indicating with this comment. The caption appears to be complete in the ACPD version online. No change was made.