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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Agricultural ammonia emissions in China: reconciling bottom-up and top-down estimates" *by* Lin Zhang et al.

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Comment: This manuscript first derives top-down estimation of growing season NH3 emissions in China using TES satellite NH3 retrievals and GEOS-Chem adjoint model. Based on published methodology, it then develops an improved bottom-up NH3 inventory from fertilizer application and animal wastes in China. It finally applies both the top-down and bottom-up NH3 inventories in the GEOS-Chem forward model to compare with in situ surface measurements of ammonia and ammonium wet deposition, showing that both the inventories improve the model performance. The manuscript is well motived, scientifically sound, and well written. I recommend publication after the following comments are addressed.
Response: We thank the reviewer for the valuable comments. All of them have been

addressed in the revised manuscript. Please see our itemized responses below.

Comment: First, there is a lack of detailed comparison between the top-down inventory and the bottom-up inventory developed by the authors (e.g. spatial, seasonal), as well as a lack of discussion if the improved bottom-up inventory would match better with the TES retrieval. They showed that both inventories improved model simulation of surface wet deposition fluxes of ammonium, but this is indirect evidence and hard to interpret with regards to the emissions effect.
Response: Thank you for the suggestion. We have compared model results using our bottom-up emission inventory with TES retrieved NH3 columns. We now state in Sect. 5.3: "this strong seasonality is consistent with the adjoint optimized emission totals for March–October (50% higher in summer than spring and fall) considering uncertainties in the inversion results and satellite retrievals. The improved bottom-up Chinese NH3 emissions are 15% higher than the top-down estimates in May and June, and 20% lower in other months. This can also be seen from the comparison of simulated NH3 columns using the improved bottom-up inventory with the TES measurements (Supplemental Fig. S1)". The revised manuscript also includes more information on comparison of our bottom-up emission inventory with previous estimates for the spatial and seasonal variations as described below.

Comment: Second, it would help future studies if the bottom-up inventory developed by this study can be compared more quantitatively with the existing ones analyzed in the manuscript. A good place would be to plot that inventory in Figure 1 in comparison with the other ones displayed in the Figure.
Response: Thank you for the suggestion. We have now plotted our bottom-up NH3 emissions in Figure 1 for comparing with previous emission estimates. We add the following text in Sect. 5.3: "The spatial distribution and seasonal variations of our bottom-up NH3 emission inventory are also presented in Figure 1 for comparison with previous estimates. We can see that our bottom-up estimates show similar

spatial features compared with Huang et al. (2012) and REAS v2, but also with some differences regionally. The total anthropogenic emission estimate of 11.7 Tg a$-$1 is in the middle of previous bottom-up estimates as summarized in Table 1, however, our NH3 emissions show much more distinct seasonal variations than previous estimates (e.g., Streets et al. (2003)) with emissions a factor of 3 higher in summer than winter."

Comment: Third, on line 109 they stated that the difference in bottom-up inventories is due to different base year, but in later places they stated that satellite data do not show a large trend of NH3 emissions in China and their model simulation was for the year of 2008 only, in spite of the use of multi years of TES observations. So my question is whether emissions would differ significantly by year, and if so, it would improve the scope of the manuscript if discussion could be added on the representativeness of year 2008 emissions they developed as the bottom-up inventory for other years, as well as offering suggestions on how scaling factors can be applied if their inventory is applied to other years.
Response: We stated on line 109 that "The factor of 2 difference is NOT likely due to the different base years". We also state "Analyses of historical NH3 emissions in China show relatively stable or weak increasing trends (less than 3% per year) since 2000 (Xu et al., 2016; Kang et al., 2016), consistent with trends in atmospheric NH3 concentration observed from satellites (Warner et al., 2017; Fu et al. 2017)". Thus we think that 2008 can be a representative year for the TES observational constraints. We are extending our bottom-up NH3 emission inventory to other years and planning to report it in a separated study.

Comment: Finally a technical issue about the GEOS-Chem model. It states that the model uses RPMARES as its thermodynamic module (line 172). I think the GEOS-Chem standard version uses ISORROPIA II thermodynamic equilibrium model. Is there a particular reason why the standard model setting is not used?
Response: We now state here: "GEOS-Chem also includes the ISORROPIA II ther-

modynamic equilibrium model (Fountoukis and Nenes 2007). We find that differences in simulated monthly NH3 concentrations over China by the two equilibrium models are less than 5%, and RPMARES runs about 30% faster in the GEOS-Chem adjoint."

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