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

Lin Zhang et al.

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Comment: This manuscript uses both top-down and bottom-up methods to investigate the spatial and temporal variations of agricultural ammonia emissions in China. The top-down estimates of NH_3 emissions, constrained by TES satellite NH_3 observations and optimized by GEOS-Chem adjoint model, show a summer peak that is underestimated in current bottom-up emissions inventories. To resolve the seasonal difference, the authors construct a new bottom-up inventory that takes account of seasonal variability in fertilizer application rates and emissions factors. The improved bottom-up inventory is broadly consistent with the top-down inversion results; both are validated by surface concentrations of NH_3 and wet deposition fluxes of NH_4+. Overall, I think the paper reads well, provides interesting results and deserves publication. I include

some minor comments and suggested revisions in the following text.

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: 1. Inverse method. The TES satellite NH3 columns are included in the observation vector, and these measurements are the basis for deriving seasonal variations in inverted NH3 emissions. Given the essential role of observational constraints, it is necessary to discuss in detail the influence of different satellite observations on seasonal variations of inversion results. It is good to see that "observations from AIRS, IASI, and CrIS" will be included in future studies. I suggest authors, at least in current state, to compare the seasonal cycle of NH3 columns measured by all the satellite sensors and to discuss the potential influences of using different data. Besides, it is not clear what means the offline NHx simulation for the iterative adjoint inversions. Please clarify it.

Response: We thank the reviewer for the suggestion. TES NH3 measurement is the only satellite dataset available to us when the study was conducted. We also think a comparison of different satellite retrievals from TES, AIRS, IASI, and CrIS will help us better understand the spatial and seasonal patterns of NH3 over China. This requires a deep analysis of different satellite datasets (with retrieval vertical sensitivity, i.e. averaging kernel matrix)). We think it is beyond the scope of this paper and should be a separated study.

As for the offline NHx simulation, we now state in the text "To lower the computational expenses, we follow the approach of Paulot et al. (2014) and use an offline NHx (NH3 + NH4+) simulation for the adjoint inversion that only calculates the physical and chemical transformation of NHx driven by hourly simulated sulfate and total nitrate (HNO3 + NO3−) concentrations archived from the standard simulation".

Comment: 2. Bottom-up method. There have been several recent studies that

use bottom-up method to establish high resolution emission inventory for NH3 in China. Most of these inventories peak its emissions during summer months, as shown in the literature review part of this paper. Therefore, in my opinion, improving NH3 inventory with strong seasonal cycle is not completely novel. The paper readers may ask what are the improvements and new points of this study in terms of approaches taken with the inventory development. These concerns are suggested to be clearly clarified in the revised manuscript.

Response: Based on our review in Sect. 2, the commonly used Chinese NH3 emissions are rather inconsistent with respect to the summer peak. We now state in Sect. 2: "Huang et al. (2012) suggests a weak summer peak in Chinese NH3 emissions, while the MASAGE inventory (Paulot et al., 2014) indicates largest emissions in April and July. NH3 emission estimates of Streets et al. (2003) have a strong peak in June, and are much higher than Huang et al. (2012) and Paulot et al. (2014) in winter". That is why we need to better constrain the Chinese NH3 emissions using both top-down and bottom-up approaches. Out bottom-up emission inventory as we state in the abstract and in the manuscript that "includes more detailed information on crop-specific fertilizer application practices and better accounts for meteorological modulation of NH3 emission factors in China".

The revised manuscript also includes more information on comparison of our bottom-up emission inventory with previous estimates as described in the response below.

Comment: 3. Results. I think the paper would be stronger if the improved emission inventory is compared in detail with previous bottom-up inventories. Table 1 presents comparison of national emission totals. Because this paper shows more concern on seasonal variations of NH3 emissions, more comparisons are needed to evaluate the new emission inventory, especially for seasonal variability.

Response: We have now added in Figure 1 our improved bottom-up NH3 emission inventory. We also state in Sect. 5.3: "The spatial distribution and seasonal variations of our bottom-up NH3 emission inventory are also presented in Figure 1 for compar-

ison 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: 4. Evaluation. I have significant concerns about the emission inventory evaluation with surface measurement of NHx. If I understand correctly, the GEOS-Chem model results for 2008 are directly compared against measurement data over 2008-2012 period. If this is the case, it may involve large uncertainties due to varying meteorological conditions and varying concentrations of SO2, NOx and oxidants in the atmosphere from year to year. It would be better to conduct an air quality modeling for 2008-2012. The NH3 emission used for the 5 years of model simulations can be fixed at 2008 because of small interannual variations. Or if the authors would not like to do this time-consuming work, I suggest only the measurement data for the year of 2008 can be used for model evaluation.

Response: Thank you for the suggestion. We have now conducted a GEOS-Chem model simulation over 2008–2012 with the improved bottom-up NH3 emissions for comparison against surface measurements. We fixed anthropogenic emissions of NOx and SO2 to the year 2008 conditions (due to a lack of relevant interannual variations in this model version) for testing the influences from varying meteorological conditions. We now state in Sect. 5.3: "To test the influences from varying meteorology, we have conducted a model simulation over 2008–2012 with our improved bottom-up NH3 emissions and other SO2 and NOx anthropogenic emissions fixed to the year 2008 conditions. Our results show small differences in simulated seasonal mean NH4+ wet deposition fluxes and NH3 gas concentrations between the 2008 and 5–year averaged model results except for the wintertime surface NH3 concentrations that the 2008 model results are 14% lower (Figs. 8 and 9 as discussed below vs. Supplemental Fig.

S2)".

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