

General comments:

This manuscript presented a new bottom-up estimate of ammonia emissions in China that emphasized some improvements on the estimates of fertilizer application-induced ammonia volatilization. The authors compared their Chinese ammonia emission inventory with a commonly-used emission estimate (MEIC), and evaluated the resulting model simulations using the WRF-Chem air quality model with surface and satellite ammonia measurements.

The manuscript is in general well conducted and organized, and it meets the scope of ACP. I have some comments below that suggest the authors better clarify the improvements of this ammonia emission inventory relative to previous studies. I think these comments should be addressed before considering publish.

Response: We really appreciate the valuable and constructive comments from the reviewer. We have made changes to both the main text and the supplemental information. Detailed responses are shown below.

Specific comments:

1) Page 5: It appears that most of the methods and datasets applied in this NH₃ emission inventory have followed previous studies (Huang et al., 2012; Kang et al., 2016; Zhang et al., 2018), except here as described in this section the improvements on fertilizer application-related NH₃ emission estimate. It is important to better identify the fertilizer application timing and proportion throughout the planting growing season. The key improvement in this ammonia emission inventory is depicted in Figure 1 that accounts for the spatial differences in fertilizer application timings for the three main crops (maize, wheat, and rice). From Figure 1, it is not clear what these fertilizer application dates are for each crop and each region. I think this information shall be presented, e.g., as a table in the manuscript or in the Supplement.

In addition, how the fertilizer amounts are distributed in each month according to the application timing needs some description in the text. Did you use a uniform fertilizer application rate for each crop in each month and region?

Response: We sincerely thank you for providing this constructive comment. As suggested, we added one Table (please see the Table S1 in the Supplementary materials) to show the basal and topdressing fertilization dates for different crops in different provinces.

In addition, we also reorganized the Section 2.1 and added one equation (equation (2)) to show how the N fertilizer amounts are distributed in different months.

“The respective amounts of each N fertilizer, applied in different months to each crop category, can be calculated using the following formula:

$$A_{i,j} = A_{total} \times P_j \times x_{i,j} \quad (2)$$

where i indicates the month. A_{total} indicates the total amount of each N fertilizer (ABC, DAP, NPK, urea, and other) applied to the different crop types in each province in 2016, which were calculated as a product of the planted cropland area and the fertilizer application rate per unit area of cropland,

based on data from MARA (2017) and NDRC (2017). Over the whole growth period, wheat, maize, and rice generally need three applications of N fertilizer (j , namely the basal dressing, first topdressing, and second topdressing). P_j represents the proportion of the total annual fertilization amount applied in the j -th dressing. The variation in P_j across the different regions was also considered, based on farmer survey data (Wang et al., 2008) and other studies in the literature (Zhang et al., 2009; Zhang and Zhang, 2012). $x_{i,j}$ represents the probability of the j -th dressing in month i , and is calculated as the proportion of days in the i -th month during the window period of j -th dressing. Table S1 lists the basal and topdressing fertilization dates for different crops in different provinces.

An example: in 2016, winter wheat was sown and basal fertilizer was applied in late September to mid-October. So, the dates of basal dressing in Hebei province span two months; $x=1/3$ in September, while $x=2/3$ in October. Similarly, the other two top dressings were applied at the jointing and booting stages, i.e., in late March to early April and late April to early May, respectively. According to the proportion of basal dressing and top dressing for wheat in Hebei, we identified the proportion of total fertilizer applied in each month (basal dressing: 0.2, September; 0.4, October; top dressing: 0.1, March; 0.2, April; and 0.1 in May).”

2) Page 9, Section 2.4: The results of Monte Carlo calculation were not presented in the manuscript. What are the uncertainties and probability distributions of ammonia emissions from fertilizer and livestock?

Response: We sincerely thank you for this critical comment. We added one sub-section (Section 3.4.1) to present the uncertainty assessment of NH₃ emission based on the Monte Carlo simulation.

“3.4.1 Uncertainty

Uncertainty in the estimated NH₃ emissions results from both the activity level and EF input data. We ran 10000 Monte Carlo simulations to estimate the range of NH₃ emissions from each source with a 95% confidence interval. The estimated total NH₃ emission range was 10.5-16.0 Tg. The 95% confidence intervals of fertilizer application, livestock waste, and others ranged from -20.5% to 64.41%, -23.0% to 37.1% and -42.9% to 62.4% (Fig. 6). Due to the large amounts of NH₃ emitted by fertilizers and livestock waste, the uncertainty of total NH₃ emissions is mainly caused by the uncertainties of these two sources. The uncertainty of fertilizer application was slightly greater than that of livestock waste. The emission factors, especially the corrected EF, were the largest contributors to the uncertainties of fertilizer application emissions. Additionally, it is clear that NH₃ emissions from other sources exhibited the largest uncertainty (-42.9% to 62.4%), mainly due to the high degree of uncertainty resulting from the many sub-sources, such as -77.1% ~ 96.9% of the transportation sector and -79.4% ~ 122.7% of the industrial sector. In comparison, the emissions from other sources were relatively small; hence, the large uncertainties of other sources did not have a significant impact on the uncertainty of total NH₃ emissions.

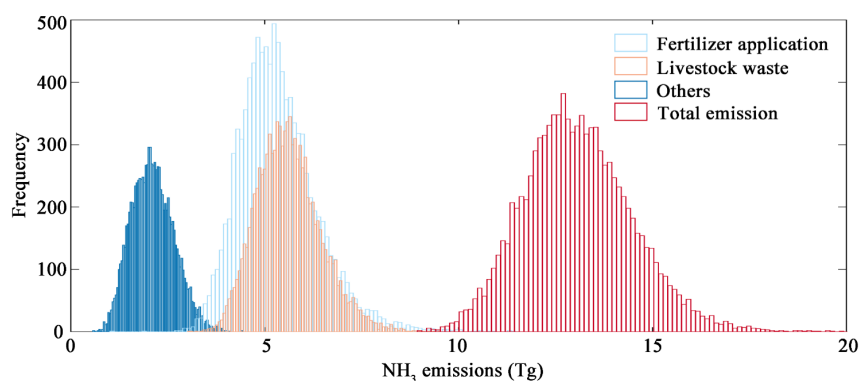


Fig. 6. Uncertainties of NH₃ emissions sourced from fertilizer application, livestock waste, and others.”

3) Page 17, Section 3.4: The study pointed out that Chinese ammonia emissions are high in summer and low in winter, and confirmed the results using WRF-Chem model simulations and ammonia measurements. This result did not seem to be much improved compared with previous estimates that all suggested higher ammonia emissions in summer than winter. I think that only analyzing the two months (January and July) could not provide sufficient information on the improvements of the ammonia emission inventory. The new fertilizer-induced ammonia emissions (Figure 5) also high values in April and October? Can you also evaluate improvements in these two months? This will provide valuable information to understand ammonia in spring and fall seasons.

Response: We appreciate the reviewer for providing this comment. As suggested, we further evaluated the spatial accuracy of the NH₃ emission inventory established in this study and the MEIC inventory in April and October 2016. It should be note that for October, we only used the IASI satellite observations to evaluate the spatial accuracy of the two inventories, because ground observation data were not available for October. The detailed accuracy evaluation was described in the Section 3.4.3.

4) Page 18, Fig. 6: The color scale for the left panel of Fig 6 could be misleading. It shows that at many sites, the model results are too high over the North China plain, however, the model results (contours) and measurements (dots) have different color scales. Suggest put them on the same color scale.

Response: Thanks for your valuable comment. According to your advice, we used the same color scale in Fig.7 (the original Fig.6).

5) Page10, Line 220: Here “9.29-15.54 Gg” should be in unit of “Tg”.

Response: We are sorry for this mistake. we have corrected it.

6) Page 11, Line 245: “replacing complex fertilizers with ABC might reduce fertilizer type-related NH₃ emissions”. Should here be “replacing ABC with complex fertilizer”?

Response: we have corrected it.

7) Page 17, Line 337: “R²=0.85” should be 0.84 as shown from Table 4. Was the R² value calculated by integrating January and July measurements?

Response: We have corrected it. The R^2 value was obtained by fitting the 2016 monthly values between IASI satellite observations and the NH_3 emissions from two inventories (MEIC and our study). To avoid this confusion, we revised the relevant sentence and added a Note in Table 4.

“We verified the accuracy of the monthly NH_3 emission trends obtained in this study by determining the correlation between the monthly IASI satellite observations and the monthly emissions of two inventories (MEIC and our study) in 2016.”

“Table 4. Comparison of the monthly NH_3 emission trends corresponding to IASI satellite observations with the two inventories.

Region	R^2		July/Jan ratio		
	IASI vs. OUR	IASI vs. MEIC	IASI	OUR	MEIC
...

Note: R^2 value was obtained by fitting the 2016 monthly values between IASI satellite observations and the NH_3 emissions from two inventories (MEIC and our study).”