Response to Referee #1

Received and published: 3 May 2019

This manuscript estimated global surface NH_3 concentration based on satellite retrievals and the temporal variation of NH_3 concentration was also presented. The study was well designed and the results are also important for evaluate NH_3 pollution in the world. The major comments to the manuscript are as follows.

The authors appreciate the valuable suggestions given by Referee #1 for improving the overall quality of the manuscript. In this document, we describe how we addressed the reviewer's comments. Detailed responses to each comment are given below (in blue).

1 Lines 278-280, for comparing the satellite-derived and measured surface NH_3 concentrations, are there any criterions to choose the sites which measured surface NH_3 concentrations? This is because satellite-derived surface NH_3 concentration in a grid (0.25^o latitude × 0.25^o longitude) is a reflection of the averaged NH_3 concentration in this grid area, but the NH_3 concentration measured at a site may only represent a limited area. For a grid with different sources of NH_3 (e.g., cropland, animal house or feedlot), the NH_3 concentration in this grid may have large spatial heterogeneity, then how to find a site with the surface NH_3 concentration to represent a grid area?

A point-to-grid verification strategy is adopted here, i.e. comparing the measurements at the monitoring stations with the grid values of satellite-derived estimates. We have to admit that this is the uncertainty of our analysis for comparing the satellite-derived and measured surface NH_3 concentrations since the monitoring site may not be representative of a given grid cell for an average retrieved value. We have added the following text to discuss this potential uncertainty in the section of "Validation of satellite-derived surface NH_3 concentrations":

"Notably, we compared the surface NH_3 concentrations at the monitoring stations with the grid values of satellite-derived estimates directly. This point-to-grid verification strategy may cause uncertainty since the monitoring site location may not be representative of a given grid cell for an average retrieved value.”.

2 Lines 282-284, for comparing NH_3 concentrations with different methods, the information on how many measuring sites, and where the sites located should be given for each country or region.

Thanks very much for this good suggestion. We have added the number of measuring sites in each region by the following text:

“IASI-derived surface NH_3 concentrations gained higher consistency with the ground-based measurements in China (R^2=0.71 and RMSE=2.6 μg N m^{-3} for 43 sites) than the US (R^2=0.45 and RMSE=0.76 μg N m^{-3} for 67 sites) and Europe (R^2=0.45 and RMSE=0.86 μg N m^{-3} for 43 sites) at a yearly scale”.

The sites locations have been given for each region in Fig. 2 in the manuscript .

3 Lines 284-286, as mentioned in comment 1, the spatial heterogeneity of NH_3 concentration in a grid and the measuring sites location may also cause the differences between satellite-derived and ground-based NH_3 concentration. Thus, this discussion should be added here.

This concern has been addressed in the response to comment 1. Please refer to it.

Besides, the detection limit and precision for deriving NH_3 concentration using the satellite should be given.

Thanks very much for this good suggestion. We have added the following text for clarifications:

“The satellite-derived NH_3 has a detection limit of 0.0025 μg N m^{-3} (2.5 ppb) (Graaf et al. 2018; Van Damme et al. 2014).”.

4 Lines 318-320, More details of the location of NH_3 hotspots should be given. In China, where is the eastern China? It seems that there were also NH_3 hotspots in Shannxi, Shanxi, Gansu and Hubei provinces, and there were no hotspot (> 8 ug N m^{-3}) in Xinjiang province in Fig. 4?

Thanks very much for this good comment. We have revised the “eastern China” as “eastern China (109-122^{o} E, 28-41^{o} N)”.

To show more details of the locations of NH_3 hotspots, we have revised the descriptions by the following text:

"We found large areas in eastern China (109-122^{o} E, 28-41^{o} N), Sichuan Basin, Hubei (including Wuhan, Xiangyang and Yichang), Shaanxi (including Xi'an, Baoji, Hanzhong, Weinan), Gansu (Lanzhou and its surrounding areas), Shanxi (including Yuncheng and Changzhi) and northwestern Xinjiang with surface NH_3 concentrations greater than 8 μg N m^{-3} y^{-1}.".

5. Lines 321-324, in fact, more than half the NH_3 emissions in China is caused by animal production. The higher NH_3 concentration in eastern China can also be caused by animal production. More discussion and supporting data should be provided to strengthen the contribution of animal production on NH_3 concentration. This is also true for US and Europe.

Thank you very much for this suggestion. We have carefully checked the NH_3 emissions. In addition to N fertilization, N manure is another major source of NH_3 emissions in China, and the percentage of N manure to NH_3 emissions exceeds 50% (Kang et al. 2016). So we have added the N manure into our analysis in Fig. 4 and Fig. 5 and revised related text in the discussion.

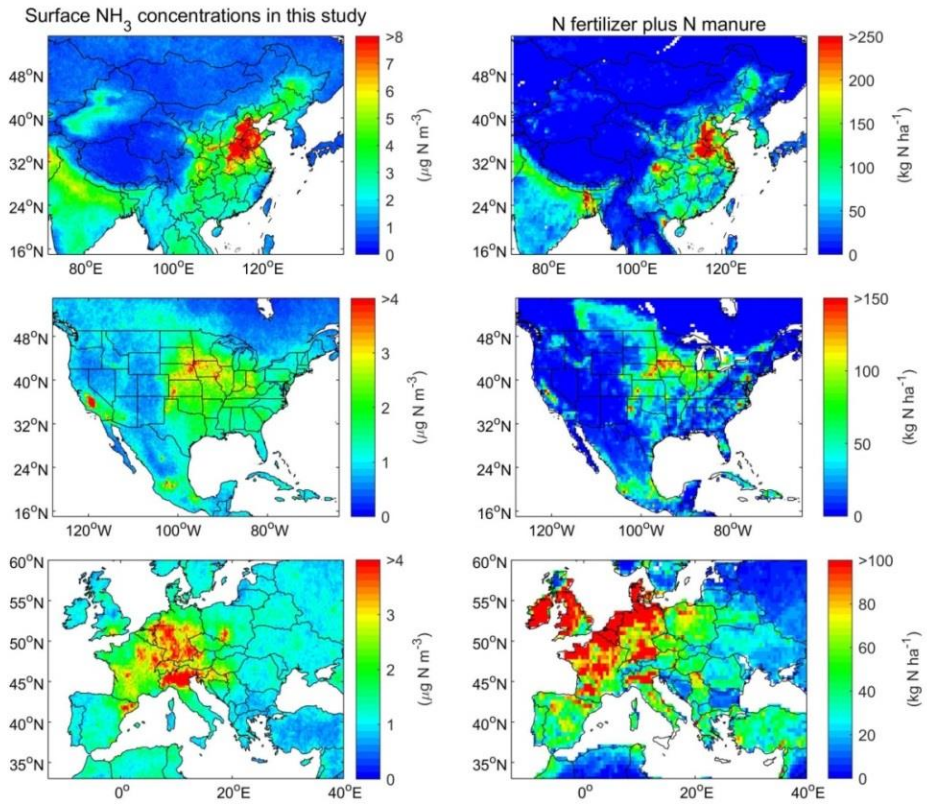

Fig. 4 Spatial distribution of IASI-derived surface NH_3 concentrations, and N fertilizer plus N manure in China, Europe and US.

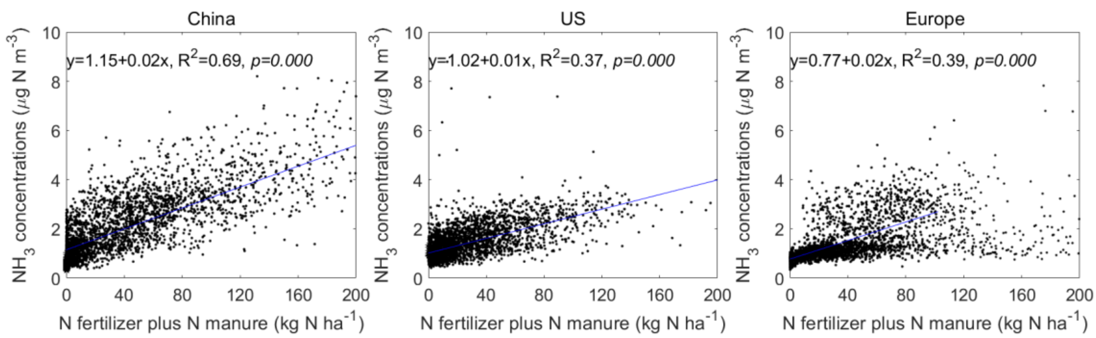

Fig. 5 Comparison of satellite-derived surface NH_3 concentrations, and N fertilizer plus N manure in China, US and Europe. The spatial resolution of satellite-derived surface NH_3 concentrations and N fertilizer plus N manure is 0.25° and 0.5°, respectively. We firstly resampled the satellite-derived surface NH_3 concentrations to 0.5° grids, and then compared it with N fertilizer plus N manure by each grid cell. We obtained the N fertilizer and N manure data produced from McGill University (Potter et al. 2010).

6. Lines 398-402, are there any differences for the seasonal variation of NH_3 concentration in different regions?

Yes. We take a case study on the seasonal NH_3 concentration in two hotspots of eastern China and Guangdong. The maximum surface NH_3 concentration in eastern

China occurred in June and July, which coincided with the planting, fertilization time and higher temperature of the main crops in the region (Van Damme et al. 2015). The maximum surface NH_3 concentration appeared in March in Guangdong, which was also closely related to crop fertilization in these areas (Shen et al. 2009; Van Damme et al. 2014). We have added the following text for clarifications:

“Notably, there is a difference in the seasonal variations of surface NH_3 concentrations between ECH (peaking in June and July) and GD (peaking in March), which was likely related to different crop planting, N fertilization time as well as meteorological factors (Shen et al. 2009; Van Damme et al. 2014; Van Damme et al. 2015).”.

7. Lines 486- 488, which sector (crop or animal production) did cause the increase of NH_3 emissions in China in 2008-2015?

We have added the following text to explore the potential reasons:

“The increase of surface NH_3 concentrations in eastern China was consistent with the trend of NH_3 emission estimates by a recent study (Zhang et al. 2017). Approximately 85% of the inter-annual variations was due to the changes of human activities, and the remaining 15% resulted from air temperature changes. Agricultural activities is the main drive of NH_3 emission increase, of which 43.1% and 36.4% were contributed by livestock manure and fertilizer application (Zhang et al. 2017).”.

Reference

Graaf, S.C.v.d., Dammers, E., Schaap, M., & Erisman, J.W. (2018). How are NH_3 dry deposition estimates affected by combining the LOTOS-EUROS model with IASI-NH3 satellite observations? *Atmospheric Chemistry and Physics, 18*, 13173-13196

Kang, Y., Liu, M., Song, Y., Huang, X., Yao, H., Cai, X., Zhang, H., Kang, L., Liu, X., Yan, X., He, H., Zhang, Q., Shao, M., & Zhu, T. (2016). High-resolution ammonia emissions inventories in China from 1980 to 2012. *Atmospheric Chemistry and Physics*, *16*, 2043-2058

Potter, P., Ramankutty, N., Bennett, E.M., & Donner, S.D. (2010). Characterizing the Spatial Patterns of Global Fertilizer Application and Manure Production. *Earth Interactions, 14*, 1-22

Shen, J.L., Tang, A.H., Liu, X.J., Fangmeier, A., Goulding, K.T.W., & Zhang, F.S. (2009). High concentrations and dry deposition of reactive nitrogen species at two sites in the North China Plain. *Environmental Pollution, 157*, 3106-3113

Van Damme, M., Clarisse, L., Dammers, E., Liu, X., Nowak, J., Clerbaux, C., Flechard, C., Galy-Lacaux, C., Xu, W., & Neuman, J. (2014). Towards validation of ammonia (NH_3) measurements from the IASI satellite. *Atmospheric Measurement Techniques, 7*, 12125-12172

Van Damme, M., Erisman, J.W., Clarisse, L., Dammers, E., Whitburn, S., Clerbaux, C., Dolman, A.J., & Coheur, P.F. (2015). Worldwide spatiotemporal atmospheric ammonia (NH_3) columns variability revealed by satellite. *Geophysical Research Letters, 42*, 8660-8668

Zhang, X., Wu, Y., Liu, X., Reis, S., Jin, J., Dragosits, U., Van Damme, M., Clarisse, L., Whitburn, S., Coheur, P.-F., & Gu, B. (2017). Ammonia Emissions May Be Substantially Underestimated in China. *Environmental science & technology, 51*, 12089-12096