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Interactive comment on "Quantification and evaluation of atmospheric ammonia emissions with different methods: A case study for the Yangtze River Delta region, China" *by* Yu Zhao et al.

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Title: Quantification and evaluation of atmospheric ammonia emissions with different methods: A case study for the Yangtze River Delta region, China

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We thank very much for the valuable comments and suggestions from reviewer #2,

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Discussion paper

which help us improve our manuscript significantly. The comments were carefully considered and revisions have been made in response to suggestions. Following is our point-by-point responses to the comments and corresponding revisionsãĂĆ

0. This paper compares and contrasts two methodologies for estimating emissions of NH3 in China, and illustrates the consequence through model simulations. The paper deals with an important subject, since the large uncertainties surrounding ammonia emissions need to be understood by modelers and policy experts. The paper is generally well written, and generally sound.

Response and revisions:

We appreciate the reviewer's positive remarks on the importance of the work.

1. I miss consideration of many of the factors omitted from the emission estimation procedure. This study basically used temperature, and agricultural statistics, to calculate emission factors (EFs). However, with respect to emissions from livestock/poultry, wind-speed is also a very important factor (e.g. Gyldenkaerne et al., 2005, Skjoeth et al., 2011, Flechard et al, 2013). Many other factors should also impact NH3 emissions, such as radiation, rainfall (and other precipitation), leaf-wetness, atmospheric stability, large uncertainties in the so-called Gamma factors, or bi-directional exchange in general (Bash et al, 2013, Flechard et al., 2013, Massad et al, 2010, Wichink Kruit et al., 2012). Consideration of such factors might also help to explain some if the model discrepancies outlined in Section 3, and should at least be considered before trying to explain all such discrepancies in terms of temperature and a few selected variables only

Response and revisions:

We thank and agree the reviewer's important comment. In this work, we mainly compared the magnitude and the spatial and temporal distribution of the YRD NH3 emissions estimated with two different methodologies, and evaluated the two inventories through air quality modeling based on available satellite and ground observation within the region. Compared to E1, in particular, E2 included the impacts of the growing and farming cycles, soil properties (pH) and selected meteorological factor (temperature) on NH3 emissions for fertilizer using sector, and those of manure management processes and ambient temperature for livestock/poultry breeding. Besides the parameters we are concerned with, however, some other factors and processes also play important roles on atmosphere-land exchange of NH3, as pointed by the reviewer. Those factors/processes that were not considered in this work include given meteorological factors (e.g., wind speed, precipitation and leaf surface wetness), surface layer turbulence, air and surface heterogeneous-phase chemistry, and plant physiological conditions (Flechard et al, 2013). In general, those factors/processes could be integrated in the bi-directional surface–atmosphere exchange module coupled in the air quality modeling, and improved estimation of NH3 flux (emissions and depositions) were expected. The modeling system with the bi-directional NH3 exchange were reported to be able to reduce the biases and error in simulation of NHx (NH3 +NH4+) wet deposition and ambient aerosol concentrations for both US and Europe (Bash et al, 2013; Wichink Kruit et al., 2012). Limited studies on the bi-directional NH3 exchange were found for China (e.g., Fu et al., 2015). Out of the scope of current work, we did not focus on the bi-directional NH3 exchange module and did not include the module for emission evaluation and comparison. We agree with the reviewer that the ignorance of given parameters/process in the estimation could potentially further explain the discrepancy between the simulation and observation. A more comprehensive evaluation and comparison in NH3 emission inventories was thus suggested in the future, including the bi-directional NH3 exchange and the top-down constraint with inversed modeling. We have discussed this limitation and added relevant literatures in lines 561-580, page 18 in the revised manuscript.

2. The authors use meteorology from ECMWF for their emissions, but why not the WRF model, since that is obviously available and is used for their CMAQ runs?

Response and revisions:

We thank the reviewer's comment. We do not have very specific reason for using the ECMWF instead of WRF. When calculating the emissions, the underlying data open to the public were preferentially selected. ECMWF provided daily average data that satisfied our need of emission estimation and they were open to the public, thus we selected the dataset.

3. The equations used are generally clearly written out, although it isn't always clear where they are coming from. For example, is it correct that equations 2 & 3 are a mixture of methods from Huang et al 2012 and EEA 2013? On the other hand, I read in various sections of EEA 2013 that temperature functions could not be provided (e.g. chap. 3.D crop production and agricultural soils) If from EEA, then it would also be good to cite the scientific papers underlying the EEA guidelines, and to be more specific as to which sections of EEA are being cited (it is a monster document).

Response and revisions:

We appreciate the reviewer's comment. The specific EEA guidelines (EEA 2013a; 2013b; 2009) were provided in the revised manuscript. For Eq. 2 & 3, in particular, the linear relationships between NH_3 volatilization rate and temperature/soil pH were described in Chap. 4.D crop production and agricultural soils of EEA (2009)/Huang et al. (2012), and we specified them respectively in lines 227-228, page 8 and lines 211-212, page 7 in the revised manuscript.

4. Some other points:

P2. The abstract is rather long, and should be shortened for clarity.

Response and revisions:

We thank the reviewer's comment and the abstract was shortened.

P3, L67. NH_3 is said to react with NO_x, but NO_x usually means $NO+NO_2$. I think the authors mean HNO_3?

Response and revisions:

We thank the reviewer's reminder and it is corrected as nitric acid (HNO_3) in the revised manuscript.

P3, L78-81. The sentence is a little unclear. Clarify.

Response and revisions:

We thank the reviewer's comment. We mean that SO_2 and NO_X emissions have gradually decreased due to improved control, thus the NH_3 emissions was found to play a greater role on the secondary aerosol formation and nitrogen deposition, compared to previous years. The sentence is rewritten in lines 72-76, page 3 in the revised manuscript: Recently the SO_2 and NO_X emissions have gradually decreased due to implementation of various pollution control measures in China, thus NH_3 emissions were found to play a greater role on secondary aerosol formation and nitrogen deposition compared to previous years.

P4, L112. Methods of including meteorology in NH_3 emissions have been around for some time and should be mentioned, e.g. Gyldenkaerne et al., 2005, Skjoeth et al., 2011, Wichink Kruit et al., 2012, Bash et al., 2013.

Response and revisions:

We thank and agree the reviewer's comment. We have added the relevant papers and description in lines 110-111, page 4 in the revised manuscript.

P5, L148. Another source of human-related NH_3 emissions is pets. As shown in e.g. Sutton et al 1995, 2000, human pets can be as significant as human metabolism with regard to NH_3 emissions.

Response and revisions:

We thank and agree the reviewer's comment. Due to lack of detailed statistic, we did not include pet emissions in current NH3 inventories. Given the relatively small fraction in total emissions (e.g., less than 2% for United Kingdom by Sutton et al.), we believe that the uncertainty was limited. We have added the explanation in lines 149-152, page 5 in the revised manuscript.

P6, L168. Using should be used.

Response and revisions:

We thank the reviewer's reminder and it is corrected in the revised manuscript.

P7, L187. Give reference for radiometer

Response and revisions:

We thank the reviewer's reminder and the reference for radiometer is given in the revised manuscript (Davies et al., 2009).

P7, L202. The study of Huang et al 2012 uses a linear relationship between pH and EF. Why is the relation here said to be near-linear?

Response and revisions:

We thank the reviewer's reminder and it is corrected as linear in the revised manuscript.

P7. What is the time-resolution of the EF calculations?

Response and revisions:

The time-resolution of EF calculation is monthly. In the method, the fertilization method (top or basal dressing) was month-dependent, and monthly average temperature was applied for the EF calculation. We have added the information in lines 212-213, page 7 and line 220, page 8 in the revised manuscript.

P8, L232. Surely fertilizer application at 15-20cm affects the pH of the soil; doesn't this affect the assumptions made when using global pH data from IIASA?

Response and revisions:

We thank and agree the reviewer's comment. Previous domestic experimental studies in China (e.g, Zhong et al., 2006) indicated that the fertilizer application would increase the soil pH, particularly for the acidic soils. Bias thus existed in soil pH from the global database, without considering the detailed schedule and method of fertilizer application. As the quantitative relation between the fertilizer application and soil pH was still lacking at the regional scale in China, we ignored the interaction between the fertilizer application and soil pH in Eqs.(2). We acknowledged the limitation and added the explanation in lines 243-248, page 8 in the revised manuscript.

P9. The basic references of the CMAQ model should be given, not just a web-address.

Response and revisions:

We thank the reviewer's reminder and the basic operational guidance of CMAQ by University of North Carolina was provided in the revised manuscript (UNC, 2010).

P10. Which version of MEGAN was used? Did you use data provided by Sindelarova, or did you use the MEGAN model itself? If the latter, a Guenther et al ref would seem more

Response and revisions:

We thank the reviewer's comment. We used the MEGAN 2.1. The literature (Guenther et al., 2012) has been added in the revised manuscript.

P10. Again, give reference to the model developers - this time for WRF.

Response and revisions:

We thank the reviewer's reminder and the reference of WRF is provided in the revised manuscript (Skamarock et al., 2008).

P11. The Lanciki 2018 reference for MARGA is missing.

Response and revisions:

We thank the reviewer's reminder and the information of Lanciki (2018) is provided in the revised manuscript.

P15. The citation of Wei et al (2015) is in Chinese, and thus not helpful for most authors. This instrument has been around for many years, and the artifacts documented elsewhere. Please find some citations in English for the problems mentioned.

Response and revisions:

We thank the reviewer's comment and provided English papers for the problem in lines 484-485, page 16 in the revised manuscript. (Chen et al., 2017; Schaap et al., 2011; Stieger et al., 2018).

P28, Use molecule not "mole.", to avoid confusion with the mole unit.

Response and revisions:

We thank the reviewer's reminder and molecule is used in the revised manuscript.

P31. Table 3. Correlation coefficients should be added, and the time-resolution of the statistics mentioned.

Response and revisions:

We thank the reviewer's comment. The correlation coefficients between the observation and simulation were added in the revised Table 3, and the time-resolution of the statics was hourly, as mentioned in the revised caption of the table.

P31 cont. for all Tables make it clear if statistics are calculated from hourly, daily or monthly values.

Response and revisions:

We thank the reviewer's reminder. The statistics in Tables 3 and 6 were calculated based on the hourly values, and those in Tables 4 and 5 were from the daily values (the value of one hour (9:30am for satellite observation and the average of 9:00am-10:00am for simulation) per day). The information has been added in the revised captions of Tables 3-6.

There are small English misses throughout, for example with regard to singular or plural, or omission of the definite article (the).

Response and revisions:

We thank the reviewer's comment and the grammar errors are corrected in the revised manuscript.

References

Bash, J. O., Cooter, E. J., Dennis, R. L., Walker, J. T., Pleim, J. E.: Evaluation of a regional air-quality model with bidirectional NH3 exchange coupled to an agroecosystem model, Biogeosciences, 10, 1635-1645, 2013.

Chen, X., Walker, J. T., Geron, C.: Chromatography related performance of theMonitor for AeRosols and GAses in ambient air (MARGA): laboratory and field-based evaluation, Atmos. Meas. Tech, 10, 3893-3908, 2017.

Davies, D. K., Ilavajhala, S., Wong, M. M., and Justice, C. O.: Fire Information for Resource Management System: Archiving and Distributing MODIS Active Fire Data, IEEE Geosci. Remote Sens., 47, 72-79, 2009.

European Environment Agency (EEA): EMEP/CORINAIR Air Pollutant Emission Inventory Guidebook-2013, 3.B Manure management, available at: https://www.eea.europa.eu/publications/emep-eea-guidebook-2013/part-b-sectoral-guidance-chapters/4-agriculture/3-b-manure-management/view (last access: 25 Feb 2020), 2013a.

European Environment Agency (EEA): EMEP/CORINAIR Air Pollutant Emission Inventory Guidebook-2013, 3.D Crop production and agricultural soils, available at: https://www.eea.europa.eu/publications/emep-eea-guidebook-2013/part-b-sectoral-guidance-chapters/4-agriculture/3-d-crop-production/view (last access: 25 Feb 2020), 2013b.

European Environment Agency (EEA): EMEP/CORINAIR Air Pollutant Emission Inventory Guidebook-2009, 4.D Crop production and agricultural soils, available at: https://www.eea.europa.eu/publications/emep-eea-emission-inventory-guidebook-2009/part-b-sectoral-guidance-chapters/4-agriculture/4-d/4-d-crop-production-and-agricultural-soils.pdf/view (last access: 25 Feb 2020), 2009.

Flechard, C. R., Massad, R.-S., Loubet, B., Personne, E., Simpson, D., Bash, J. O., Cooter, E. J., Nemitz, E., Sutton, M. A.: Advances in understanding, models and parameterizations of biosphere-atmosphere ammonia exchange, Biogeosciences, 10, 5183-5225, 2013.

Fu, X., Wang, S., Ran, L., Pleim, J. E., Cooter, E., Bash, J. O., Benson, V., Hao, J.: Estimating NH3 emissions from agricultural fertilizer application in China using the bi-directional CMAQ model coupled to an agro-ecosystem model. Atmos. Chem. Phys., 15, 6637-6649, 2015

Guenther, A. B., Jiang, X., Heald, C. L., Sakulyanontvittaya, T., Duhl, T., Emmons, L. K., Wang, X.: The Model of Emissions of Gases and Aerosols from Nature version 2.1 (MEGAN2.1): an extended and updated framework for modeling biogenic emissions. Geosci. Model Dev., 5, 1471-1492, 2012.

Lanciki, A.: 2060 MARGA Monitor for AeRosols and Gases in ambient Air. Metrohm Process Analytics, Switzerland, available at: https://www.metrohm.com/en/products/process-analyzers/applikon-marga/ (last access: 10 Feb 2020), 2018.

Schaap, M., Otjes, R. P., Weijers, E. P.: Illustrating the benefit of using hourly monitoring data on secondary inorganic aerosol and its precursors for model evaluation. Atmos.

Chem. Phys., 11, 11041–11053, 2011

Skamarock, W. C., Klemp, J. B., Dudhia, J., Gill, D. O., Barker, D. M., Duda, M. G., Huang, X.-Y., Wang, W., Powers, J. G. A Description of the Advanced Research WRF Version 3. NCAR Tech. Note NCAR/TN-475+STR, 113 pp.doi:10.5065/D68S4MVH, 2008.

Stieger, B., Spindler, G., Fahlbusch, B. Muller, K., Gruner, A., Poulain, L., Thoni, L., Seitler, E., Wallasch, M., Herrmann, H.: Measurements of PM10 ions and trace gases with the online system MARGA at the research station Melpitz in Germany - A five-year study. J. Atmos. Chem., 75, 33-70, 2018.

University of North Carolina at Chapel Hill (UNC): Operational Guidance for the Community Multiscale Air Quality (CMAQ) Modeling System Version 4.7.1 (June 2010 Release), available at http://www.cmaq-model.org (last access: 10 Feb 2020), 2010.

Wichink Kruit, R. J., Schaap, M., Sauter, F. J., van Zanten, M. C., van Pul, W. A. J.: Modeling the distribution of ammonia across Europe including bi-directional surface atmosphere exchange, Biogeosciences, 9, 5261-5277, 2012.

Zhong, N., Zeng, Q., Zhang, L., Liao, B., Zhou, X., Jiang, J.: Effects of acidity and alkalinity on urea transformation in soil, Chinese Journal of Soil Science, 37, 1123-1128, 2006 (in Chinese).

Interactive comment on Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2019-689, 2019.