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Atmospheric Chemistry and Physics Discussions

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 #3,

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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 revisions.

0. The manuscript develops and presents two gridded NH3 emission inventories, one based on emission factors from the literature and a second with more process information. The two are compared against one other, as well as to two ground sites. CMAQ output was also be compared against satellite columns. This is a good exploration of what is known about NH3 emission patterns in the heavily populated Yangtze River Delta region. That said, without well understanding the methods E1 and E2, it was difficult to fully review manuscript.

Response and revisions:

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

1. Emission inventories with general emission factors or more detailed process have always been used, so at first read I am not sure why this is considered as case study of the methodology versus something like "Quantification and evaluation of atmospheric ammonia emissions for the Yangtze River Delta region, China". The exact methods used for E1 and E2 are fairly confusing. The "constant emission factors" method that is referenced throughout are actually based on annual emission factors, with a monthly and spatial allocation schemes given on L179-L188. This needs to be clearer early on in the manuscript. Also, to confirm, neither allocation affects the total yearly emission? Are the activity factors different in E1 than what are used in E2?

Response and revisions:

We appreciate the reviewer's important comment and acknowledged that some descriptions on the principles of the two methods were unclear in the original manuscript. The reviewer was correct for E1. It was developed based on the constant annual emission factors at the prefectural city level (as most of the activity data could be obtained at

the prefectural city level). Spatial and monthly allocations of emissions were then conducted, without changes in total yearly emissions. Following the reviewer's suggestion, we mentioned this at the beginning of Section 2.1 (lines 143-145, page 5 in the revised manuscript). It is a relatively quick and simple method, based on the previous understanding of NH3 emissions at regional scale (both the emission factor and temporal distribution). The effects of actual environmental conditions and agricultural activities on emission rate were not considered at a high temporal and spatial resolution.

In E2, the method for fertilizer application and livestock/poultry breeding (the main source categories of NH3) was improved. In particular, the emission factors were developed at the monthly resolution, integrating the effects of soil, meteorology and agricultural processes. Therefore the method did not only change the temporal pattern of emissions but also the magnitude of the annual emissions, as the emission factors developed in this method varied from the ones applied in E1, which were directly taken from previous studies.

It should also be noted that the annual activity data were the same for the two inventories at the prefectural city level, although the monthly distributions were different. We have clearly stated the relation between the activity data in the two inventories in lines 196-197, page 7 in the revised manuscript. Please also see our response to Question 2 of the reviewer.

2. Sect 2.2.1 about E2: please check each use of 'corrected' to make sure it is clear what/how/why something is being corrected. Specifically, L198 why does the fertilizer use need to be corrected? Where do the relationships in Table S2 come from?

Response and revisions:

We appreciate the reviewer's comment and acknowledged some "corrected" were confusing. In E2, the emissions from fertilizer use and livestock/poultry breeding were recalculated with a different method from E1. In general, it is not a correction of E1. Therefore we deleted unnecessary "corrected" in the methodology section of E2 (Sec-

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tion 2.2). Regarding the word “corrected” fertilizer as pointed by the reviewer, in particular, we actually means that the monthly fertilizer used was estimated in E2 combining the information of investigated farming cycles. The annual total fertilizer was the same as E1. We also added an example of early-season rice for better understanding the method. The relevant texts have been revised in lines 199-209, page 7 in the revised manuscript.

Regarding Table S2, we clearly stated that the annual total amount of fertilizer used were the same by prefecture city and type in the two inventories in lines 196-197, page 7 in the revised manuscript. The method of estimating the annual amount of fertilizer by prefecture city and type in Table S2 was described in lines in lines 165-171, page 6 in the revised manuscript.

3. Specific technical/style: L206 EFbase -> EFbasal and Tbase -> Tbasal

Response and revisions:

We thank the reviewer’s reminder and terms are corrected in the revised manuscript.

L206-207 Are Tbasal and T0 in different units? Otherwise, 273.15 wouldn’t be needed

Response and revisions:

We thank the reviewer’s reminder and 273.15 was deleted in the revised manuscript.

L213 ‘method’ -> ‘application method’? (if I’m guessing correctly). What are the possible methods?

Response and revisions:

We thank the reviewer’s reminder and it is revised as application method (basal dressing).

L347-L349 where is this shown about the EFs being from hot seasons?

Response and revisions:

We appreciate the reviewer's comment. Most of the measurements on emission factor of ammonia from fertilizer application were conducted in summer or late spring (Cai et al., 2002; Huo et al., 2015; Su et al., 2006), especially those using micrometeorological method. It is expectable since that the basal dressing of single-season rice and maize as well as top dressing of wheat are usually conducted in late spring or summer. However, the crop rotation varies a lot in China, and part of the nitrogen-containing fertilizer actually is not applied in hot seasons. Emission estimation based on those emission factors may thus overestimate the emission intensity of ammonia (Huo et al., 2015; Wang et al., 2011; Zhang et al., 2010). We have provided relevant literatures and added the above discussion in lines 361-369, page 12 in the revised manuscript.

L518-L519 Please reword. IASI is an instrument, so it cannot 'provide' an averaging kernel.

Response and revisions:

We thank the reviewer's reminder and the sentence is rewritten in lines 537-538, page 17 in the revised manuscript:

As the ESPRI product of NH3 VCDs we applied in the study does not provide the averaging kernel. . .

Figure 1 caption: "Studying area and research domain" -> aren't study area and research domain the same?

Response and revisions:

We thank the reviewer's reminder, and they are the same. We delete the "Studying area" in the figure caption.

Figure 3 and Figure 6: 'Januray" -> "January"

Response and revisions:

We thank the reviewer's reminder and the errors are corrected.

Figure 4: emissions misspelled in the y-axis label Figure 4: Suggest giving fertilizer and livestock consistent colors, then keeping E1 as solid fill but E2 as hatched for easier reading

Response and revisions:

We thank the reviewer's reminder and the figure is improved as required.

Figure 6: colorscales could have greater consistency

Response and revisions:

We thank the reviewer's reminder and the figure is improved as required.

Figure 9: the subplots should have a consistent axis font size

Response and revisions:

We thank the reviewer's reminder and the same font size is applied in all the subplots.

Figure 10; from caption, shouldn't axis limits be same as Figure 9? Also, helpful to add the border lines like in Figure 9 so one is orientated where they are looking

Response and revisions:

We thank the reviewer's reminder and revised the axis limits. The border lines have also been added in the revised Figure 10.

Figure S4: there is one main cluster of data along the black line, but why is there seem to also be a second one? Also, what is the significance of the red dots, which do not fit well especially for the ABC panel?

Response and revisions:

The black line is obtained through linear regression based on all the blue dots (including the "second cluster" mentioned by the reviewer).

Different from blue dots that are calculated for all the grids within the research domain

of this study, the red dots are taken from available field measurement studies, as we mentioned in the figure caption. The gap between them, in particular at lower soil pH, explained the possible uncertainty in current method, i.e., the current linear assumption between the soil pH and NH3 volatilization rate might not be appropriate for soil with low pH values for eastern China. We discuss it in lines 553-560, page 18 in the revised manuscript.

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