

Interactive comment on "Estimating NH₃ emissions from agricultural fertilizer application in China using the bi-directional CMAQ model coupled to an agro-ecosystem model" by X. Fu et al.

Anonymous Referee #4

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Review of Fu et al., "Estimating NH3 emissions from agricultural fertilizer application in China using the bi-directional CMAQ model coupled to an agro-ecosystem model" for ACPD.

This manuscript couples various models to resolve the spatial and temporal emissions of NH3 in China. Quantifying NH3 emissions is critical towards mitigating fine particulate matter as NH3 plays important roles in aerosol composition and growth. China, in particular, is expected to have large emissions from past studies but there are large

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uncertainties as well. Part of the problem is the paucity of NH3 (and related aerosol pre-cursor, aerosol composition) measurements in this region due to measurement challenges intrinsically and lack of publically-available datasets. The authors state that agricultural fertilizer emissions from China are 3 Tg in 2011, an emission that is the lower than all previous studies but still on the same order of magnitude with them.

Clearly, the manuscript is on a subject matter appropriate for and of interest to ACP. However, I struggle to understand the significance and context of the results in this study. For example, despite the huge uncertainties in the various input data for the models, there is no estimate of the corresponding uncertainties in the overall emission rate. Sensitivity studies are needed to show how uncertainties in various quantities would result in changing the total emission rate. While I recognize that the input data also likely doesn't have intrinsic uncertainties, the authors need to provide uncertainty estimates of these input variables or at least show how the final emissions would change for a given range of uncertainty. What parameters are most sensitive to the final number and to what extent?

Furthermore, more thorough discussion is needed to compare to prior studies other than a vague statement that emission factors from the US or UK are not applicable to China. While this may be true, what aspects of the emission factors are not relevant? How are management practices so different that the US/UK emission factors would not be representative of those practices in China?

Regarding data availability, it is unclear to me which datasets on Chinese agricultural practices (fertilizer use, crop use, etc.) are available to the wider public so that future studies may improve upon this study. One of the citations was a thesis study in Chinese (Zhang, 2008) – is this available to the broader community? What about other data sources?

Finally, the manuscript needs a thorough read by a native English speaker. There are many cases with extra or missing definite articles ('the'). Clauses starting with "which"

are frequent, sometimes properly and many times improperly. The co-authors from the US should be able to provide and correct these oversights or questionable grammar aspects.

Particular details/comments:

1. First sentence of the abstract regarding ammonia's importance to atmospheric chemistry – I would be more specific here and emphasize its importance in aerosol composition/chemistry instead. Gas phase atmospheric chemists have long ignored NH3 because it really doesn't matter. While I agree on balance it is very important, for the first sentence in the abstract I would refine the focus slightly.

2. p. 748, line 5: inorganic aerosol is more convention, rather than "non-organic"

3. p. 749, lines 4-7: "...correction factors are empirical and too simple." More elaboration is needed here – what parts of the emission factors and why are they not appropriate?

4. Section 2.2.1: Is there a database of the cropland area for 2710 counties? If not available publically, can the authors post their datasets in such a location for others?

5. Section 2.2.2 and 2.2.3: since everything is related to the 36 x 36 km CMAQ grid, also specify in parentheses the scale in km after the native unit resolutions for the soil database (arc-second) and MERRA (0.5 degree x 0.667 degree) to help the reader

6. Section 2.4.4: It is unclear why the EPIC model uses heat units in some cases and unpublished data in other cases. What is the justification to selectively use on metric over the other? Why not use the best database, which appears to be the unpublished Chinese Academy of Sciences research? Can the authors provide a link to that data used in this study?

7. Section 3.2.2 (Comparison to other studies) was lacking, as noted in my most significant comments above. No detailed discussion of the differences between studies are noted, other than a generalized statement that "the parameters were set based on

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conditions in UK, which may be very different from China". Likewise, a similar statement is made about US experiments. What happens the parameters were similar to the US, Europe, or UK? Then how can one explain the different results?

8. Section 3.3, CMAQ and ground observations: Is the assumption that if CMAQ models NO3- correctly that NH4+ would also be OK? This argument is flawed. Fore example, see Schiferl et al., JGR, 119, 1883-1902, 2014 on the difficulties of modeling NO3- and NH4+ with other constituents and trying to match observations. I found this entire section to be speculative and not add much to the paper. I suggest removing or significantly revising.

9. Section 3.4: The uncertainty analysis here is actually just a qualitative discussion of potential biases, and a discussion in the most general of sense. This section needs to be greatly expanded so others can assess how changes in the input parameters will result in changes to the total emissions. A full sensitivity analyses is needed here, particularly if datasets are not readily available. Otherwise, it will be difficult for others to ever compare to this study.

10. Conclusions: What are the larger implications of your work? If NH3 is less from fertilizer than others report, does this mean that animal systems are more important to controlling ammonia? What is the comparable scale between such inventories? Some larger perspectives are needed here.

Overall, while the manuscript has interesting results that may be valuable to the atmospheric chemistry and aerosol communities, a major re-write is needed to make this publishable in ACP. Certainly, monthly and spatial distributions of NH3 emissions will be valuable – but not in its present form.

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 745, 2015.