

Interactive comment on “Development and uncertainty analysis of a high-resolution NH₃ emissions inventory and its implications with precipitation over the Pearl River Delta region, China” by J. Y. Zheng et al.

Anonymous Referee #1

Received and published: 16 February 2012

1 Overall Comments

This manuscript addresses an important issue air quality, ecosystems, and understanding the composition of the atmosphere. They present an ammonia emission inventory for China with high spatial resolution and a representation of temporal variability. This is an important step forward and is within the scope of “Atmospheric Chemistry and Physics”. However, I have very serious concerns about the quality of the analysis and the interpretation of the results. In order to gain acceptance, I strongly recommend that

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the authors make these substantial changes and improvements.

ACPD

1.1 Analysis of the trend in precipitation concentrations

About the precipitation samples – how frequently are the samples retrieved from the field and analyzed? Does the time in the field change the H⁺ balance? How accurately are the concentration of sulfate, nitrate, and ammonium measured? These are likely to be more stable than H⁺.

"In Table 7, the mean long-term trend of the ratio Ca²⁺+NH⁺₄ /SO₂⁻ 4 +NO₃⁻ 3 (NP/AP) was around 0.94 during 1998 – 2006, which was lower than that of some northern Chinese cities (e.g. Beijing) (Yang et al., 2004). This indicates that more inputs of alkaline species like NH₃ into the precipitation in Beijing than that in the PRD region."

I don't think you can make this conclusion, because many relevant cations are excluded from Table 7. Beijing also has more dust, while the coastal cities in the south likely have more sea salt.

I recommend removing this discussion of acidity and Figure 9, as they are a distraction from the main purpose of this paper which is ammonia.

1.2 Analysis of the temporal trends

I find Figure 8 very difficult to understand. After pondering this for a while, I determined that the blue line must be plotted on the left axis. This is very confusing. Please plot this as a stacked bar chart.

The temporal trends in fertilizer seem reasonable. But please provide more information about how this is calculated in the methods section. However, I'm not sure that the livestock monthly variability is reasonable. Is it based on consumption of livestock? This is what is described in the text, but we would expect livestock animal population to

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be out of phase with consumption, because the animals have to be raised for several months before they are consumed. The emissions should be more closely tied to temperature, which is the most important factor affecting volatilization. This is alluded to, but it is not clear how this is incorporated in these estimates. Please include a discussion of how the temporal variability is calculated.

1.3 Analysis of the trend 1998 – 2006

The conclusions state:

“A significant long-term correlation between agricultural NH₃ emission and pH value was observed reflecting the fact that the local NH₃ emissions influence the precipitation characteristics.”

This is not correct. I could find no such statistically significant correlation described in the results. Furthermore, looking at the data in Table 7 and Figures 11 – 13, there does not appear to be any trend in the cation ratio or NH₄⁺ in precipitation.

The analysis of the trend 1998 – 2006 is not useful and should not be presented. According to the emission estimates, there is relatively small trend in emissions over this period. According to the NH₄⁺ concentration data, there is no trend over this period. There are some very high values reported in 1999 and 2005. From this, it is not possible to conclude that there is a meaningful relationship between the measured NH₄⁺ at this estimate of NH₃ emissions. The best is that there is no real trend in either dataset. The authors should simply state this and move onto a different approach for the analysis.

There is however a substantial difference in winter and summer NH₃ emissions estimated by this inventory. Is it possible to segregate the winter / summer NH₄⁺ precipitation concentration? Do you see a similar winter / summer difference in the measurements? This would give us confidence that the inventory is capturing the seasonal

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variability.

There is some evidence that the spatial patterns in the NH4+ in precipitation is influenced by local NH3 emissions as quantified by this work. However, this relationship is weak (R-squared of 0.203). This is to be expected, though. The NH4+ in precipitation would be expected to be influenced by a broader area than just local emissions. Also – why are the data log-transformed?

To build confidence in the results, the authors should compare their emission inventory to surface measurements of NH3 in the gas phase, such as those recently published by Hu et al. (2008); Meng et al. (2010); Shen et al. (2011).

1.4 Characterization of uncertainty

I applaud the authors for characterizing the uncertainty. This is an important goal. However, the results from this characterization should be propagated through all of the results. An example is shown in Figure 2, but then single value, deterministic results are presented in all of the rest of the figures.

From your uncertainty characterization (Section 3.5) – what are the most important uncertainties? Where should efforts be focused for the biggest benefit? Some of the largest uncertainties are in sectors that contribute relatively little. The research needs are presented at the end, but it seems that these are based on data availability, rather than importance to reducing uncertainty.

2 Editorial Issues

Please address these editorial issues. I recommend an editor read the document for other errors I have missed.

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L24, P33735 “few works are reported regarding how ammonia emissions” consider “few studies have reported how ammonia emissions...”

L15, P33737 “examination of literatures” consider “previous studies”

L19, P33737 what is “scalper”?

L13, P33750 what is “dairy life”?

Table 1 convert from 6 columns to 2 columns to make this more clear.

Figure 8 There is a semicolon in the middle of “Agricultural” on the left axis.

references I was not able to find documentation on AuvToolPro. Please provide a reference or describe more completely.

References

Min Hu, Zhijun Wu, J. Slanina, Peng Lin, Shang Liu, and Limin Zeng. Acidic gases, ammonia and water-soluble ions in pm2.5 at a coastal site in the pearl river delta, china. *Atmospheric Environment*, 42(25):6310 – 6320, 2008. ISSN 1352-2310. doi: 10.1016/j.atmosenv.2008.02.015.

Zhao-Yang Meng, Xiao-Bin Xu, Tao Wang, Xing-Ying Zhang, Xiao-Lan Yu, Shu-Feng Wang, Wei-Li Lin, Yi-Zhen Chen, Yi-An Jiang, and Xing-Qin An. Ambient sulfur dioxide, nitrogen dioxide, and ammonia at ten background and rural sites in china during 2007–2008. *Atmospheric Environment*, 44(21–22):2625 – 2631, 2010. ISSN 1352-2310. doi: 10.1016/j.atmosenv.2010.04.008. URL <http://www.sciencedirect.com/science/article/pii/S1352231010002906>.

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