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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Role of ambient ammonia in particulate ammonium formation at a rural site in the North China Plain" *by* Zhaoyang Meng et al.

Anonymous Referee #1

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Overview

This paper as the other reviewer has pointed out potentially has an amazing dataset which is really needed for greater understanding of air pollution and its impacts in agricultural regions. The paper could be hugely improved by moving away from the gas-particle ratio analysis to more detailed atmospheric chemistry and physics which would allow insight into the processes occurring and whether current understanding of emission, transformation and deposition can explain the observations. There is an overuse of "the data suggests. . ." and "this indicates. . ." without backup of information.

Major comments:

Introduction:

I think the ambition of the paper (as described in the last paragraph) needs to be more detailed and then the critical analysis done in the paper.

P3 More details of instrumentation is needed, in particular the calibration and response time of the NH3 instrument is required. Did the authors see an influence on the response time from PM deposition on the inlet and instrument filters (see Bobrutski et al 2009 and other papers for details of this issue). Some raw data and calibrations would be useful – ACP is not figure limited. Though rainfall is mentioned as a key meteorological driver, the method of measurement and the data are not shown at all.

P 11 section 3.4: Relationship between ammonium and ammonia: This discussion is very brief and limited. In particular after noting previously (and probably correctly) that local ammonia emissions dominate the chemical speciation observed, the authors then infer "NH3 dominates NHx deposition". With the dataset they have they could have performed calculations of deposition vs emission over the 4 month period would have given much more insight, i.e. the process is bidirectional therefore it is uncertain whether any net deposition would occur under the ambient conditions. This is a missed opportunity to explore the atmospheric chemistry and physics of the system

Given that there are several thermodynamic models freely available, it would have been useful to explore the dataset against what is predicted by models. What is the ion balance – are dicarboxylic acids needed to explain aerosol neutralisation? (is it neutral?). Is the atmospheric chemistry at the site dominated by thermodynamic equilibrium or is there kinetic limitations on the processes? The authors have missed an opportunity with the dataset to fully understand the chemistry and rely in the results and discussions to discuss the ratios between gas and aerosol partitioning to explain scientific concepts which are known and therefore not surprising.

In the places in the manuscript which use atmospheric chemistry to explain data (e.g. 3.4.2), there are no calculations to check if what is observed is what would be expected under the conditions. Is ozone being lost to the surface or is there a haze which allows

aqueous processing in the atmosphere, what might be the role of organics. . .), what is the surface area of PM (given composition and RH) and hence can N2O5 hydrolysis explain the observations completely? There are lots of questions which are not touched upon, though they are key to understanding the role of NH3.

Having read the paper I am still not sure what the authors want a reader to learn from the gas-particle ratios. I would suggest the authors revise to include pollution/wind rose diagrams to look at the pollution footprint (e.g. ones are available on Open air and other packages), use current thermodynamics and kinetics of the system to see if current models would accurately represent the observations, if not what may be missing?

Oxidised nitrogen chemistry and the gas-aerosol partitioning dynamic are mentioned in passing but are key to understanding whether NH3 is driving the PM formation or it is a reservoir gas which grows PM when the presence of the other pollutants is there. Biomass burning is mentioned and K and CO as the indicators. With the dataset they could estimate the fraction of PM due to biomass burning, how much of the PM is explained by biomass burning, and does the biomass burning "seed" larger PM events. Finally the discussion and conclusion would be enhanced if some discussion about the impacts and potential solutions to the impacts. What is the evidence that limiting the NH3 emission would improve the air quality - it may well achieve this, but to make the case, evidence or hypothesis is needed to back the statements up.

Minor corrections:

P1 Line 22 The observation that NH3 drives NH4 content of PM is not new, so I do not think the word " suggesting" is appropriate

P1 Line 24: This is a percentage not a ratio.

P1 Line 25: use previous NCP abbreviation

P2 line 15: Actually most atmospheric chemistry text books discuss this, would cite them rather than research papers.

P5, line 19: asymmetric errors would be more appropriate given that one cannot have negative concentrations.

P25 Figure 2: given that it rained during the 4 months, why does the RH never go above 90%?

P27: for the PM composition it would be useful to have them as stacked so that one can see the variation of composition through time References: There are not many references from 2015 and 2016 despite many papers being published on this subject area. I would suggest the authors review the recent literature.

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