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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Effectiveness of Ammonia Reduction on Control of Fine Particle Nitrate" *by* Hongyu Guo et al.

Anonymous Referee #1

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This paper explores the sensitivity of NH4NO3 concentration to gas phase NH3 and NOx control for a number of contrasting locations and provides a comprehensive evaluation of the effectiveness of ammonia reduction on control of fine particle nitrate. The authors first developed a conceptual basis (S curve) to evaluate the effectiveness of ammonia control on partial nitrate through aerosol pH using a thermodynamic model ISORROPIA-II. Then, they use observation data to calculate the aerosol pH and the S curve for contrasting locations in Netherlands, US and China, and assessed the effectiveness of ammonia reduction in those places. More comprehensive simulations are conducted to investigate the sensitivities of pH and nitrate partitioning to NH3 concentration, as well as the effectiveness of NH3, NOx and SO2 control in reducing fine particle mass for contrasting locations and in different seasons. The authors conclude

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Discussion paper

that NH3 emissions control would be only be effective in reducing PM2.5 mass when ambient particle pH drops below approximately 3.

The question about how effectiveness is ammonia reduction on air quality amelioration is an area of active research, and the paper adds new results to the literature. In particular, I am very impressed by the conciseness of the S curve and the way it links up the control effectiveness with relating factors. I think this paper fits well into the scope of ACP and will interest its readers. In general, this paper is well-written, and I recommend it to be published in ACP after the following weakness/questions are addressed:

Major comments:

1. Pg 3, line 19: The authors offered clear explanation for the decreasing tendency of SO2 and NOx, which is a result of regulation. However, it seems less clear to me why NH3 is increasing, although the authors have tied NH3 emissions with population growth previously. It would be better to explicitly state that the increase of NH3 emissions is due to the increase of farming activities and fertilizer applications, in order to support the growth of population. I would also suggest adding something about the potential increase of ammonia emission due to global warming, such as the study of Skjøth and Geels 2013. Skjøth, C., and Camilla Geels. "The effect of climate and climate change on ammonia emissions in Europe." Atmospheric Chemistry and Physics 13 (2013): 117-128.

2. Pg 5, line 5. “With high NH3 concentration, it is somewhat representative of northwestern Europe.” I would suggest the authors to provide additional evidence for this claim. Perhaps, some reference which indicate that northwestern Europe is normally have high NH3 concentration. Or, maybe provide the averaged NH3 concentration value on northwestern Europe and compared it with the averaged NH3 concentration in Cabauw.

3. Pg 6, line 15. “Inorganic ions are also assumed to be only in the aqueous phase.”

Does the model assume that all aerosol species are in the aqueous phase or it also consider some of the species in solid state? Please clarify.

4. Pg 6, line 29. Other studies show that existence of organic phase could also impact the NH3 and NO3 partition as some SOA could react with NH3 and reduce the NH3 concentration. Add comments.

Zhu, S., Horne, J.R., Montoya-Aguilera, J., Hinks, M.L., Nizkorodov, S.A. and Dabdub, D., Modeling reactive ammonia uptake by secondary organic aerosol in CMAQ: application to continental US.

5. Pg 7, line 4-5. The authors used two "discussed below" in this sentence. It would be better to give the exact section or location of the discussion instead. Does it refer to the first paragraph of 2.3?

Actually, there is research showing that different mixing assumption could have significant impact on NO3- and NH4+ partition, especially on NO3-:

Zhu, S., Sartelet, K., Zhang, Y. and Nenes, A., 2016. ThreeâĂŘdimensional modeling of the mixing state of particles over Greater Paris. Journal of Geophysical Research: Atmospheres, 121(10), pp.5930-5947.

6. P11, line 15. The authors should provide more details regarding to the nature of "particle artifacts in the gas collection system" that is affecting the measurement of HNO3 and HCl.

7. Pg 17, line 19. Since the calculations are based on site measurement in this study, does it suggest that the pH calculated here is closer to the reality than the one calculated by Pozzer et al., (2017). Or, on the other hand, is it possible that the measurements site is not representative enough for the larger domain used in the global model calculation due to its coarse resolution? Are there any regional simulation results that is consistent with the pH prediction presented here?

8. Pg 18, line 13-14. This conclusion looks not very convincing to me. Since the particle

composition is so different between SE US and NE US, the author should justify how the SE US could be a representative case for the eastern US in the summer, and how the NE US could be representative case for the eastern US in the winter before drawing such a conclusion. Or latest explain the cause of such a high sulfate composition (76%) in the SE US case.

Minor Comments:

Pg 5, line 13, the word “alternatively” here is confusing. Do you mean it is the first hour measurement is for PM1 and the next hour will be for PM2.5? In that case the measurement interval will be 2 hours for either PM1 or PM2.5, is that the case? Please clarify.

Pg 7, line 9. “In Cabauw, it has been reported . . .”Could reference be provided for this report?

Pg 7, line 25. It would be better to specify the “coarse mode salts” that HNO3 evolved into.

Pg 9, line 14. “0.987x10-14 is a unit conversion factor” I would better to specify which units are being converted with this factor.

Pg 10, line 3. Could the authors be more specific on how the “approximately 0.6” non-ideality shifts are calculated? Or provide a reference S curve without the non-ideality effect?

P11, line 17. Could the authors provide the references for those “previous studies” mentioned here?

P12, line 2. Could the authors provide the exact hour ranges used in this study to define “night” and “daytime”?

Pg 12, line 13. I found it confusing that the authors keep changing between “NE US” and “WINTER” for the Guo et al., (2016) case, for example, “WINTER” is used in Figure

2, but “NE US” is used here in the text. I suggest the authors use more consistent expression.

Pg 19, line 4. The previous discussions in this paragraph are based on Cabauw winter and Beijing, while the 19% ε(NH4+) value used here are from one-year Cabauw, would you explain why?

Pg 20, line 12. What does “further from the actual ambient particle pH” referred for? Do you mean the region 2 of the curve is further from the ambient particle pH?

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