

Interactive comment on “Modelling the impacts of ammonia emissions reductions on North American air quality” by P. A. Makar et al.

Anonymous Referee #2

Received and published: 27 March 2009

This paper presents the results of modeling studies analyzing the effect of ammonia emission reductions on ambient concentrations of PM_{2.5} and its chemical precursors, for a one-year period over North America, and considering two different ammonia reduction scenarios. Ammonia has been given perhaps too little attention in the scientific literature and as a management strategy for addressing PM, so I applaud the authors for choosing to address this topic. Their finding that ammonia emission changes affect PM concentration over a larger area than they affect ammonia concentration is an interesting one, supported by their analysis.

The modeling results appear to be sound, or at least I don't see indications to question whether they are comparable to the current state of the art. The paper is within the scope of ACPD, and is well written. However, I think that the authors are missing an op-

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portunity to explain better why they see the results they do, in terms of the fundamental processes. I think there is too little analysis of the simulations that are conducted, and either the authors lack a detailed understanding of the processes involved in inorganic aerosols, or they have not done a good job of communicating them.

Because the modeling results appear to be good, I judge this paper to be publishable with minor revisions. However, I would also encourage the authors to go beyond the minimum required for publication and to explore more fully the processes involved in, and explanations for, their model results.

In particular, I don't think that the paper gives a clear answer to how the different components of PM respond to the change in ammonia emissions. Does PM decrease because ammonium, nitrate, sulfate or some other components decrease? Important here is the concept of "ammonia limitation" which deals with gas-aerosol chemistry. There is a discussion of ammonia limitation on p. 5374, and the concept of ammonia limitation comes back later. What is not clear from the presentation is what is being limited by the lack of ammonia. I also think that the authors have oversimplified the effects of changes in ammonia on PM by referring to "ammonia limitation" as they do. In fact, I suspect that the picture of the response of inorganic PM to changes in ammonia is more complicated than suggested by the simple "ammonia limited" versus "non ammonia limited" designations. In particular, this picture is more complicated where nitrate aerosol is high, and where other inorganic components are also in play.

The authors could divide their thinking into two steps: what is the response of gas-aerosol chemistry to changes in ammonia, and second, given those changes in the chemical forms of gases and aerosols (particularly for ammonia and nitrate), what is the effect on the lifetime and concentrations of the different species given different removal rates. The responses of the inorganic chemistry are discussed at length in several papers that the authors do not reference - although the focus is on changes in sulfate rather than ammonia, similar concepts would apply - Ansari and Pandis (EST, 1998), West et al. (JAWMA, 1999), and Blanchard et al. (JAWMA, 2000). Papers by R.

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Pinder and colleagues (e.g. EST, 2007) then apply this in a similar model framework as the current paper.

On p. 5374, I feel that the authors explain this system insufficiently. More importantly, they present effects on PM_{2.5} without exploring fully the changes in particulate sulfate, nitrate, and ammonium. While they do present changes in ratios (e.g., figure 12), I find these insufficient to understand the process. A more clear way to present the results might be through separate tile plots for the change in PM_{2.5} sulfate, nitrate, and ammonium (all in ug/m³).

That is, the conceptual model in Fig. 16 can be tested by showing how the relative abundance of the different species changes from the top picture to the bottom in a quantitative way, also analyzing how that picture might differ in different seasons or regions of the country. Currently, the conceptual model in section 4.2.6 is presented in a vacuum, without any quantification and without any reference to the model simulations or results presented previously. It might be correct, but the point is that there is an opportunity to test this model that is missed here.

In this spirit, in Fig. 7 and 8, the changes of 5 ug/m³ at the high end of the distributions are huge, far exceeding the median and probably accounting for much of the total PM_{2.5}. Some explanation of whether these large changes are possible, and under what conditions they result, would help. Likewise, although increases in PM_{2.5} are rare, can the authors provide a theoretical basis why this would happen? In Fig. 3, why do we see these seasonal and spatial patterns in ammonia/sulfate? Part of the reason would be changes in emissions - what else is going on?

Specific comments:

Table 1 - the term "PM_{2.5}-NH₄" has not been used elsewhere in the paper and should be defined. I thought you were subtracting NH₄ from PM_{2.5}, but then realized you're using it like p-NH₄.

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Fig. 2 and similar 4-panel figures. I think it would help to label the panels on the figure (JJA) rather than in the caption, and remove the cryptic labels on the figures.

Fig. 3 - the color scale is labeled "M8 mole"?

Fig. 8 should be stretched horizontally - it's really small.

p. 5394, line 25 - it would help to say Figures 7 and 8 rather than the "frequency distribution figures".

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 5371, 2009.

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