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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Global simulations of nitrate and ammonium aerosols and their radiative effects" *by* L. Xu and J. E. Penner

Anonymous Referee #1

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In this manuscript, Xu and Penner applied the hybrid dynamic method, in which the mass transfer to the fine aerosol is simulated using the bulk equilibrium assumption and to the coarse aerosol using a dynamic approach, into a global chemistry transport model in order to estimate the global annual budgets of ammonium nitrate and nitric acid and their direct and indirect radiative forcing. The paper is, in general, well written, and contains relevant scientific information that will add to the current level of understanding the aerosol climatic effects. I recommend the paper for final publication, after some issues are addressed.

General Comments:

1. Page 10117, lines 5-6 and Page 10120, lines 3-4: The authors state that thermodynamic equilibrium overestimates the portion of ammonium nitrate in coarse mode (and

underestimates the fine mode). Nevertheless, later in the text (page 10118, lines 23-25) they say that equilibrium method neglects differences in the chemical driving forces of the specific aerosol size bins. This means that during equilibrium most of ammonium nitrate will condense in the fine mode, which has most of the surface area, resulting in an underprediction in the coarse mode (and overprediction of the fine mode). In fact, Capaldo et al. (2000) and other studies (i.e. Karydis et al., 2010; Karydis, Tsimpidi, et al., 2011) have reported an underprediction of nitrate in the coarse mode (and overprediction of fine mode), using the equilibrium method, and a significant increase on coarse nitrate after using the hybrid approach (and a decrease of the fine mode). With that said, I think it would be useful to identify the source of this discrepancy between these studies and the Feng and Penner (2007) study (and presumably the current study too), and include a discussion that explains why the use of hybrid approach decreases the ammonium nitrate in the coarse mode in contrast to the Capaldo et al. (2000) and Karydis et al. (2010; 2011) findings.

2. Section 3.4: I believe that there are two important issues that are needed to be addressed in this section. 1/ How much the performance of the model improved, compared to the previous study (FP07), by the improvement of the sulfate aerosol dynamics? and 2/ why nitrate is so seriously overpredicted over North America? The latter is also discussed in FP07 study but since the main focus of the current paper is the ammonium nitrate, I think it would be helpful to include a short discussion here as well.

Specific Comments:

1. Page 10116 line 14: I suggest to add a short explanation on how nitric acid contributes on indirect radiative forcing. For example, the statement can be rewritten as: "almost all of which is due to condensation of nitric acid gas onto growing droplets (-0.08 W m-2)".

2. Page 10117 lines 15-19: Please include a short comment on why this will be the case. I assume that after the reduction of SO2 emissions, the free NH3 in the atmo-

sphere will be increased resulting in an increase of ammonium nitrate formation.

3. Page 10119 lines 3: I would rephrase the statement to: “because it takes from several hours to up to a week to reach equilibrium”. What is important here is that, at any case, it takes more than an hour (which is the time step of the model) to achieve equilibrium.

4. Page 10122, first paragraph: How did you choose the prescribed background size distributions? Are they based on measurements? If so, please add the corresponding citations.

5. Page 10131, line 6: replace “though” with “through”

6. Page 10133, lines 8-9: Please add references for the aerosol hygroscopicities (especially for organic mass and dust as they seem to be in the high end of the reported values in literature).

7. Page 10136, lines 16: Why for clear-sky conditions there is no heating over the biomass burning regions of central Africa?

8. Page 10138, lines 24: The CDN concentrations are predicted high over the southern remote oceans (more than 100 cm-3) and slightly low over the polluted regions (less than 500 cm-3). How are the predictions of the model compared to other published global CDN fields (i.e. Penner et al., 2006; Merikanto et al., 2010; Chen et al., 2010; Karydis, Kumar, et al., 2011)?

9. Page 10139, line 28: Why the additional HNO3 gas in the TN case results in a significant negative response of sulfate in Northern America?

10. Page 10142, line 8: replace “frame work” with “framework”

11. Page 10143, line 16: Add “at” before the “surface”

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