Reply to review comments by anonymous referee #2 on "Global simulations of nitrate and ammonium aerosols and their radiative effects" by L. Xu and J. E. Penner

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We appreciate the comments of this reviewer, which have allowed us to improve the manuscript and clarify ambiguities. We address each comment below. The reviewer's comments are in bold followed by our response.

1. It is interesting that the first indirect forcing of nitrate (-0.09 W m^{-2}) is mainly from nitric acid gas (-0.08 W m^{-2}). On the other hand, I notice that there is another paper on ACPD (Makkonen et al. ACPD, 2012) that shows a much larger indirect effect (-0.46 W m^{-2}) due to nitric acid gas. I understand that the discrepancy can be largely due to the different parameterization of nitric acid on cloud activation in two models, but this indeed deserves some insightful discussion.

Reply: The first indirect forcing of total nitrate (-0.09 W m^{-2}) refers to the forcing of anthropogenic nitrate (gas plus aerosol) changing from the pre-industrial to the present-day condition while the indirect effect (-0.46 W m^{-2}) reported in Makkonen et al. (2012) is actually the effect of present-day nitric acid gas and refers to the forcing difference in the simulation with and without nitric acid gas. Thus, these two values are not comparable in terms of the definition of indirect forcing. The global present-day total nitrate (gas plus aerosol) effect from Makkonen et al. can be compared to our simulation with and without nitric acid gas and aerosol, which was estimated to -0.23 W m^{-2} (see Figure 16 top panel). This large difference between our calculation and Makkonen et al.'s could result from not only the different parameterizations of the effect of nitric acid gas on aerosol activation but also on differences in the simulated nitric acid mass concentrations (which can differ significantly between different atmospheric chemistry and transport models). Although HNO_3 variations might be smaller or larger than those for NO_2, it has been documented that there are factor of two or larger variations in the calculation of tropospheric NO_2 column densities for different regions (i.e., Eastern U.S., Europe, East Asia, Africa, South America) (van Noije et al., 2006). A discussion of the results of Makkonen et al. will be added to the paper, but we cannot understand the cause of the differences, since the study of Makkonen et al. (2012) did not provide a global budget and burden of nitric acid gas.

2. I am also curious about how well this model simulates nitric acid. In my understanding, nitric acid gas is often overestimated in some models.

Reply: The simulated nitric acid is close to that in the previous study (Feng and Penner, 2007) since similar NOx emissions were used as well as the same simple nitrogen chemistry mechanism. Comparing the nitric acid mixing ratio near surface with two other global model studies (Adams et al., 1999; Liao et al., 2003), the nitric acid gas in the present study shares a similar spatial pattern as these two studies though the global annual average nitric acid gas is about 60% and 36% lower than Adams et al. (1999) and Liao et al. (2003), respectively. In addition, the annual average tropospheric burden of nitric acid gas of 0.30 Tg N estimated in this study is about a factor of 3 and 2 smaller than that in

Bauer et al. (2007) (0.86 Tg N) and Pringle et al. (2010) (0.55 Tg N), respectively. We will add this discussion in the revised manuscript.

3. Please comment on why the indirect effect of nitric acid gas is most significant over the coastal regions. Is it because of the high concentrations of nitric acid, sea salt aerosols, or other things?

Reply: The large anthropogenic indirect forcing of nitric acid gas over the coastal regions is due to changes in the number of cloud droplets associated with sulfate, fossil fuel and biomass burning aerosols when nitric acid gas is added (see Figures 12 and 13). The reasons for these differences are discussed during the discussion of these figures, but will be made clearer, and will be referred to when the indirect effect of nitric acid gas is covered.

4. I agree with reviewer #1 that, the authors should show how the model simulation is improved compared to Feng and Penner (2007) study, by incorporating the sulfate aerosol dynamics. Any comment on this would be useful.

Reply: Compared to the study of Feng and Penner (2007) that assumed an internally mixed chemical composition within each size bin (i.e., in total 4 size bins for the radius from 0.05 to 10 um), we examined the interaction of nitrate and ammonium on the 11 externally mixed pre-existing aerosol populations listed in Table 2. The fundamental framework of the aerosol model (i.e, Liu et al., 2005) used in this study is more realistic than that in the Feng and Penner (2007) study that only simulates sulfate mass. We will add the following discussion in the revised manuscript.

"Compared with the previous study (FP07), this work predicts a 10% and 4% lower burden of nitrate and ammonium, respectively, higher total sources and sinks and therefore shorter lifetimes by 25% and 18% for aerosol nitrate and ammonium due our different treatment for the interaction between nitric acid and ammonia and pre-existing aerosols as well as different aerosol deposition schemes (since our scheme assumes external mixtures and FP07 do not)." A detailed discussion of these differences is reported in Xu (2012)."

5. Is it possible that the overestimate of nitrate over North America is partly due to ignoring organic nitrates in the model?

Reply: We are not sure if the overestimation of nitrate over North America is partly due to ignoring organic nitrates in the model. However, one possible reason, as stated in Adams et al. (1999), is that particulate nitrate may evaporate from the measurement filter packs, resulting in a low bias for the values from the measurements (Adams et al., 1999; Pakkanen et al., 1999). We will add this short discussion in the revised manuscript.

6. Page 10122 Line 13, "Table 1" should be "Table 3".

Reply: We will change from "An assumed size distribution (Table 1, Liu et al., 2005) is used…" to "We used an assumed size distribution (see Table 1) following (Liu et al., 2005) within each size bin…"

Reference:

van Noije, T. P. C., Eskes, H. J., Dentener, F. J., Stevenson, D. S., Ellingsen, K., Schultz, M. G., Wild, O., Amann, M., Atherton, C. S., Bergmann, D. J., Bey, I., Boersma, K. F., Butler, T., Cofala, J., Drevet, J., Fiore, A. M., Gauss, M., Hauglustaine, D. A., Horowitz, L. W., Isaksen, I. S. A., Krol, M. C., Lamarque, J.-F., Lawrence, M. G., Martin, R. V., Montanaro, V., Müller, J.-F., Pitari, G., Prather, M. J., Pyle, J. A., Richter, A., Rodriguez, J. M., Savage, N. H., Strahan, S. E., Sudo, K., Szopa, S., and van Roozendael, M.: Multi-model ensemble simulations of tropospheric NO_2 compared with GOME retrievals for the year 2000, Atmos. Chem. Phys., 6, 2943-2979, doi:10.5194/acp-6-2943-2006, 2006.

Xu, L.: Global simulation of nitrate and ammonium aerosols and their radiative effects and comparison of satellite-based and modeled aerosol indirect forcing, Ph.D. thesis, Department of Atmospheric, Oceanic and Space Sciences, University of Mcighian, Ann Arbor, 306 pp., 2012.