

Interactive comment on “Simulation of nitrate, sulfate, and ammonium aerosols over the United States” by J. M. Walker et al.

Anonymous Referee #2

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Walker et al. aimed to explain model biases of nitrate aerosol concentrations over the US (underestimates in California, and overestimates in the East and Midwest) using the GEOS-Chem chemical transport model. They examined the uncertainties in ammonia emissions by comparing with satellite measurements, and also tested the nighttime nitric acid formation by heterogeneous N_2O_5 hydrolysis. They showed reducing the N_2O_5 uptake coefficient did not explain the nitrate overestimates in the East and Midwest, while the underestimates in California is likely due to underestimates of ammonia emissions. The study is well conducted and well fits the scope of ACP. A similar study was conducted independently at the same time by Heald et al. (ACPD 2012; cited in the manuscript). Heald et al. (ACPD 2012) focused on explaining the model nitrate overestimate in the East and Midwest, and this study focused on the underestimate of nitrate aerosol in California.

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My main comment on the manuscript is whether it can achieve a more quantitative conclusion. The manuscript conclude that the underestimate of nitrate aerosol concentrations in California is likely due to underestimate of ammonia emissions. It does not answer how much increase in California ammonia emissions is needed to reproduce the measured nitrate concentrations, and whether the increase is consistent with constraints from satellite measurements. I suggest add some discussions on it and also discuss other possible factors that might contribute to the model bias.

I have a few more comments listed below:

Specific comments:

1. Page 19501, Introduction: I do not think that the Introduction section provides enough background knowledge for this study. More sentences on our current understanding of aerosol simulation (capability and uncertainty), and understanding of precursors emissions in particular ammonia emissions over the US would help readers to understand the context better.

2. Page 19503, Line 17: This paragraph and Fig. 2 shall cite Evans and Jacob (GRL 2005) that describes the parameterization of N_2O_5 hydrolysis in GEOS-Chem.

Evans, M. J. and Jacob, D. J.: Impact of new laboratory studies of N_2O_5 hydrolysis on global model budgets of tropospheric nitrogen oxides, ozone, and OH, Geophys. Res. Lett., 32, L09813, 2005.

3. Page 19504, Line 12-14: Does GEOS-Chem only simulate fine particulate matter of sulfate, ammonium and nitrate? Please check Pye et al. (JGR 2009) (first paragraph of section 2.3) that "ISORROPIA II is implemented in GEOS-Chem to compute gas-aerosol equilibrium partitioning of nitric acid and ammonia. Particles in this study are not size-resolved; however, they can be generally assumed to represent $\text{PM}_{2.5}$ since formation of sulfate-nitrate-ammonium on coarse mode sea salt and dust is excluded."

Pye, H. O. T., et al.: Effect of changes in climate and emissions on future sulfate-

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nitrate-ammonium aerosol levels in the United States, *J. Geophys. Res.*, 114, D01205, 2009.

4. Page 19505, Line 11-12: Model sulfate aerosol concentrations are biased low by 34

5. Page 19505, Line 25-27: Can you discuss other possible factors that may contribute to the nitrate overestimates in the East and Midwest in the model?

6. Page 19506, Line 2: Does Table 3 show the biases in the annual concentrations or the averaged normalized mean biases in the monthly concentrations? The comment also applies to the title of Table 3.

7. Page 19506, Line 22-25: Can you explain the statement "either the thermodynamic partitioning between the gas and aerosol phases or the removal of atmospheric HNO₃ is the limiting factor"? In the following discussion, you also attributed the limiting factor to the availability of ammonia.

Fig. 9 also shows there is a large fraction of gas-phase ammonia at both California sites. Is it inconsistent with the conclusion of limited availability of ammonia? Would the model underestimate of ammonium aerosol be in part caused by the underestimate of sulfate?

8. Page 19508, Section 4.2: The comparison with IASI measurements of ammonia column seems not consistent with Heald et al. (ACPD 2012) that I think follows the appropriate way to compare with satellite measurements. In their manuscript (Fig. 4) they applied the IASI retrieval averaging kernel and a priori to the GEOS-Chem simulation and showed significant changes in the model results. Their Fig. 4 showed that GEOS-Chem ammonia column concentrations were much higher than IASI before applying the averaging kernel and a priori over the US Midwest. That reflects either IASI a priori profiles are very different from GEOS-Chem profiles or the averaging kernel is not uniform vertically. This is not consistent with Fig. 11 in this manuscript that directly comparing the GEOS-Chem model results with IASI measurements. Please check.

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9. Page 19525, Fig 10: It is not clear to me that in Fig. 10 plotting the hourly time series is very helpful. What drives the variability in the hourly nitrate time series? Does the ammonia emissions in the model have a hourly resolution? Would it be more helpful if reducing the resolution to daily and over-plotting the measurements for comparison?

10. Page 19517: In the legend of Fig. 2, the red dashed line should be "Sulfate, T > 282K".

11. Page 19522: Fig. 7 is not clear to read. I suggest make the symbols thicker or add lines over the symbols, and also make the labels of x- and y-axis larger.

12. In the captions of Fig 6, 7, 8, and 9, please state the comparison year 2009.

Interactive comment on *Atmos. Chem. Phys. Discuss.*, 12, 19499, 2012.

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