

Interactive comment on “Nitrate aerosols today and in 2030: importance relative to other aerosol species and tropospheric ozone” by S. E. Bauer et al.

S. E. Bauer et al.

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We would like to thank the three reviewers of our manuscript, for their thoughtful comments. Many of the reviewers comments lead to changes in the manuscript, but we would like to discuss some comments in this reply.

Model emissions: Methane emissions are prescribed in this model run, this is said in the model description, but it was misleading to have the methane emissions listed in the emission table. We removed that part from the table. We used the EDGAR emissions for the year 2000 and the Streets et al 2003 projection for the A1B 2030 simulation. Therefore NO_x, SO₂, NH₃, BC/OC emission changes are all consistent with each other including biomass burning emissions. The original manuscript cited a wrong data

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sets. One reviewer raised the question about the difference between prescribed and interactive DMS emissions. Regarding DMS emissions, a more complete description is given in Koch et al., 2006: "As in the work of Koch et al 1999, we base our DMS emissions on the sea-water DMS concentrations of Kettle et al. [1999]. However, we now make use of the sea-to-air transfer function of Nightingale et al. [2000], together with model generated surface (10 m) windspeeds. Since the dependence of the flux on the square of the windspeed is increased, our DMS emission (see Table 1 in Koch et al 2006) is approximately double that in the work of Koch et al 1999. We use this larger emission in order to increase the model sulfate production in remote regions closer to the observed levels." In Koch et al 1999, Liss and Merlivat 1986 for sea-air DMS transfer is used instead of Nightingale in order to increase DMS/sulfate in remote regions. We refer to those studies regarding the effect of DMS emission parameterization on global modeling.

We decided to perform the sensitivity experiments around the year 2030, because like that we could study which of the expected changes in the various emitted species most strongly effects nitrate aerosol formation.

The question was raised, how does the solubility/insolubility of mineral dust would leads to more wet deposition of dust? Several processes are considered to calculate the wet removal of aerosol in our GCM. First the aerosol dissolves in cloud water. This process is proportional to the tracers solubility. However we assume uncoated mineral dust to be insoluble. The deposition of hygroscopic material like sulfate or nitrate will transform the hydrophilic dust into a soluble particle, and hence increase the wet deposition of mineral dust. This process is studied in more detail in Bauer et al, 2005 and 2007.

One reviewer commented: "The radiative effect of nitrate and sulfate in coarse mode are not taken into account, the paper mentioned that is due to the coarse nitrate are attached to mineral dust particles. " We published a whole paper on that issue Bauer et al. 2007: Do sulfate and nitrate coatings on mineral dust have important effects on radiative properties and climate modeling? Where we demonstrated that the optical

properties of coated dust particles only significantly change compared to uncoated dust, if coating layers are thicker than 10% in respect to the particles radius. Coating layers were much thinner in our simulation as well as in the observational data sets we found in literature. Therefore Bauer et al 2007, concludes that coated dust particles can be treated as external mixtures for radiative calculations, and the coating material has negligible optical effects on the global scale.

One reviewer raised the question if the dramatic underestimation of nitrate aerosol in East Asia could be partly caused by the transport scheme. Yes, that is of course always a possible factor of uncertainty, however the huge underestimation, by an order of magnitude that is not only seen in comparison to the few surface observation, but as well as compared to the aircraft measurements, that cover quite a large special range over and downwind of East Asia, let us conclude that there is a dramatic lack of nitrate material lacking in our simulation in East Asia. Such huge differences must be mainly caused by incorrect emission sources in that area.

The following comment was made by a reviewer: “These results give a strong indication that including heterogeneous dust chemistry clearly improves the simulated nitrate concentrations. I am just curious that how important this effect to sulfate is? Since many models haven’t included this effect yet.” Bauer et 2005: Impact of heterogeneous sulfate formation at mineral dust surfaces on aerosol loads and radiative forcing in the Goddard Institute for Space Studies general circulation model’, exactly studied that question, and comes to the conclusion that heterogeneous reactions on dust surfaces strongly impact sulfate aerosol simulations. However, as H₂SO₄ is a condensate it is very likely to deposit on various aerosols present in the atmosphere, therefore this interaction is not limited to dust surfaces.

“In several campaigns, the modeled total nitrate (red line) is significantly higher than fine mode of ammonium nitrate (black dotted line). What could be the possible sources and/or processes contributing such a significant coarse mode ammonium nitrate, even up to very high altitude in a few campaigns?” Yes, that is an interesting question we

would like to answer. The paper gives the results of our simulation, showing that compared to the PEM tropics data sets coarse mode nitrate provides the major contribution of nitrate in the upper troposphere. PEM tropics, measured the air over the South Pacific, which shows very low nitrate concentration. The dust load in these regions is very low, however we assume that nitrate material sticking to background concentrations of dust is still dominating the nitrate aerosol mass in that region. Only detailed size and composition resolving measurements would allow us to verify those results.

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