

Interactive comment on “Sensitivity of nitrate aerosols to ammonia emissions and to nitrate chemistry: implications for present and future nitrate optical depth” by F. Paulot et al.

Anonymous Referee #1

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In this study, the authors describe an updated version of the nitrate aerosol scheme included in the GFDL atmospheric model, evaluate that scheme against observations, and analyse sensitivity simulations where emissions and chemistry options are varied.

The paper is interesting and well-written. Figures illustrate the discussion well. The authors give an exhaustive view of the nitrate modelling capability of their model, and demonstrate deep understanding of the model and its sensitivity to changes. It is particularly useful to give budgets. I recommend publication after minor revisions are made to address the following comments.

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1 Main comments

- Like most other nitrate modelling papers in the literature, the authors do not discuss and evaluate the diurnal cycle of nitrate aerosols in their model. It is unfortunate because the ability of nitrate aerosols to dissociate back to the gas phase means that their diurnal cycle is quite pronounced (Dall’Osto *et al.*, 2009). It is important to reproduce that diurnal cycle well in a model because when nitrate is in the gas phase (which includes most daylight hours), it will not exert radiative effects and forcing by interacting with radiation. The authors may not have the diagnostics required to evaluate the diurnal cycle, but may have looked at it in another context and could discuss it briefly in the paper.
- In an otherwise rigorous modelling of nitrate aerosols, the authors are surprisingly lax about nitrate aerosol optical properties. They are simply taken as identical to sulphate aerosols (page 25743, lines 20–23). Why not do things properly? Refractive index datasets are available, as are hygroscopic growth curves. The authors state that using sulphate optical properties yields errors of 20% in extinction (page 25743, line 22). How was that estimated? The paper should clearly state that that 20% is a big number compared to the sensitivities in emissions and chemistry explored later. As such, the paper would have benefited from including nitrate optical properties in the sensitivity analysis.

2 Other comments

Page 25751, line 11: The quality of the simulation of precipitation rates will also matter.

Page 25753, line 16: Using sulphate aerosol hygroscopic growth curves may also be a problem.

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Page 25758, line 20 to Page 25759, line 9: Those changes are due to changes in the strength and location of emissions alone. It would be useful to speculate on the effect of climate change as well. Climate change would presumably increase transport to the free troposphere, thus making NH₃ limitation bite even more than suggested by those simulations.

3 Technical comments

- Page 25742, lines 9–11: Several acronyms to define here: ACCMIP, IPCC, and AR5.
- Page 25747, line 22: “The volatilization of NH₄NO₃”.
- Page 25756, line 17: Delete extra comma.
- Page 25758, line 14: Missing space in “surface NH₄NO₃”.
- Figure 2: The black cross indicating the location of Bondville is not easy to see.
- Caption of Figure 8: Delete repeated word “the”.

4 References

Dall’Osto, M., Harrison, R. M., Coe, H., Williams, P. I., and Allan, J. D.: Real time chemical characterization of local and regional nitrate aerosols, *Atmos. Chem. Phys.*, 9, 3709-3720, doi:10.5194/acp-9-3709-2009, 2009.

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