

Interactive comment on “Modelling of nitrate particles: importance of sea salt” by G. Myhre and A. Grini

Anonymous Referee #2

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General The paper describes an interesting experiment on nitrate formation on the global scale. Although the subject is of general interest for ACP readers, there are a number of issues that need significant improvement. My main comments are associated with validation, the influence of kinetics (vs thermodynamics) and the influence of the model resolution on the results. Further, I would encourage the authors to let a native speaker check the paper on English language.

Comments The validation of the model results is very limited and not presented in a quantitative way. To supply the reader with a sense to which extend reality is reproduced by the model, a comparison of modelled (total) nitrate, (total) ammonium and sulphate concentrations to the data from the network in the US and Europe should be

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given and discussed. The comparison should be given for both the simulation with and without sea salt to see the (large) influence of sea salt in comparison to observations. For Europe, a host of valuable data are available for total nitrate and total ammonium in the EMEP network.

The authors use the EQSAM module for thermodynamic calculations. This module describes the thermodynamics in an appropriate way and the choice for this fast scheme does not affect the results much as it is very close to other schemes like Isorropia. The introduction of two size ranges (sulphate and sea salt dominated) for the calculation of fine and coarse mode nitrate is not perfect but appropriate as fine and coarse mode particles are by definition externally mixed. The authors indicate that mass transfer is important but do not implement it. The issue of mass transfer (kinetics) is probably most important in coastal areas where fine and coarse mode nitrate co-exist. The chosen approach to apply the equilibrium module to the coarse mode using the excess nitric acid, effectively transfers the excess nitric acid to the coarse mode. Hence, the equilibrium assumption causes the “kinetics” to be a function of the time step used in the study as each time step the available ammonium nitrate supplies the nitric acid in the gas phase which is then effectively transferred to the coarse mode (in the presence of enough sea salt). To my opinion the inclusion of kinetics for the coarse mode should be discussed. As there is a large discussion in literature whether or not the quenching of nitric acid by sea salt has a limit (kinetically) it would be interesting to learn the authors opinion about the time scale involved and how it would influence the nitrate partitioning in coastal areas?

The role of ammonia in this study does not receive a lot of attention, although the treatment of ammonia has a large influence on the results for nitrate. I agree with the authors that the seasonal cycle of ammonia should not be weighted with the sunlight hours to the power 4. In my opinion the power 4 relation by Adams et al was indeed tuning. Regional models use seasonal cycles which reflect to some extent a scaling with sunlight hours and are able to reproduce the amount and seasonal cycle of ammo-

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nium nitrate. Tests reveal that using different seasonal cycles may have large effects (up to a factor 2) on seasonal average nitrate levels. The background for the power 4 relation was probably the coarse model resolution, and associated underestimation of NH_3 and NO_3 formation in summer, used in the study by Adams et al (2001). The authors also use a coarse resolution model and may have similar problems (not clear in the paper). Hence, a discussion on the influence of the model resolution and the uncertain ammonia emissions and their influence on the results seems appropriate (see also next point).

1463 26-29: These statements indicate that the authors feel that it is not appropriate to model nitrate distributions in a global model. As stated above the resolution effects and the validity of the resolution used should be discussed in the paper.

1461 fine mode nitrate is also greatly influenced by relative humidity. 1461 the excess ammonium is an assumption in the thermodynamical module. Small amounts of nitric acid may dilute into the aerosol without excess ammonium.

1463 line 5: the model represents rural areas. Hence, the gradients between rural and natural areas can be described, not the urban-rural ones. For rural to natural the statements are valid. For urban to rural a large number of studies show that the gradients of primary BC en OC are larger than for nitrate. The observation that nitrate is much higher in urban areas than in rural areas is probably biased due the locations of the sites in the Putaud et al publication and to my opinion not valid for European conditions.

1465 13 The definition of NP appears to me to be the fine mode nitrate concentration in molar units. Is that so? If so, defining NP does not make a lot of sense to me

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