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Interactive comment on “A global model simulation of present and future nitrate aerosols and their direct radiative forcing of climate” by D. A. Hauglustaine et al.

Anonymous Referee #1

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This is a solid paper that accumulates the evidence for an important role of nitrate aerosol in present and future conditions. I particularly appreciate the careful job that the authors did trying to understand similarities and differences with other studies. The emphasis on the key role of agricultural (NH₃) emissions in future is justified, but perhaps some more emphasis should be placed on the strong limitations resulting from the use of the RCP emissions dataset. For almost all pollutant emissions, except for SO₂, they are to some extent assuming Kuznets assumptions, and it is very questionable what pollution levels in 2100 could be. Why not focus on 2050, a somewhat more foreseeable future?

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On the more technical level, the authors have formulated a simplified thermodynamic aerosol formation scheme. While it has been demonstrated in the past (e.g. Metzger et al), that such parameterizations are relatively accurately mimicking more accurate schemes, such evaluation is missing in the present manuscript. I suggest that the authors present such evaluation in the supplement, especially because the whole paper is building on the accuracy of this scheme. Some simple plots scanning concentration/T/RH space would be indicative of the model's performance.

The amount of figures is quite excessive (26). On the hand there will be always somebody who will appreciate particular plots- but it may preclude others to start reading the paper. I would encourage the authors to place less essential plots in the supplementary material, and work on some good summary plots and tables. For instance I have seen a number of scatter plots, but it difficult to grasp if the overall picture is consistent among these figures.

With regard to the measurements evaluation, especially the ammonium-nitrate-sulfate system, the authors should be more explicit on the limitation of the measurements that they have used. For instance it is known that filter-pack measurements at temperatures above 20 C, are prone to large sample losses, and at most present a lower limit to the model. Can this explain the overestimates of nitrate/nh4 reported in the paper?

Likewise an evaluation sub-regions- (perhaps in a table, regions from Chin et al??) could give more specific information on issues and model performance. We don't need more figures- but something that could summarize North-South differences in Europe/USA, for instance.

The authors mention a potential issue with heterogeneous reactions of N₂O₅, which is much lower than in other studies, but even compared to the same model (Bauer et al. 2007). I appreciate the honesty, but it is something that has to be clarified, since it could influence the model performance especially in winter.

Finally, I appreciate the attempts of the authors to compare to other studies. Never-

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theless it remains difficult to do this comparison more systematically due to a lack of standard evaluation sets and methods. To my opinion AEROCOM could play a more a substantial role in defining such benchmark sets- that would allow easy comparison of model results (beyond the joint exercises that take place from time to time), and monitor progress. I would appreciate some paragraph on this as well.

Despite these criticisms, I recommend the paper to be published in ACP, as a welcome addition to the not very extensive literature on nitrate aerosol, after taking into account the remarks above, and the detail comments below.

Detailed comments:

p. 6864 l. 8 same magnitude as ; represent=>compares to

p. 6864 l. 18 All nitrates, or mainly NH_4NO_3 ?

p. 6864 l. 20 RCP scenarios only?

p. 6867 l. 7 The thermodynamic scheme is used in some regional/global models. Was it evaluated against more comprehensive scheme? How good was it?

p. 6868l. 5-20 What is the thickness of the surface layer? How can the mixing of the model be characterized. These are essential parameters to understand NH_3 .

p. 6870 l. 11 I am not sure, but to me equation 3 just looks like a rewrite of equation 2? Or is the point that the factor Beta is independent evaluated?

p. 6871 Given the equations in R1 to R9: what is the number of N_2O production?

p. 6872/6873 could probably move to supplement together with a more thorough evaluation.

p. 6874 9- and further Only on dust and seasalt? Did the authors consider uptake on sulfate? And on NH_4NO_3 ?

p. 6877 I assume one year was used as spin-up?

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p. 6878 I found the color scheme giving too little information in the regions below 1 ug/m³

p. 6878 It is important to tell how the data were used. E.g. can summer measurements on filters be used? Is there a threshold of # days to calculate a monthly average. Were there quality issues, and how were they used?

p. 6879 I. 29 and further. The authors seem to suggest that it is physics of global models that are causing the issue. What about resolution? What about measurements that may be biased? This aspect needs to be more expanded also for abstract and conclusions.

p. 6882 I. 27 I am confused about the sulfate remark, since previously the authors only talk about reactions with dust and seasalt.

p. 6883 I 4 ensured=>caused, or most nitrate is lost by wet deposition

p. 6885 cloudiness formed?

p. 6888 Future evolution of what?

p. 6893 It is good that the authors perform sensitivity studies, but we should realize that the RCP scenarios are not internally consistent. Possibly more useful is to increase/decrease within a scenario the emissions of specific components.

p. I found the 'future' section relatively lengthy- and as I explained before, very dependent on relatively similar scenarios. Perhaps this is a section that could be reduced, while retaining the main points: the increasing importance of nitrate under future conditions.

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 6863, 2014.

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