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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 #3

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Review of “A global model simulation of present and future nitrate aerosols and their direct radiative forcing of climate” by Hauglustaine *et al.*, submitted to *Atmos. Chem. Phys.*

In this study, the authors document the addition of a nitrate aerosol scheme in the LMDz-INCA model, and use that model to highlight the dramatic change in aerosol composition that would happen over the 21st century if RCP are followed. The paper is well written and presents interesting results, notably that ammonium burden remains fairly constant in the future as ammonium nitrate formation replaces ammonium sulphate formation; that ammonia is the limiting factor explaining differences between dif-

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ferent RCPs; and that the change in aerosol composition has important consequences on direct radiative forcing (Figures 24, 26, and 27). The authors also do a good job at placing their study within the existing scientific literature, although they could be bolder in highlighting the strengths of their study.

For those reasons, I recommend publication of the manuscript, with minor changes to address the comments below.

1 Main comments

- The authors state several time that their results replicate those by previously published papers. Replication is important, but the authors should evaluate previous results more critically. For example, some of the previous studies did not include coarse-mode nitrate, or did not model ammonium explicitly. Do those limitations matter? The scheme implemented by the authors is fairly complete – at least from a global modelling point of view – and I believe the results in this paper can be seen with more confidence than some of the previous, more simpler, analyses.
- One defining characteristics of nitrate aerosols is their ability to easily dissociate back into the gas phase. This means that nitrate aerosols have a very pronounced diurnal cycle, spending most of daytime hours in the gas phase (Dall’Osto et al., 2009). This is crucial for their direct radiative effects, which is exerted by the aerosol phase only. Previous studies have overlooked that aspect, so it would be very interesting to add a short discussion of the diurnal cycle in LMDz and demonstrate that it follows the observed behaviour.
- There is a large number of figures in the paper, some of which being only discussed briefly in the text. I would suggest merging some of the figures together, especially Figures 1, 2, and 6; Figures 4 and 5; Figures 11 and 12; and possibly Figures 22 and 23.

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- Furthermore, the same kind of information (change in surface concentrations) is presented in different ways across Figures 16 to 18. The reason for that choice is unclear: Why doesn't Figure 17 show RCP4.5? Why does Figure 18 show timeseries rather than distributions? With regard to the subject matter, a Figure similar to Figure 21, but showing in addition regional changes, would be very helpful. By showing the evolution of all the species involved in nitrate formation (NH_3 , HNO_3 , SO_4) it would provide an efficient way of identifying the causes for the change in nitrate concentrations.

2 Other comments

Page 6866, line 1: Are the radiative forcings reported in this sentence for the direct effect only, or is it a mixture of direct-only and direct+indirect studies?

Page 6868, line 8: Out of curiosity, what does the z in LMDz stand for?

Page 6869, lines 6–7: The distinction between anthropogenic and natural aerosols made here is a bit arbitrary, since sulphate aerosols can be both, for example. I suggest rephrasing that statement.

Page 6872, line 1: It is worth stating here that the fact that sulphate formation takes precedence over that of nitrate is justified by the low vapour pressure of sulphuric acid.

Page 6874, line 13: In the introduction, limitations to the first-order removal method have been mentioned, with Feng and Penner (2007) as a reference. In section 2.2, it would be useful to shortly summarise what those limitations are.

Page 6878, lines 4 and 23: Does “agreement” refer to patterns, magnitudes, or both?

Page 6878, line 19: What do the authors mean by “anthropogenic nitrates” in that context?

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Page 6879, line 24: Another difficulty when comparing with measurements of nitrate aerosol concentrations is that they are difficult to measure, because nitrate aerosols dissociate easily. See for example Schaap *et al.* (2002). It is likely that those difficulties translate into underestimated concentrations in the observations.

Section 3.1, and Figures 2,4,5,6: It is worth clarifying early in the text and figure captions the units used for concentrations and burdens, i.e. whether they are given as [S] or [N], or as [SO₄], [NO₃], etc.

Page 6881, line 12: I'm not sure I understand the statement "nitrate on pure sulfates" here. By redoing the calculation, it seems that it means "fine mode nitrates".

Page 6886, line 13: A global-, annual-averaged total AOD of 0.059 is a surprisingly small number. Is that really correct? Looking at Table 5, it seems it is in fact not the total AOD, but excludes mineral dust and sea-salt aerosols.

Figure 13a: looking at Figures 5, I would have expected a more equal contribution of fine and coarse mode nitrate to total column around 50N, but it is difficult to judge by eye.

Figure 13b: Has the y-axis really been multiplied by 100? The magnitudes and the lines 18–20 on page 6887 suggest that is not the case.

Page 6891, lines 17–25: The discussion of Figure 20 is confusing, since it mixed hemispheric averages (which decrease) with regional changes that are of varying signs. It would be clearer to say that although Figure 20 seems to show an increase in nitrate column in most regions of the north hemisphere, that increase is in fact more than compensated by strong decreases over Europe and China, where present-day nitrate concentrations are large. Having said that, I am surprised by how localised the European and Chinese changes are: Why don't they extend downwind?

Page 6895, lines 5–8: I would rephrase this statement slightly as "because deposition of nitric acid, which dissociates readily in water, causes a significant drop in pH, but

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deposition of NH_x increases the water alkalinity.”

Page 6901, line 16: At this stage, I would think that understanding the sources of uncertainty is a more likely outcome of an intercomparison project than reducing those uncertainties.

3 Technical comments

The level of English language is very good, but there are recurring mistakes when plurals are used as adjectives, and a few other grammatical mistakes. To list but the first instances:

- Page 6864, line 2: fine nitrate particle formation
- Page 6865, line 2: nitrate exerts; line 9: nitrogen oxide emissions; line 10: soil emissions.
- Page 6870, line 14: organic carbon

I encourage the authors to pay particular attention to these matters when proof-reading the paper.

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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