

We thank both reviewers for their constructive comments and suggestions. Replies and corrections (in italics) are listed thereafter

Reviewer 1

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

We have included a comparison between observed and simulated diurnal cycle of NO_3^- at the YRK site from the SEARCH network. The following text has been added to the manuscript.

Note that the diurnal cycle of NH_3 emissions has a small impact on the simulated mean surface $[\text{NO}_3^-]$ concentration, but reduces surface $[\text{NH}_3]$ and increases its export to the free troposphere. Fig. S20 shows the observed and simulated diurnal cycle of $[\text{NO}_3^-]$ at the YRK SEARCH site. NO_3^- exhibits a pronounced diurnal cycle with a maximum in the early morning and a minimum in the late afternoon (as a result of both thermodynamics and boundary layer height). AM3N and AM3N_diu capture the timing of the diurnal cycle well. As NH_3 emissions peak in the afternoon, the magnitude of the NH_4NO_3 diurnal cycle in AM3N_diu is lower than in AM3N. Higher daytime concentrations of NH_4NO_3 in AM3N_diu suggest that accounting for the diurnal cycle of NH_3 emissions may increase the magnitude of the radiative forcing associated with NH_4NO_3 .

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

As shown in Fig. S1, below (added to supplementary materials), the mass extinction of NH_4NO_3 and $(\text{NH}_4)_2\text{SO}_4$ at 550nm calculating using standard Mie calculations differs by less than 20% below 95% relative humidity (we do not consider hygroscopic growth beyond that point) and by less than 10% between 90% and 95%. Consistent with a small impact of this simplification on the simulation NO_3^- optical depth, the simulated burden and optical depth for NO_3^- over the 2008–2010 period are very similar to those reported by Hauglustaine et al. (2014), who treated the optical properties of NH_4NO_3 explicitly. Other uncertainties such as the degree of internal mixing between NH_4^+ , SO_4^{2-} , NO_3^- , and BC, the cap on aerosol hygroscopic growth or even as highlighted in our work the simulation of RH are likely to be much more important in determining NO_3^- optical depth at 550nm.

However, we agree with the reviewer and we recognize that this simplification has important limitations. As a result we chose to only present the simulated AOD at 550 nm rather than the radiative forcing associated with nitrate. This simplification was motivated by current limitations in the treatment of aerosol optical properties in the GFDL-AM3 model associated with the mixing of NH_4^+ , SO_4^{2-} , NO_3^- , and BC. Revisions to the radiative code in the next GFDL atmospheric model (AM4), will allow us to use observed refractive index and hygroscopic growth for ammonium nitrate. The main findings of this study, such as the importance of the convective transport of NH_3 to the free troposphere for future projections of NO_3^- burden and optical depth are not expected to change.

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

We agree with the reviewer. However, the change in the convective scheme between AM3 and AM3N has little impact on the precipitation rates while it increases the efficiency of both convective removal and convective transport. Further analysis suggests that the finer discretization of the convective plumes in AM3N improves the stability of the tracer convective transport scheme. We have modified the text as follow:

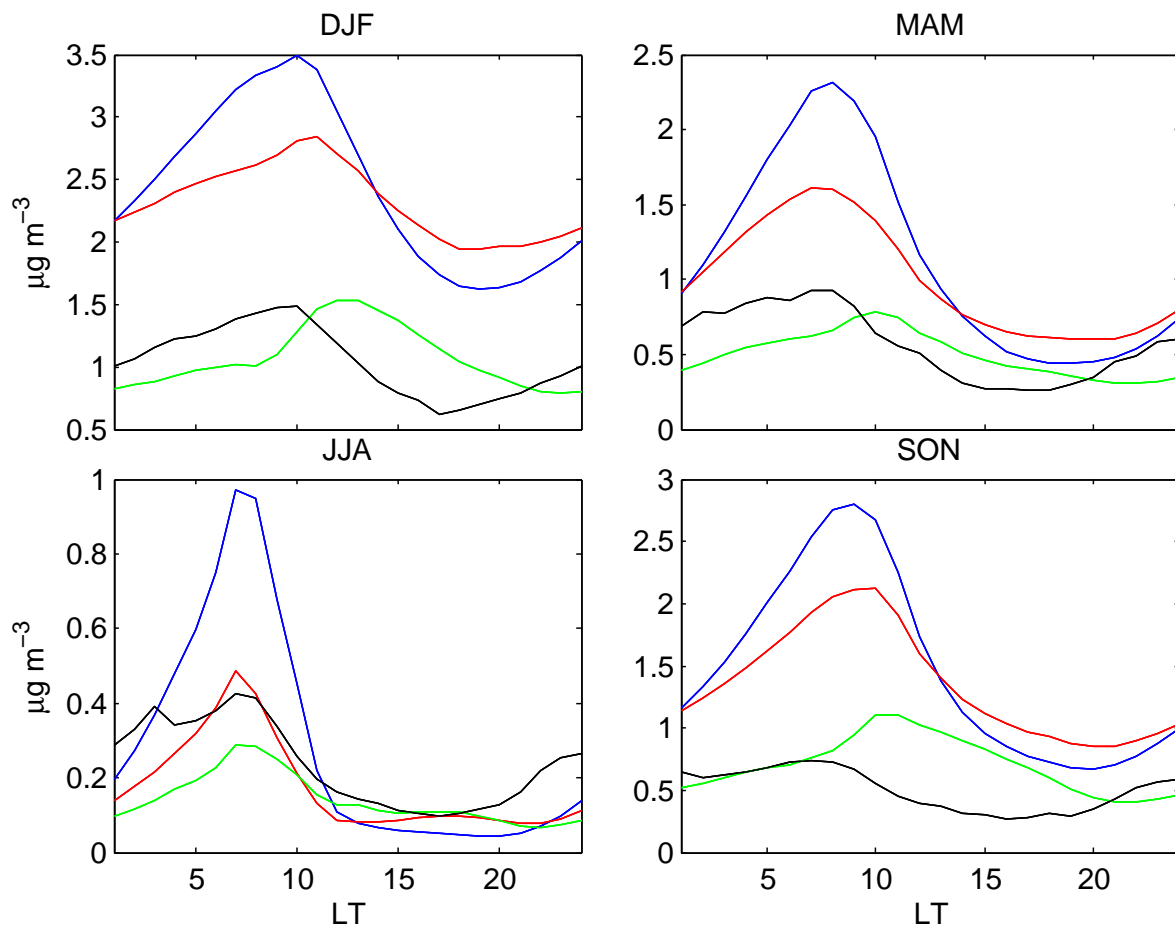


Figure S20: Average diurnal profile of NO_3^- at the YRK SEARCH site (33.93N, 274.95E, 395m asl) in 2008. Simulated nitrate diurnal profiles from AM3N, AM3N_diu, and AM3N_fdep_diu are shown in blue, red, and green respectively. Observations are shown in black

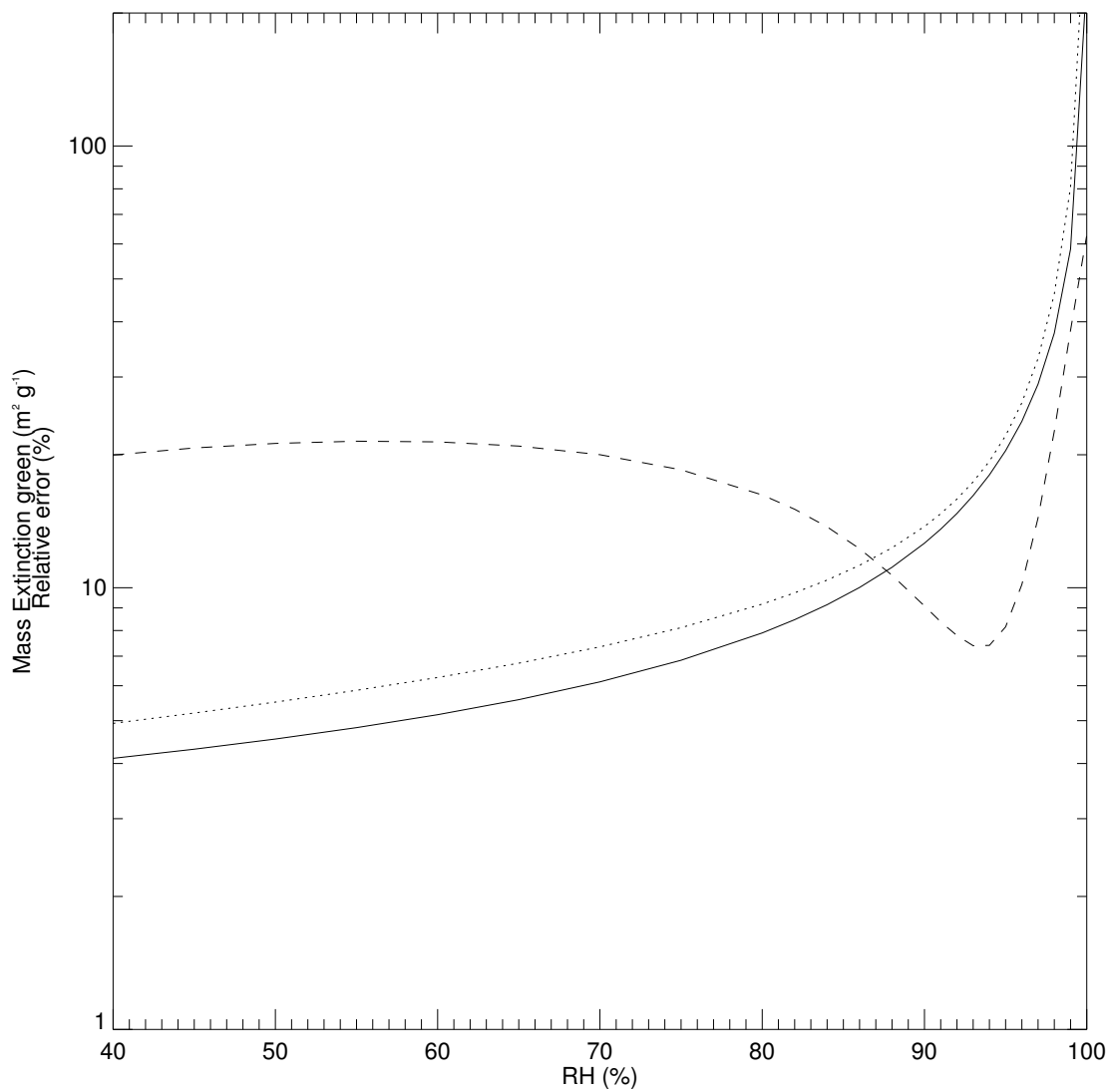


Figure S1: Mass extinction of $(\text{NH}_4)_2\text{SO}_4$ (dotted line) and NH_4NO_3 (solid line) at 550nm. The dash line shows the relative error between $(\text{NH}_4)_2\text{SO}_4$ and NH_4NO_3 mass extinctions. Refractive index and hygroscopic growth for NH_4NO_3 are taken from Gosse et al. (1997) and Tang (1996) respectively.

The improved discretization of the convective plume has little impact on precipitation at the surface but increases the convective wet removal of tracers as we will show [...]

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

The use of sulfate optical properties for nitrate introduces a small error at high RH (<10% for RH>90%), much less than the discrepancy between observed and simulated AOD at Bondville. The error is likely to be larger (<20%) under the low RH conditions (RH<40%) used to measure aerosol dry extinction. Thus the model high RH bias in winter is likely to cause the high bias in simulated AOD in all AM3N configurations. Excessive hygroscopic growth also helps explain that AM3 captures winter AOD remarkably well in spite of neglecting nitrate aerosol and underestimating sulfate surface concentrations. In summer, nitrate aerosol is very low and cannot explain the large underestimate of the aerosol optical depth (see Fig. S2).

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 simulation.

We have performed a new simulation with reduced convective removal of NH₃. In this configuration, the simulated nitrate burden is found to be similar to that reported by Hauglustaine et al. (2014) in the tropics and exhibits more sensitivity to changes in anthropogenic emissions. Fig. 12 and its description in the last section have been revised (see reply to the last comment of reviewer 2) to emphasize a) that the response of convection to climate change has important implications for future projections of nitrate burden and b) that differences in the representation of convective transport and removal across models contribute to the large variability in the modeled response of nitrate to projected changes in anthropogenic emissions.

Reviewer 2

Considering the comparison of the response of NO₃ AOD to future emissions (compared to that of Hauglustaine), for this simulation you start with a model in which NO₃ is already biased high, and compare to a set of scenarios in which NH₃ levels are already much higher, muting the response of the system to the emissions changes. It thus seems that some of the differences in the projected changes in AOD between the two studies may be owing to different starting conditions.

We have clarified that there is a large absolute difference between the projections for the NO₃⁻ burden in 2050 obtained from most AM3N configurations and that reported by Hauglustaine et al. (2014) for 2050. In particular, when using AM3N with RCP8.5 emissions for 2050, NO₃⁻ optical depth is 0.0076, 36% less than reported by Hauglustaine et al. (2014). One notable exception is AM3N_ndust, which produces a similar NO₃⁻ optical depth in 2050 (0.01) to the one reported by Hauglustaine et al. (2014).

We have added the following text in the 2050 emission section:

The response of NO₃⁻ to changes in anthropogenic emissions is weaker than reported in recent studies. For instance, Hauglustaine et al. (2014) reported a NO₃⁻ optical depth of 0.01 for 2050 and an increase of the conversion rate from NH₃ to NO₃⁻ from 0.36 day⁻¹ to 0.57 day⁻¹ from 2000 to 2050. Using the same anthropogenic emissions, the simulated NO₃⁻ optical depth in AM3N in 2050 (the configuration closest to that used by Hauglustaine et al. (2014)) is 0.077 and the conversion rate from NH₃ to NO₃⁻ is 0.33 day⁻¹.

It seems a bit odd to show the model evaluation for one of the sensitivity simulations (AM3N_fdep_diu) but then report results for the AM3N simulation for subsequent analysis. It seems like whichever simulation is justifiably ‘best’, both in terms of the details of the mechanisms included as well as the model evaluation, would be most suitable for reference in the rest of the work. I think this may actually be the case, but there is some ambiguity in the text and certainly some ambiguity in the figures and Tables 1 somebody who looks at Table 2 and then Fig 9 would never suspect that here “AM3N” actually means (I think) “AM3N_fdep_diu”.

Fig. 9 shows AM3N_fdep_diu not AM3N and we have corrected the caption. In the manuscript, we chose to focus on the comparison between observations and AM3N_fdep_diu as it better captures surface NO₃⁻ concentration than AM3N. However, in the analysis we use AM3N as our reference for two reasons: a) this facilitates the analysis of the impact of different processes as the sensitivity tests performed in this study (Table 2) are all based on AM3N,

b) the magnitude of the perturbations are large and often represent upper bounds. This is especially the case for the simulations with increased deposition of NO_3^- and no heterogeneous production of HNO_3 .

We have added AM3N_fdep_diu to Fig. 12. It can be seen that the difference between AM3N and AM3N_fdep_diu cannot explain the differences with the simulation performed by Hauglustaine et al. (2014). Further revisions are detailed in the response to the reviewer's last comments

Abstract: “only find a modest” I was confused about how this was presented. 30% changes in nitrate AOD in response to 30-40% changes in SO₂ and NH₃ seems significant. I guess after reading the paper I understand that this may seem like a small change, relative to some other studies, but by itself it doesn't seem modest. It might be best to just remove such qualifiers and just present the quantitative results, or alternatively explain the context a bit more.

We agree with the reviewer and we have removed references to previous studies from the abstract. In the main text, we now emphasize absolute differences more. The abstract was revised as follow:

Simulated nitrate optical depth increases by less than 30 % (0.0061–0.010) in response to projected changes in anthropogenic emissions from 2010 to 2050 (e.g., –40 % for SO₂ and +38 % for ammonia). This increase is primarily driven by greater concentrations of nitrate in the free troposphere, while surface nitrate concentrations decrease in the midlatitudes following lower concentrations of nitric acid. With the projected increase of ammonia emissions, we show that better constraints on its vertical distribution (e.g., convective transport and biomass burning injection) and on the sources and sinks of nitric acid (e.g., heterogeneous reaction on dust) are needed to improve estimates of future nitrate optical depth.

Abstract: The focus here is on changes in global budgets. Given that secondary inorganic aerosol though is rather regional, is that the best way to summarize the impacts? Later in the manuscript evaluation is considered for the top percentiles of locations. would that be more suitable here as well, or in addition?

We have added the following sentence in the abstract

In wintertime, nitrate aerosols are simulated to account for over 30% of the aerosol optical depth over western Europe and North America.

Introduction: It seems like the set of references cited need to either be more comprehensive or “i.e.” inserted when citing only representative studies.

corrected

25741.15: Also see West et al., AE, 1998, Marginal direct climate forcing by atmospheric aerosols, and Henze et al., ES&T, 2012, Spatially refined aerosol direct radiative forcing efficiencies.

Both references have been added

Section 2.1: I was expecting a description of heterogeneous chemistry here. Granted, it comes later. Maybe that can be indicated?

We have clarified that the parametrization of the heterogeneous chemistry is based on Mao et al. (2013). We have also expanded the description of changes to the heterogeneous chemistry to include changes in the γ for N_2O_5 , NO_2 , and NO_3 between AM3 and AM3N (see revised Table S1).

We also reduce the reaction probabilities (γ) of N_2O_5 , NO_2 , and NO_3 on aerosols relative to AM3 (Mao et al., 2013) (see Table S1 and Sect. 2.3.2). The implications of these changes for the budget of HNO_3 and aerosol NO_3^- are described in Sect. 2.4.

Section 2.2: Similarly, I was expecting a description of NH₃ emissions, but that came later

We have added the contribution of the different sectors to the emissions of NH_3 in Table 1.

25743.23: Why is this neglected?

Coarse-mode nitrate is expected to make a small contribution to present-day nitrate optical depth and contributes little to the response of NO_3^- optical depth to future anthropogenic emissions (e.g., Hauglustaine et al. (2014)). We have modified the text as follow:

The optical depth of NO_3^- associated with dust is expected to be small relative to fine-mode NO_3^- (e.g., Hauglustaine et al. (2014)) and it is not considered here.

25747.1: That production of HNO3 from N2O5 is the dominant pathway in the norther mid latitudes in the winter has been known much earlier than 2010.

We have added references to Dentener and Crutzen (1993) and Tie et al. (2003)

25747.10, 25747.14: why are these neglected?

We have revised the text to clarify that the AM3N_nhet and AM3N_ndust configurations are designed to quantify the impact of different treatments of heterogeneous chemistry on the simulated NO_3^- optical depth.

Wintertime production of HNO_3 in the northern midlatitudes boundary layer is dominated by the uptake of N_2O_5 on aerosols (e.g., Dentener and Crutzen (1993); Tie et al. (2003); Lamsal et al. (2010)). The probability for the heterogeneous conversion of N_2O_5 to HNO_3 (γ) remains uncertain (Chang et al., 2011) with field and laboratory observations showing that it is inhibited by aerosol nitrate and organics (Brown et al., 2009; Brown and Stutz, 2012; Wagner et al., 2013; Gaston et al., 2014), but enhanced by cold temperatures (Griffiths and Anthony Cox, 2009; Wagner et al., 2013). To quantify the impact of the heterogeneous production of HNO_3 on aerosol NO_3^- , we neglect the heterogeneous production of HNO_3 via N_2O_5 aerosol uptake in AM3N_nhet. We also neglect the productions of HNO_3 by NO_3 and NO_2 reactive uptake, as they may modulate the wintertime budget of NO_y in polluted region (Paulot et al., 2013). Note that previous characterizations of NO_3^- optical depth also neglected the heterogeneous chemistry of oxidized nitrogen (e.g., Bellouin et al. (2011)). We also evaluate the impact of the heterogeneous chemistry on dust as it is not included in all model (e.g., Pye et al. (2009); Bellouin et al. (2011)). In AM3N_ndust, we neglect the uptake of HNO_3 , N_2O_5 , NO_3 , H_2SO_4 , and SO_2 on dust.

25748.20: Also agrees well with observed values (e.g., Lee et al., JGR, 2011, SO2 emissions and lifetimes: Estimates from inverse modeling using in situ and global, space-based SCIAMACHY and OMI observations

Thank you. We have added this reference to the manuscript.

Section: Description of biases are largely qualitative. Without changing this to be an extensive numerical catalog, it would be good in places to be more specific about what is a low or high bias, small or large, more quantitatively

We have incorporated some key numbers from Table 3 and S3 in the text.

25754.15: Does increased convective precip also increase loss of HNO3?

Yes it does. We have modified the text as follow:

Increased convective removal of HNO_3 and NH_3 also reduce the low bias in simulated summer wet deposition NO_3^- (-50% to -23%, Fig. S9) and NH_4^+ (-46% to -16%, Fig. S10).

25755.3: The same was shown in Zhu et al., ACP, 2015, Global evaluation of ammonia bi-directional exchange, which also found bigger impacts on surface NO3 than indicated in the present work. Also, on the following page the impact of NH3 diurnal variability is said to impact NH4NO3 (25757.2), so it seems odd to say here that the effect on NO3 is small.

We have clarified here that we only refer to surface concentrations. The impact on the NO_3^- burden (mentioned in line 25757.2) is more significant but it is primarily driven by an increase in the export of NH_3 to the free troposphere, consistent with the large sensitivity of NO_3^- to the convective removal of NH_3 . We have added a reference to the study of Zhu et al. in the section devoted to NH_3 emissions. See also the replay to reviewer 1 first comment regarding the diurnal cycle of NH_4NO_3 .

Abstract, Conclusions, etc: The authors purportedly evaluated the sensitivity of nitrate with respect to “uncertainties in NH₃ emissions”, but it seems that they only investigated but it seems that they only investigated the sensitivity with respect to the diurnal or seasonal variability in NH₃ emissions. The net emissions themselves are probably uncertain by x2 globally, and maybe much more than that regionally. So it seems that either the authors can’t really claim to have investigated the impacts of NH₃ emissions uncertainties in the most general sense. Either more numerical experiments are required, or more precise language is called for.

Throughout: Bidirectional exchange of NH₃ is never mentioned. Should it be included in discussion of uncertainties, since it isn’t considered here?

We agree with the reviewer that there are considerable uncertainties in NH₃ emissions. We believe that differences between RCP8.5 NH₃ emissions and HTAP emissions capture some of this uncertainty. These inventories differ significantly both spatially (see Fig. 1) but also temporally (RCP8.5 NH₃ emissions have no seasonality). For instance, India anthropogenic emissions of NH₃ in HTAPv2 are more than twice as large as in RCP8.5. These large differences have a small impact (<10%) on the NO₃⁻ burden simulated for 2050, consistent with the diminishing sensitivity of NO₃⁻ to NH₃. In AM3N, ammonia remains confined to the surface. As NH₄NO₃ becomes more and more limited by HNO₃ following projected changes in anthropogenic emissions, the representation of 2050 ammonia anthropogenic emissions has little impact on the simulated nitrate burden in our model. Models with more efficient transport of NH₃ to the free troposphere may exhibit a greater sensitivity to NH₃ emissions including to the bidirectional exchange of NH₃. This has been clarified in the conclusion.

The text was modified as follow:

2050 emissions:

Figure 12 shows that the NO₃⁻ burden is projected to shift equatorward in the Northern Hemisphere in response to changes in anthropogenic emission from present-day to 2050. NH₄NO₃ increases in the free troposphere but decreases near the surface, a vertical redistribution also noted by Hauglustaine et al. (2014). The decrease of surface NO₃⁻ in the midlatitudes is primarily driven by lower NO emission. Large differences in the seasonality, spatial distribution, and magnitude of anthropogenic NH₃ emissions in RCP8.5 (dotted line) and scaled HTAPv2 for 2050 have little impact on the simulated NO₃⁻ burden (<10%), which reflects the diminishing sensitivity of surface NH₄NO₃ to NH₃. However, NO₃⁻ remains sensitive to NH₃ in the free troposphere, where it can persist longer than in the boundary layer thanks to lower temperature. The solid line in Figure 12 shows the impact of lower convective removal of NH₃ (achieved by neglecting the impact of pH on NH₃ solubility) on the NO₃⁻ burden. Over the 2008–2010 period, this results in a 40% increase of the NO₃⁻ burden with a near quadrupling in the tropics, qualitatively matching the results of Hauglustaine et al. (2014) in this region. In 2050, the impact is much more pronounced and the simulated burden is more than twice as large as in 2010, a similar response to that found by Hauglustaine et al. (2014). Note that increasing NH₃ emissions from biomass burning and distributing these emissions vertically (Naik et al., 2013) also increases tropical NO₃⁻ (not shown) but to a much lower degree (<50%). These results suggest that differences in the transport of NH₃ to the free troposphere across models contribute to the variability in the projected NO₃⁻ burden and optical depth. Such differences may arise from differences in the parameterizations of convection (Folkins et al., 2006) as suggested by the much lower tropical NO₃⁻ burden in AM3N than in LMDz-INCA model (Hauglustaine et al., 2014) but also from changes in the tropical circulation in response to climate change (e.g., Ma et al. (2012)).

Conclusion:

We have examined the response of simulated NO₃⁻ optical depth to projected changes in anthropogenic emissions from 2010 to 2050 in RCP8.5. Depending on the configuration of AM3N (Table 2), NO₃⁻ optical depth varies from 0.0061 to 0.01 in 2050. The increase of NO₃⁻ (< 30 % relative to 2008–2010) is partly inhibited by greater limitation of NH₄NO₃ production by HNO₃ at the surface due to lower NO emissions, more efficient removal of HNO₃ by dust, and a large decrease in the heterogeneous production of HNO₃ by N₂O₅ (associated with lower aerosol surface area). In the Northern Hemisphere, the NO₃⁻ burden is projected to shift southward, following the increase of tropical NH₃ emissions and the decrease of NO emissions in the midlatitudes. This shift is associated with an increase of the NO₃⁻ burden in the free troposphere, where NH₄NO₃ formation is limited by NH₃. We suggest that the convective transport of NH₃ and its response to climate change (not considered here) play an important role in modulating the response of NO₃⁻ optical depth to changes in anthropogenic emissions. The complexity of the response of NO₃⁻ to changes in surface processes, chemistry, and convection indicates that the global trends of NH₃ emissions may not be a suitable proxy to estimate the future forcing from NO₃⁻ aerosols (Heald and Spracklen, 2015). We conclude that in addition to improvements to NH₃ emission inventories (e.g., bidirectional exchange of NH₃ (Zhu et al.,

2015)), *observational constraints on the processes controlling the vertical redistribution of NH₃ and the response of NO₃⁻ to NH₃ in the free troposphere (e.g., magnitude of NH₃ emissions in the tropics (Aneja et al., 2012; Whitburn et al., 2015), biomass burning injection height (Val Martin et al., 2010), transport and removal of NH₃ in convective updrafts, heterogeneous chemistry on dust) and sensitivity studies to characterize their response to climate change are needed to improve estimates of present and future NO₃⁻ optical depth.*

Fig 2: by a black cross in the upper left panel.

corrected

Fig 9: “Across AM3N” It isn’t clear what is mean here. Across what? Seasons? Different model configurations?

we have specified that we refer here to the different AM3N configurations

25741.11: However, recent

corrected

25741.17: In this study

corrected

25747.3: It seems rather obvious that it would depend on the choice or reaction probability suggest rewording.

this subsection has been rewritten see above.

throughout: It is odd to present numbers as “X %” rather than “X%”.

this was done during typesetting by copernicus staff. Thus we have kept all percent numbers as “X %”

25747.16: slow, similar

corrected

25747.16: results

corrected

Other corrections

The following additional corrections have been made:

1. We corrected an error in the labeling of the tick marks in Fig. 8.
2. We have added seasalt and dust optical depths for the LMDz-INCA model in Fig. 9.
3. We have added that revisions to the convective removal of ammonia in the GISS model result in a much lower simulated present-day nitrate optical depth (0.005, S. Bauer, personal communication).

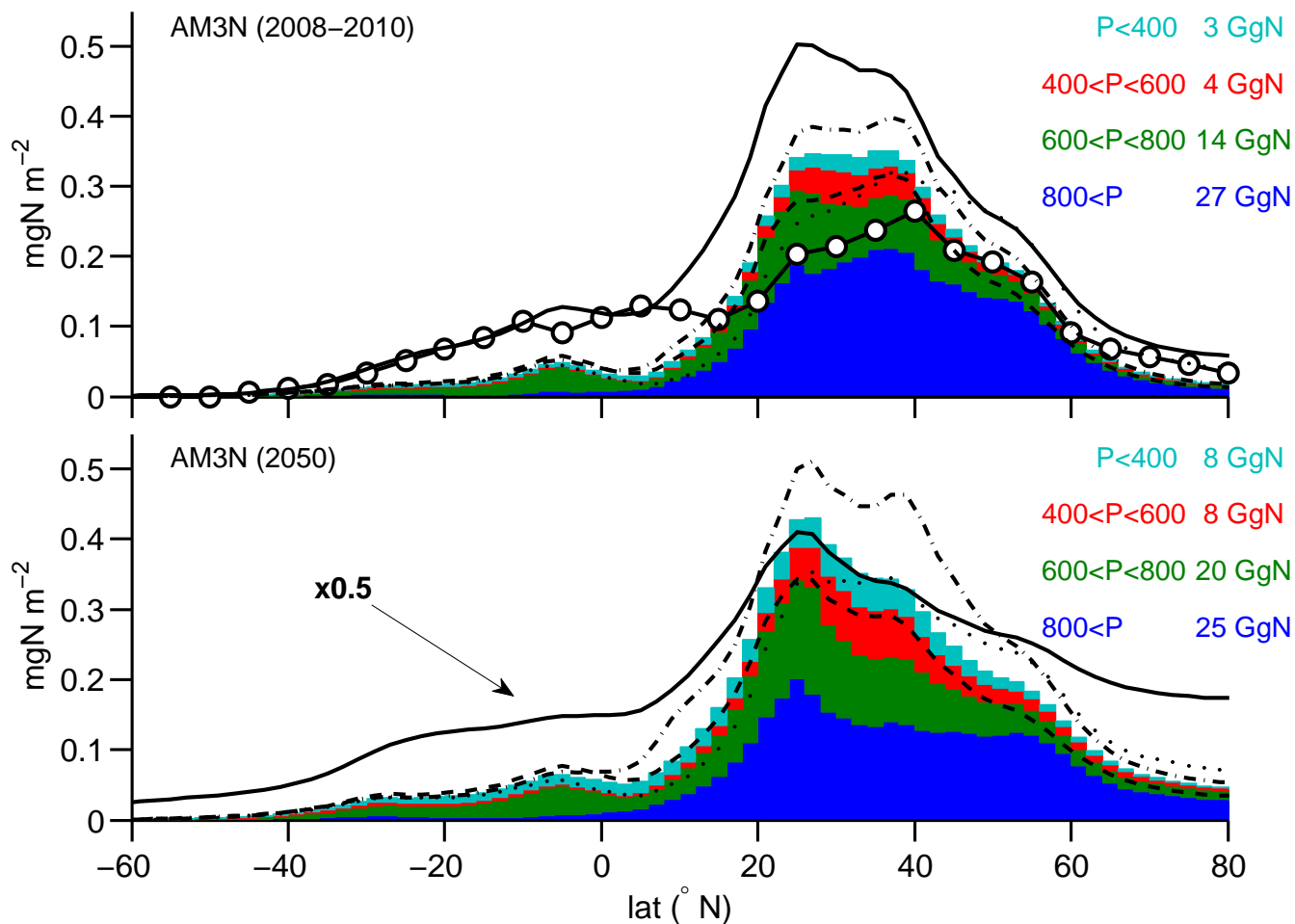


Figure 12: Annual zonal mean distribution of NO_3^- in AM3N with 2008-2010 anthropogenic emissions (top) and 2050 anthropogenic emissions (from RCP8.5 except for NH_3 , see text). The blue, green, red, and cyan regions denote the NO_3^- burden located above 800 hPa, between 600 and 800 hPa, between 400 and 600 hPa, and below 400 hPa, with the partial burden in each pressure range indicated inset. The annual mean zonal burdens of NO_3^- simulated using AM3N_fdep_diu (dash line), using AM3N with anthropogenic emissions from RCP8.5 for NH_3 (dotted line), using AM3N_ndust (dash dot line), and using AM3N with reduced convective removal of NH_3 (solid line) are also shown. The white circles in the top panel indicate the 2000 annual zonal mean NO_3^- burden simulated by Hauglustaine et al. (2014).

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