Atmos. Chem. Phys. Discuss., 15, C9687–C9690, 2015
www.atmos-chem-phys-discuss.net/15/C9687/2015/

Atmospheric Chemistry and Physics Discussions

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 #2

Received and published: 24 November 2015

The manuscript by Paulot et al., Sensitivity of nitrate aerosols to ammonia emissions and to nitrate chemistry: implications for present and future nitrate optical depth, presents updates to the AM3 simulation of nitrate aerosol. The study evaluates the impacts of update treatment of aerosol thermodynamics, heterogeneous processes, and emissions on nitrate concentrations and aerosol optical depth. Overall, the study is fairly rigorous and the model responses are well explained through application of several sensitivity studies. While several significant sources of model bias and differences compared to other studies in the literature are evident, these differences are generally well explained. The manuscript is suitable for publication following minor revisions to address the comments below.

Comments

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.

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 âĂŤ somebody who looks at Table 2 and then Fig 9 would never suspect that here "AM3N" actually means (I think) "AM3N_fdep_diu".

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.

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 âĂŤ would that be more suitable here as well, or in addition?

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

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.

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

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

25743.23: Why is this neglected?

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.

25747.10, 25747.14: why are these neglected?

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.

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.

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

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.

Abstract, Conclusions, etc: The authors purportedly evaluated the sensitivity of nitrate with resect to “uncertainties in NH3 emissions”, 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?

Editorial

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

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

25741.11: However, recent

25741.17: In this study

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

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

25747.16: slow, similar

25754.19: results

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 25739, 2015.