Atmos. Chem. Phys. Discuss.,
doi:10.5194/acp-2016-426-RC2, 2016

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Chemistry-climate interactions of aerosol nitrate from lightning" *by* H. Tost

Anonymous Referee #2

Received and published: 10 September 2016

1 Summary

This study performs a series of sensitivity simulations in a global coupled chemistry-climate model (CCM) aimed at characterizing the impact of lightning NO_x emissions on the composition of the atmosphere and climate, with particular focus on the role of nitrate aerosol. Within the EMAC CCM, the study has performed decadal time-slice simulations for the preindustrial and present. For each time period, simulations with zero lightning emissions were compared to simulations containing lightning emissions. All simulations were done with two separate mechanism for cloud-aerosol interactions.

The paper is of scientific merit and the results interesting. However, I think that in order to be suitable for publication, there needs to be a careful look at the statistical

significance of the results over such a short simulation period (see general comments). I'm not convinced that the study period is of appropriate length for the system being examined.

2 General Comments

1. I'm concerned that 10-year time-slice simulations are too short a period over which to significantly quantify lightning-aerosol impacts in a free-running chemistry-climate model. Both lightning and aerosols have strong sensitivity to clouds, which are highly variable in space and time. Whereas the global chemical tendencies and their physical explanations as argued here are probably correct, I'm not sure how well we can trust the reported magnitudes without an analysis of how statistically significant the changes are relative to the natural climate variability over the period. The weak pattern correlation reported in Section 3.4.1 and the large variability relative to the lightning NO_x forcing observed in the latter figures imply that there is still low signal-to-noise. The figure that does show significance tests (Fig. 7), has few locations with statistical significance, which are generally not regions with largest impacts from lightning. I'm especially concerned about the cloud properties changes attributed to lightning in the difference plots. I think the manuscript would be improved if the same significance tests were done for the data in Figs. 2-6 to establish which signals are robust. Ideally, the simulations could be extended until significance was achieved in each of the examined variables. However, I realize that this is not necessarily possible, so I think at least including the statistical significance estimates are critical, if the simulated changes are to be attributed to the lightning emissions.

2. The author is correct that few global CCMs include aerosol nitrate. However, the paper neglects to mention the global and regional chemical transport models

(CTMs) studies of lightning impacts on photochemistry and aerosols, several of which include ammonium nitrate aerosol thermodynamics and chemistry. Some recent examples include

- Allen et al., ACP, 2012, doi:10.5194/acp-12-1737-2012
- Murray et al., JGR, 2013, doi:10.1002/jgrd.50857
- Holmes et al., ACP, 2013, doi:10.5194/acp-13-285-2013
- Zare et al., ACP, 2014, doi:10.5194/acp-14-2735-2014
- Gressant et al., ACP, 2016, doi:10.5194/acp-16-5867-2016

Whereas, none of those studies explicitly report the impact of lightning on nitrate aerosol, the influence of lightning via nitrate aerosol pathways on ozone, OH, methane lifetime, and/or bulk $PM_{2.5}$ were included. In particular, the conclusion stated on page 8, lines 241-244 implies that no lightning photochemistry study has included nitrate aerosol chemistry. I would recommend that the Introduction and Section 3.2.2 be rephrased to acknowledge that the ozone, OH and methane lifetime results presented here are in agreement with the CTM studies, and emphasize that what is uniquely reported here are (1) the isolation of lightning impacts on nitrate aerosol, and (2) the discussion of lightning impacts on climate-relevant aerosol properties and the climate system itself.

3. The 3-D renders in the figures are novel and interesting, but very hard to interpret in a 2-D print media, especially for Fig. 5 and 6, where information on the faces of the rectangular cube are severely distorted and blocked by the 3-D contours. In particular, I think Fig. 5 would benefit by being converted into multiple figures.

3 Specific Comments

- P2; L47 - “this” indirect effect

- P2; L53-63 - This study is still a substantially large perturbation to the reactive nitrogen budget of the free troposphere, so I would still consider its results to be strongly susceptible to errors introduced by non-linearities. To truly minimize these errors, one would need to do a small perturbation analysis (e.g., Sauvage et al., 2007; doi:10.1029/2006JD008008). I agree with the method used here, but I think this paragraph is slightly misleading with respect to the uncertainties.
- Section 2.3 - How many years was each simulation initialized over? Is methane prescribed or allowed to respond to the large changes in OH?
- P3; L82 - comma should be before “we” instead of “that”
- P5; L132 - nitrate “precursors” from
- P5; L152 - The “C-shaped” profiles are somewhat outdated. Unimodal distributions with maxima in the free troposphere suggested by top-down (Ott et al., 2010; doi:10.1029/2009JD01188) and bottom-up modeling studies (Koshak et al., 2014; doi:10.1016/j.atmosres.2012.12.015).
- P5; L161 - “also” should be moved to before “are”
- P8, L251 - do you mean “oxidation capacity of the upper troposphere”?
- P9; L289-290 - I’m not sure exactly what is meant by the clause that begins with “whereas.” But if there is a statistically significant increase in CCN over Africa (but not South American or Indonesia), that is an interesting result from the perspective of the potential role that it might play in leading to convective invigoration that might contribute to the African lightning maximum, which models seldom replicate. Aerosols have been implicated before (e.g., Jacobson et al., 2009; doi:10.1029/2008JD01147).

- P9; L334 - I interpret “back” and “front” panels as being the northern and southern faces of the rectangular cube. I would recommend switching to using “left” and “right” face, or “western” and “eastern” face for Figs. 5 and 6.
- P14; L465-466 - Lightning strongly impacts background global oxidant levels. Shouldn’t we expect significant impacts on shortwave radiation near the aerosol precursor sources in the midlatitudes?
- P15; L497 - comma should go after “NH3”
- P15; L499 - please clarify if you mean to the global aerosol nitrate burden, or the local upper troposphere
- P15; L504 - Why not represent the oxidation potential via the oxidant concentrations themselves, rather than indirectly via the lifetime? The methane lifetime is heavily biased toward the tropics due to strong temperature sensitivity of the CH_4 + OH reaction. This may underrepresent the importance of lightning on the extratropics.
- Fig. 4 - the axis labels for the contour panels are illegible. I would recommend making this a 3 x 3 panel plot, with the panels in rough geographic order.
- Fig. 7 - the units for the y-axis are missing (W m^{-2}?)
- Table 1 - “Differences due to LNO_x emissions” is ambiguous toward its directionality. I’d recommend using “Estimated contribution from lightning emissions”.

Interactive comment on Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2016-426, 2016.