Review of "Chemistry-climate interactions of aerosol nitrate from lightning" by Tost


Summary

This article shows the effect of nitrogen oxides ($NO_x$) produced by lightning on aerosol nitrate, other trace gases and aerosols, aerosol and cloud properties, and atmospheric shortwave radiation using a global chemistry-climate model. The impact of including lightning-$NO_x$ ($LNO_x$) emissions is to increase ozone and odd hydrogen (as shown via the methane lifetime), similar to previous studies. $LNO_x$ emissions increase aerosol nitrate burdens, and also aerosol sulfate burdens (somewhat) because of the effect on odd hydrogen. These changes manifest themselves in changes in aerosol size distributions and extinction, cloud drop and ice number concentrations, as well as a small effect on the radiation fluxes. The new information from this paper is the impact on aerosols. However, I am not convinced these are significant impacts because of the large internal variability and uncertainties in the modeling system.

The paper should focus on what is unique and on what the important findings are. In its current state, the paper- does not do a sufficient job of characterizing what changes are important and which ones are not. There is much refinement to the analysis on what these results mean that needs to be done. The presentation needs improvement too. I personally would not use 3-d figures, because they are difficult to understand. The English writing could be improved too.


Major Points

1. There are many discussion points illustrating small differences between simulations with and without $LNO_x$ emissions. Are these differences significant enough to be discussed? I suggest the author focus the paper on substantial (i.e. >10%) changes and what unique things we learn from these simulations. Because the effect of $LNO_x$ emissions on ozone and other oxidants has been shown before (e.g., Labrador et al., 2005), these findings should be limited to explaining why the sulfate aerosol burden increases. I think using actual oxidant burdens is preferable to using methane lifetime.

2. The paper contains discussion on the impact of $LNO_x$ emissions during preindustrial times, showing results in the supplement, but these results are similar to the present day scenario and do not even warrant mentioning in the abstract. Perhaps the preindustrial results could simply be summarized in a paragraph in the discussion or conclusions of this paper.

3. It seems to me that the loss processes should be addressed in this study. The author mentions the formation of $NH_4NO_3$ in the upper troposphere. However, considering inorganic aerosol particles are mostly washed out (e.g., Chatterjee et al., 2010, J. Atmos. Chem.; Gilardoni et al., 2014, ACP; Yang et al., 2015, JGR) and the highly soluble $NH_3$ and $HNO_3$ are likely removed by thunderstorms, it seems that there may not be enough $NH_3$ to form $NH_4NO_3$ downwind of the storms.

4. I thought many of the figures were challenging to read. The author should consider whether the figures do the best job in delivering the message of the paper

in a clear manner such that the readers can easily grasp the science learned. The author should also consider how well a colleague could explain the science in the paper using the figures provided (e.g. for teaching purposes).

Specific Comments

Lines 30-37. Is there any observational evidence of $NH_4NO_3$ formation downwind of convection?

Lines 107-118. Are aerosol-cloud interactions applied to both resolved and parameterized clouds? Please clarify.

Lines 134-136. It would be good to give a brief description of what ACCMIP present-day and pre-industrial emissions are. For example, what assumptions went into creating the pre-industrial scenario.

Lines 140-143. Are there specific years that comprise the present-day simulations? Are there specific years for pre-industrial simulations? Are all these simulations run as a climate model or are they driven from a reanalysis product?

Lines 151-152. Subsequent papers by Pickering's students have updated the vertical profiles for lightning-$NO_x$ emissions (DeCaria et al., 2005; Ott et al., 2010). These profiles exclude a NO source at lower altitudes. It seems that the consequence of the Pickering et al. (1998) profiles do not have a big effect on the model results, but I suggest that the chemistry-climate model be updated.

Lines 176-178. It seems that PAN and other organic nitrates and perhaps $NO_3$ should be included as contributors to $NO_y$ in addition to $NO_x$, $HNO_3$, and $N_2O_5$.

Lines 198-199. How long does it take for $NO_2$ to transform to aerosol nitrate?

Lines 200-203. It appears that a discussion of the results of Figure 3 was not included.

Lines 211-217. To me, a budget includes source and sink terms describing the major pathways creating and destroying a trace gas. Table 1 shows only the contribution of various species to N(V), and has no discussion of the processes affecting N(V) species.

Lines 215-217. It would be nice to see supporting information that $NH_3$ emissions are responsible for lower particulate nitrate concentrations.

Lines 211-217. It seems that $NO_3$, PAN, and other organic nitrates should be included.

Lines 218-223. I could not connect the numbers presented in this paragraph to the ones listed in the Table. Is the particulate nitrate contribution determined from dividing nitrate column burden by total N(V) column burden (143/554)?

Lines 232-235. Another recent paper also reports the effect of $LNO_x$ emissions on tropospheric ozone burden. Finney et al. (2016) ACP find a 27-30% increase in tropospheric ozone due to $LNO_x$ depending on the manner for calculating lightning flash rates. The paper tropospheric ozone burden found by Finney et al. (2016) is substantially lower than that reported here. I assume these differences are simply based on the model configuration. However, it would be good to cite the Finney et al. (2016) findings.

The author may want to also cite Finney et al. (2016) GRL, which discusses the effect of $LNO_x$ emissions on ozone among the ACCMIP models.

Lines 245-251. As mentioned in the opening remarks in this review, the methane lifetime is not really the best way to show that OH is affected by $LNO_x$ emissions and therefore sulfate burdens. I suggest removing this discussion. However, if it is kept there are a few things that need to be improved.

First, please explain the method better for calculating the methane lifetime. Second, be consistent with nomenclature. In the text it says $LNO_x$ emissions increase methane lifetime, but in the table caption it says, "increase due to neglect of $LNO_x$ emissions". Third, please explain why the change in lifetime occurs. This might be discussed in Labrador et al. (2005), but it is worth summarizing in this paper.

Lines 253-257. It is interesting to see that the $LNO_x$ emissions affect sulfate aerosol concentrations. The author attributes this to the gas-phase chemical production via OH oxidation of sulfur dioxide. However, could the aqueous-phase production also be different because its main oxidants, ozone and hydrogen peroxide, are affected by $LNO_x$?

Section 3.2. The author highlights changes of various key constituents. Is it important to highlight the change if it is less than a 10% change? Surely, there is enough uncertainty in other model parameters to complicate the interpretation of a small change in the burden of a constituent. Perhaps the author could state the statistical significance of these changes.

Lines 279-290. It would be helpful to know why the 9 regions were chosen. It appears that the regions are defined by latitude-longitude values without regard for land or ocean (which have quite different aerosol size distributions). If the data were further filtered for over land regions for the U.S., South America, Africa, Europe, East Asia, and Siberia, would there be a substantial change in size distributions?

Lines 312-315. The impact of $LNO_x$ on aerosol water uptake is not surprising since most of the $LNO_x$ effect is in the upper troposphere where it is quite dry.

Lines 342-344. It is an interesting point that the maximum enhancement in aerosol extinction occurs in the middle troposphere. I think the strength of using a global model is to show these downstream effects. Can the author say something as to why the middle troposphere is affected more than the upper troposphere? I was going to suggest the ice sedimentation to the mid-troposphere where $HNO_3$ would be degassed when the ice sublimated, but the author points out that the largest aerosol nitrate enhancement is in the upper troposphere.

Lines 371-374. I like frequency distributions because they quantify some more the changes that are occurring. It seems to me that these plots could be included as a figure, especially since it is worth discussing. The discussion seems to point to one perhaps significant difference between present day and pre-industrial scenarios.

Lines 401-402. Why is the polar latitude cloud coverage changed and is it *statistically* significant?

Lines 415-421. The discussion focuses on the cloud drop number and ice crystal number concentrations, but there should also be a few remarks about CCN and IN number concentration changes as well.

Technical Comments

L. 27. $LNO_x$ needs to be defined. I suggest doing that on Line 23.

L. 53. Insert "a" before "few".

L. 80. "NOx" needs a subscript "x" to be consistent with manuscript.

L. 84. Should it be "emitted NO" or "emitted $NO_x$"?

L. 121. "4" needs to be subscripted.

L. 153-159. This paragraph does not seem to belong in section 3.1 on lightning and $LNO_x$ emissions.

L. 161. Remove "also". I suggest a good proofreading to remove unnecessary "also"s and improve the writing in general.

L. 174. Replace "mixes with" with "along".

L. 189. Insert "of" between "factor" and "two".

L. 190. Move "globally averaged" to after "32%".

L. 192. The figure caption says it is a white isosurface, but on this line it says gray. Be consistent.

L. 200. Remove "Additionally".

L. 276. What are the values of the contour lines?

L. 277. Capitalize “for” at the end of the line. That is, start a new sentence.

L. 310. Remove “-“ and use a comma.

L. 311. Remove “e.g.”

L. 311. Remove “-“ and use a comma.

Lines 322-333. Most of this paragraph discusses the effect of $LNO_x$ emissions on AOD. Is it intended to discuss aerosol extinction in this paragraph too? It is confusing, plus the sentences should be placed after discussing global aerosol extinction changes.

L. 336. Add “of aerosol extinction” after “relative change”.

L. 339. Move “also” to just before “simulated”.

L. 342. Add “of aerosol extinction” after “enhancement”.

L. 350. “lighting” should be “lightning”.

L. 367. Is this sentence discussing tropical South America or all of South America?

L. 369. It seems unnecessary to have both “extra-tropics” and “mid-latitude”.

L. 401. Add a comma after “this”.

L. 404. Insert “by” before “local” and insert “there are” after “but”.

L. 410. Is the increase of 10% for total water content?

L. 413. Insert “ice” before “crystal”.

L. 415. “Further North” of where?

L. 424. It is better to say “model output” rather than “data”.

L. 428. Insert “By” before “analyzing”. Remove “with the help of” and put parentheses around “Fig. 6”, adding a comma after the parentheses.

L. 502. Remove “also”.

L. 503. Change “load” to “loading”.

L. 516. I suggest using “unclear” instead of “ambiguous”.

L. 558. Why is a 2014 paper (Chang et al., ACP) a discussion paper? Please update!

Figure 2. The flashrate units do not match between the color bar on the plot and the figure caption. If the units were flashes per $km^2$ per minute, then it would be easier to compare to satellite data in the literature.

Figure 4. I suggest changing the color bar to have white for the -2% to +2% region. The -5 to -15% colors are so similar it is difficult to see changes. The same is true with -30 to -40% and 18-32%. For such small plots, perhaps it is better to have just 5 colors: red, yellow, white, green, blue, and define broader regions of percent difference.

Figure 4 caption. What are the contour level values?

Figure 5. “Additionally the front panel depicts again relative percentage differences” of what? And no need for both “additionally” and “again”.

Figure 6. It should be “effective radius”. What are the values of the isosurfaces? “substantial absolute changes” is not quantitative.

Figure 6. Why are there ice crystal size changes in the 1000-700 hPa regions where it is usually too warm to support ice?