Reply to reviewer 1 of the
interactive comment on "Chemistry-climate interactions of aerosol nitrate from lightning"

I thank the reviewer for his comments which helped to improve the manuscript.
The reviewer comments are given in italics and my comments are following the individual points.

In the summary, the reviewer requests are stronger focus on the new findings on aerosols. However, as some changes in aerosols and their properties can only be derived from the impact of LNOx on the gas phase these effects have to be mentioned as well. I agree that some of the discussion on gas phase chemistry can be shortened as this has already been the focus on numerous studies.
A major concern is the statistical significance of the results which is addressed in the revised manuscript version. Many of the impacts on aerosols and their properties show are statistically robust despite the model internal and interannual variability.
I agree that the model formulation include a lot of simplifications which lead to model uncertainties, but nevertheless the model is a state-of-the-art chemistry climate model and this study is a first attempt to analyse and quantify the impact of lightning on nitrate aerosols.
The 3D visualisations, even though a bit more challenging for understanding at first glance allow the inclusion of more information in a single graph, i.e. the exact geographical location of isolated three dimensional features which cannot be depicted by 2D cross sections or 2D averages. The updated manuscript therefore still contains 3D graphs, but 2D sections are provided in the supplement which may be more convenient to visualise individual aspects. Additionally, replacing the 3D visualisations would result in substantially more pictures in the manuscript increasing its length and decreasing the readability.

Major points:
1)
There are many discussion points illustrating small differences between simulations with and without LNOx 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 LNOx 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.

The revised manuscript depicts only statistically significant differences. For nitrate almost all upper tropospheric differences show statistical robustness. Using derived aerosol properties and their impacts on the climate system the robustness becomes smaller, especially for liquid water clouds the statistical significance is not reached. Therefore this part is substantially shortened in the revised version.
The isosurfaces in the 3D visualisations always show changes larger than 10%, sometimes even up to 60%, and also the impact on CH4 lifetime for the whole troposphere is larger than 20% (which is also statistically robust). Consequently, we can follow the process chain as depicted in Fig.1 from the NOx source from lightning up to radiative flux perturbations as a result from neglecting LNOx emissions.
I agree that most of the gas phase results have been published before (and the revised manuscript will even include some more references), but few studies allow to draw conclusion about the implications of the gas phase on the aerosol phase. Furthermore, I think that the fact that considering aerosol particles does not substantially change the results from pure gas phase studies is also an important point.

2)
The paper contains discussion on the impact of LNOx 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.

I agree that the results for preindustrial conditions do not provide any new insights and a mostly suitable to determine the internal variability thus providing additional confidence in the robustness of the simulation results. Furthermore, as LNOx emission contribute more to total NOx emissions under preindustrial conditions the effects of lightning are actually even stronger in this scenario.
In the revised version the preindustrial scenario will be discussed in an appendix, and not in every section of the manuscript.

3)
It seems to me that the loss processes should be addressed in this study. The author mentions the formation of NH4NO3 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 NH3 and HNO3 are likely removed by thunderstorms, it seems that there may not be enough NH3 to form NH4NO3 downwind of the storms.

Even though nitrate particles formed near the surface will be mostly removed by wet deposition, similar to HNO3 the formation of NH4NO3 in the upper troposphere is still a real phenomenon. C-ToF-AMS Measurements during ACRIDICON-CHUVA (a campaign in Brazil with the HALO aircraft) taking place in the convective outflow in the upper troposphere report quite often enhanced nitrate values which are highly consistent with enhanced NOx and NOy data. (pers. Communication, J. Schneider, MPI for Chemistry Mainz, Nov. 2016). The stabilising cations in this altitude are either ammonium or potentially potassium from biomass burning. However, the latter source is not included in the presented model simulations, such that NH4+ ions are the only choice for neutralising cations. Furthermore, Höpfner et al., (ACP, 2016) detected NH3 in the gas phase in the outflow of the Asian summer monsoon, which then could also form the NH4+ ions. Consequently, the wet removal of NH3 might not be complete such that sufficient ammonia and subsequent ammonium can be available for the formation of NH4NO3.

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)

As mentioned above I prefer one picture using 3D visualisation over several figures using 2D graphs, since this allows a more consistent representation of the real distributions. I agree that some graphs are too busy, especially Fig.5 and 6 which I have refurbished for the revised manuscript version. Additionally, some more graphs in 2D have been added to the supplement, which are easier to read and therefore potentially for suitable for teaching purposes.
Nevertheless, I personally think that 3D visualisation offers a potential for data analysis which has often been neglected in the past because of higher level of complexity. Usually the authors know the data they plot very well, however, a reader of a manuscript who has no access to the data often is not provided with all information the authors have available to come to their conclusions. This is (in my opinion) slightly improved when 3D visualisation is used.

Specific comments:

Lines 30-37. Is there any observational evidence of NH4NO3 formation downwind of convection?
As mentioned above, observational evidence exists for nitrate from aerosol mass spectrometry, however only the anion has been detected. But as gaseous NH3 is also found in this altitude, the existence for NH4NO3 is very likely.

Lines 107-118. Are aerosol-cloud interactions applied to both resolved and parameterized clouds? Please clarify.
The current convection scheme in the EMAC model system is not aware of aerosol particles (except for vertical transport). Therefore aerosol cloud interactions are only considered for large-scale clouds and detrained cloud water/ice to form cirrus clouds in the anvil regions of convective activity. This is clarified in the revised manuscript version.

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.
The answers to this question is quite extensive. For the current simulation this is of minor importance (in my point of view), but more details on the ACCMIP simulations and the respective emissions can be found in the overview paper of Lamarque et al. (GMD, 2013).

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?
The simulations have been conducted as free running simulations with a climatological SST and sea-ice coverage (AMIP2 climatology). The simulation has been initialised with data for the year 1996, but afterwards the simulation has not been nudged towards a re-analysis product. Therefore, the processes and feedback are not weakened or amplified by an external data source for neither present nor preindustrial conditions. Chemical initialisation has been taken from the same year from a transient simulation within the ESCiMo project (Jöckel et al., GMD, 2016). The emissions have been kept constant for the whole simulation time, but of course include a seasonal cycle.

Lines 151-152. Subsequent papers by Pickering's students have updated the vertical profiles for lightning-NOx 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.
I am aware of the updated LNOx emission profiles, and currently the new distributions are implemented as alternative vertical emission distribution in the modelling system. However, from previous studies with our model system (e.g. Huntrieser et al., 2007 for the TROCCINOX campaing and Tost et al., 2010 for SCOUT-O3-DARWIN) lead to the conclusion that the Pickering vertical distribution scheme in combination with the applied convection and convective transport algorithm result in simulation results which agree well with aircraft observations.

Lines 176-178. It seems that PAN and other organic nitrates and perhaps NO3 should be included as contributors to NOy in addition to NOx, HNO3, and N2O5.
Indeed PAN is very important for long-range transport of NOy, but for the sake of limiting myself to the aerosol phase as much as possible, PAN is not considered in the analysis. Nevertheless, it is part of the chemical reaction system and therefore included in the model simulations. Higher organonitrates are currently lumped together and also contribute to the NOy reservoirs. The total PAN load of the atmosphere decreases by approx. 15% in case that LNOx emissions are neglected. This corresponds

almost perfectly to the reduction in nitrates. However, the upper troposphere reduction of PAN is with 21% substantially lower compared to the nitrate reduction with 32%. This is a consequence of the composition for PAN which does not only require NOy, but also VOC compounds. Consequently, in case LNOx emissions are taken into account, maximum PAN formation is not always limited by the amount of available NOy, but the limitation is caused by the low amount of VOCs in the upper troposphere.

Lines 198-199. How long does it take for NO2 to transform to aerosol nitrate?
The formation time is typically in the order of several hours to a few days, and therefore one of the main conversion term for NOx into NOy, especially in the upper troposphere.

Lines 200-203. It appears that a discussion of the results of Figure 3 was not included.
The discussion of Fig.3 is extended in the revised version.

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.
The loss processes for the N(V) species are tabulated in the supplement to close the budget.

Lines 215-217. It would be nice to see supporting information that NH3 emissions are responsible for lower particulate nitrate concentrations.
Lines 211-217. It seems that NO3, PAN, and other organic nitrates should be included.
The analysis of the corresponding simulation data has led to the conclusions in the manuscript (without analysing PAN in detail). Again, focussing on the aerosol and not the full N(V) budget has led to these shortenings in the manuscript, which I would like to keep this way. The response of PAN has been discussed shortly above.

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)?
Indeed, this way the numbers in this text section are determined.

Lines 232-235. Another recent paper also reports the effect of LNOx emissions on tropospheric ozone burden. Finney et al. (2016) ACP find a 27-30% increase in tropospheric ozone due to LNOx 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.
I agree that the results of (Finney et al., ACP, 2016) are interesting and related to the findings in my study. The increase in the tropospheric burden in my study is around 225 for the whole troposphere; this is in agreement with Labrador et al., 2005. For the upper troposphere the increase is more important with 27% which is comparable to the results from Finney for the whole troposphere. Depending on the model configuration, especially the vertical resolution, the transport of ozone from the stratosphere into the upper troposphere can have substantial impact on the tropospheric ozone burden. However, the effect of total NOx emissions (from all sources) as well as total VOC emissions can have a similar effect on the tropospheric ozone burden, e.g. an increase in isoprene emissions due to a slightly different leaf area index can lead to a substantial increase in the tropospheric ozone burden. Consequently, the model configuration as well as the interplay of various boundary conditions can cause the difference in the total tropospheric O3 burden. In total the EMAC model tends to have a

slight high bias in tropospheric ozone (c.f. Jöckel et al. 2016).

The author may want to also cite Finney et al. (2016) GRL, which discusses the effect of LNOx emissions on ozone among the ACCMIP models.
This study is added in the introduction.

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 LNOx 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 LNOx emissions increase methane lifetime, but in the table caption it says, "increase due to neglect of LNOx 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.
I tend to disagree in this point, as oxidant concentrations fail to consider the recycling potential of oxidants, especially OH. The OH burden of the atmosphere differs only by 10% between the simulations with lightning compared to the simulation without LNOx emissions, whereas the methane lifetime effect is twice that large. For calculating the CH_4 lifetime, the CH_4 and OH concentrations as well as temperature and the second order reaction rate which is also used in the chemistry mechanism of the EMAC model.
The table caption is changed according to the reviewers suggestion.
The CH_4 lifetime is mostly affected by the change in the O_3 mixing ratios, which directly influence the amount of OH produced from O3P (from photolysed O_3) and H_2O. Furthermore, the OH recycling reaction $NO + HO_2 \rightarrow NO_2 + OH$ also directly depends on the available NOx concentration. The impact of the second formation (or better recycling reaction) cannot be directly analysed with considering the OH concentration only (or the HOx concentrations).

Lines 253-257. It is interesting to see that the LNOx 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 LNOx?
As the effects of lightning are strongest in the upper troposphere, but the aqueous phase production is most efficient in the lower troposphere due to the amount of available liquid water (as shown e.g. by Tost et al., ICCP, 2012), the aqueous phase production is less affected by the LNOx emissions. Since less S(VI) is formed via the gas phase, even more S(IV) is available for aqueous phase oxidation. These two effects almost balance each other, such that there are only insignificant effects on aqueous phase sulfate formation.

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.
The only change that is lower than 10% is the change in the sulfate burden. Even though this change is small and in some locations not statistically significant, it is in agreement with the significant changes in the size distribution. Therefore, I came to the conclusion that the reduced sulfate production is of relevance and decided to mention it in the manuscript.

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?
The regions have been selected based on the degree of lightning activity and to a second degree on the changes in nitrate concentrations. Even though continental and oceanic boxes are combined usually the continental boxes dominate the size distribution due to the substantially higher aerosol burden, such that the distributions are representative for continental conditions. Only the marine regions in the Central Pacific and South Atlantic are representative for oceanic conditions. In the revised version the size distribution changes are also checked for statistical significance. This reveals that only fine mode differences in the middle and upper troposphere exhibit robustness.

Lines 312-315. The impact of LNOx on aerosol water uptake is not surprising since most of the LNOx effect is in the upper troposphere where it is quite dry.
I agree and the sentence will be rephrased to explicitly mention this point. Nevertheless, I also checked changes in hygroscopicity and also this revealed very little effects. As AOD is mostly affected by aerosol water, I found it important to analyse whether aerosol water had an impact on both the size distribution as well as the extinction coefficients.

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 HNO3 would be degassed when the ice sublimated, but the author points out that the largest aerosol nitrate enhancement is in the upper troposphere.
I think that nitrate sedimentation plays a minor role here, since this would be represented already in the concentration differences. As stated below, the pattern correlation of the change in extinction corresponds much better to the change in the sulfate concentrations. The change in the extinction led to the analysis of the size distributions and revealed the connection to the sulfate changes.

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.
The frequency distributions help to quantify the differences; however, in this case the plots exhibit only small differences that have not been tested for significance. In the PDFs all data points and not the mean values have been used. I found the results sufficiently interesting to mention, but they have not been conclusive to differentiate cases under present day and preindustrial conditions.

Lines 401-402. Why is the polar latitude cloud coverage changed and is it statistically significant?
The polar cloud coverage change is not statistically significant, even though it is larger than in most other regions. The wording in the manuscript has been misleading.

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.
Changes in CCN and IN can partly be deduced from the aerosol size distributions. As liquid clouds hardly show significant differences, the CCN distributions are most likely also not significant. Concerning the IN, these are not explicitly diagnosed by the model, but crystal formation is parameterised based on several aerosol species. The shape of the regions of significant ice crystal size changes leads to the conclusion that immersion freezing in mixed phase clouds is the dominant process

responsible for the differences.

Technical Comments:
L. 27. LNOx needs to be defined. I suggest doing that on Line 23.
Agreed.

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

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

L. 84. Should it be "emitted NO" or "emitted NOx"?
LNOx emissions are completely put into the tracer mixing ratio of NO.

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

L. 153-159. This paragraph does not seem to belong in section 3.1 on lightning and LNOx emissions.
The paragraph will be rephrased. It should explain the grey isosurface of elevated nitrate concentrations in the lower troposphere, as well as the nitrate mixing ratios in the LNOx emission plumes.

L. 161. Remove "also". I suggest a good proofreading to remove unnecessary "also"s and improve the writing in general.
The manuscript is going to be checked before re-submission. Before final publication the mandatory copy-editing by copernicus will also help to improve the language of a non-native speaker.

L. 174. Replace "mixes with" with "along".
done
L. 189. Insert "of" between "factor" and "two".
done
L. 190. Move "globally averaged" to after "32%".
done
L. 192. The figure caption says it is a white isosurface, but on this line it says gray. Be consistent.
corrected
L. 200. Remove "Additionally".
done
L. 276. What are the values of the contour lines?
The contour lines represent absolute number concentrations, with the thick line representing 1 particle, dashed lines 10^{-1}, 10^{-2}, etc. and continuous lines 10^1, 10^2 etc. particles per cm^{-4}. Zooming into the plot the actual values become visible.

L. 277. Capitalize "for" at the end of the line. That is, start a new sentence.
done
L. 310. Remove "-" and use a comma.
L. 311. Remove "e.g."
L. 311. Remove "-" and use a comma.
done

Lines 322-333. Most of this paragraph discusses the effect of LNOx 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.
This paragraph is rephrased taking the statistical significance into account. First AOD and afterwards extinction are discussed.

L. 336. Add "of aerosol extinction" after "relative change".
done
L. 339. Move "also" to just before "simulated".
done
L. 342. Add "of aerosol extinction" after "enhancement".
done
L. 350. "lighting" should be "lightning".
done
L. 367. Is this sentence discussing tropical South America or all of South America?
Tropcial South America has been meant.

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".
done

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

L. 413. Insert "ice" before "crystal".
done
L. 415. "Further North" of where?
Transition from tropics to mid-latitudes.

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".
done

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!
A final publication of the Change et al., ACPD manuscript is not foreseen. A new manuscript describing the approach is currently under review for Atmospheric Environment.

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 km2 per minute, then it would be easier to compare to satellite data in the literature.
Caption corrected.
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 refurbished and caption corrected.

Figure 5. "Additionally the front panel depicts again relative percentage differences" of what? And no need for both "additionally" and "again".
Caption and figure updated.

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?
Caption and figure updated; most of the changes for liquid clouds have not been significant, as well as ice crystal size changes in non-ice regions.