Reply to reviewer 2 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.

Major 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 NOx 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.

I agree with the reviewer that some of the signals do not appear to be robust. However, I have recalculated the data used in the graphs and in the revised version of the manuscript only the statistical significant changes are displayed. Nevertheless, the changes in aerosol nitrate from lightning (Fig.3) are very robust, such that the significance test shows that they are statistical significant and the graph does not substantially change. Similarly the main features in the changes of the size distributions (Fig.4) are also statistically significant. The updated version of the manuscripts marks all non-significant areas of the plot with hashes. The conclusions drawn from the previous version of the graph remain unchanged since they are focused on the large differences which exhibit statistical significance. Analysing the changes in extinction with respect to statistical significance also reveals that the influence of the lightning emissions (both the additional nitrate formation as well as the sulphate production) are robust compared to the internal interannual variability, such that the influence on extinction remains visible. However, analysing the statistical significance of the differences in column AOD reveals that these changes are mostly not significant. This is also not very surprising since the differences in extinction are located in the upper troposphere, but this region does only contribute to the total AOD to a minor degree (see back panel in Fig.5a), as the extinction rates are a factor of 10 or even 100 smaller compared to the near surface values which dominate the column AOD.
Concerning the changes in effective radius these are hardly significant for cloud droplets, but the statistical significance for the ice crystals is larger. Consequently, the impact of the cloud droplet activation scheme is of minor importance as warm clouds play a secondary role in changes due to lightning emissions. Nevertheless, clouds (also warm clouds) contribute to the statistical noise in the radiative fluxes analysed in Fig.7 such that the regions with significant changes in the radiative fluxes cannot unambiguously attributed to the lightning emissions any more. The updated version of the figure including a correction of a small mistake in the significance test also shows slightly more statistical significance especially in the regions of substantial cooling. Despite the problem with a direct

co-location of sources and effects, the global total effect is robustly negative in all simulations, both for present day and pre-industrial conditions with both warm cloud activation schemes, such that a total cooling effect can in my opinion be determined from the simulations.
I personally have doubts that extending the simulations will substantially improve statistical robustness of the results. Even though mathematically the number of data points included in the significance test scales with the power of 0.5 to the significance and the variability is supposed to not increase, the cloud effects, which still have the highest level of uncertainty with respect to process understanding, substantially contribute to the total radiative effect and this conclusion can already be drawn from the current simulation length. For a continuation of the simulation time the computing resources have not been available, such that the answer here is only speculative.

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 PM2.5 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.

Again I agree, that the impact of lightning on nitrate aerosol, especially the chemical composition the oxidation capacity of the atmosphere and PM have been previously investigated. As correctly stated by the reviewer, none of these simulations included the complex feedback on the dynamics of the atmosphere (as they have been conducted with transport models). I admit that I have not been aware of all these simulations, and I will mention some of them in the introduction. However, several of these studies have been conducted with regional models such that a global perspective cannot immediately be offered. Furthermore, some studies also neglect the particulate phase. I have shown that the impact of the particulate phase on the gas phase is minor, however the inversion of this statement is not the valid, e.g. the Allen et al. Study analyses the impact of lightning on nitrate deposition, but does not take the consequences for sulphate explicitly into account.
In the revised introduction, the complex interactions in this study allowing the multi-directional feedback between lightning, gas phase chemistry, particulate phase and the impact on the dynamics of the atmosphere via radiation and cloud processes will be better elucidated to show the novelties of the current study.

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.
I think that the 3D visualisation offers the benefits of displaying in more detail regions of interest. I agree that some of the graphs are more difficult to understand at first glance in contrast to some 2D visualisations, but they offer the potential to include more information in the same number of graphs. Otherwise, a substantial increase in the number of figures would be required to show all the conclusion drawn in the analysis, which is all included in the individual 3D visualisations. Fig.5 and Fig.6 are revised due to the results based on the statistical significance (see above) such that some of the information has been removed from the figures allowing a better visibility of the main features. Additionally Fig.5 has been replaced by two graphs, one showing the main statistical significant regions in combined two dimensional structures and the other one depicting the 3D structure of the impact of lighting on extinction.

Specific comments:
• *P2; L47 - "this" indirect effect*
corrected
• *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.*
This study is not such a strong annihilation scenario comparing simulations with nitrate to those without any nitrate; nevertheless it is still an annihilation scenario with respect to LNOx emissions.
The formulation is rephrased. I agree that the disturbance is still large due to the impact on the chemical regimes and hence the oxidation capacity of the atmosphere. A disturbance study would be better suited, e.g. 2 Tg LNOx emissions, 5 Tg LNOx emissions and 8 Tg LNOx emissions. However, the computation time for this study (which already encompassed 80 years of simulation time with a comprehensive chemistry climate model including gas and cloud phase chemistry as well as aerosol particles) has been limited such that these sensitivity studies could not have been conducted. Furthermore, for these cases most likely the signal-to-noise ratio would have been even worse such that no conclusion might have been drawn from these simulation results.

• *Section 2.3 - How many years was each simulation initialized over? Is methane prescribed or allowed to respond to the large changes in OH?*
The simulation has been initialised with data from a comprehensive transient simulation (Jöckel et al., GMD, 2016) such that no additional spin-up phase has been conducted. Methane is prescribed at the surface with observed concentrations such that the change in the loss rates is partially dampened by additional pseudo-emissions. Therefore, the direct changes in the oxidants have not been reported, but the impact on the CH4 lifetime as a measure of the oxidation capacity as this quantity is less dependent on the actual methane concentrations.

• *P3; L82 - comma should be before "we" instead of "that"*
corrected

• *P5; L132 - nitrate "precursors" from*
reformulated

• *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:10.1029/2009JD011880ls PDF) and bottom-up modeling studies (Koshak et al., 2014; doi:10.1016/j.atmosres.2012.12.015).*

Even though unimodal distributions are found to give a better representation of the distribution of LNOx in present day studies, C-shaped profiles are found to give realistic results (at least in agreement with measurement campaigns (e.g. SCOUT-O3-Darwin (see Tost et al., 2010), TROCCINOX (Huntrieser et al., 2007)). As this is the current implementation of the vertical LNOx emission distribution function, this is not going to be changed. However, I will mention the Ott and Koshak studies.

• *P5; L161 - "also" should be moved to before "are"*
corrected

• *P8, L251 - do you mean "oxidation capacity of the upper troposphere"?*
The reduction to 50% corresponds to the upper troposphere only. The total oxidation capacity of the atmosphere is not affected this drastically.

• *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).*
The changes in the size distribution in the lower part of the troposphere are found not to be statistically significant in neither South America nor Indonesia. Even though some significant changes are found in Central Africa, I do not see a direct link to a convective invigoration. On all three tropical continents the majority of the lower tropospheric aerosol particles result from biomass burning and SOA formation. The contribution of nitrates from lightning is small compared to the other sources in the lower troposphere and the feedback via the oxidation capacity and oxidative ageing of organic aerosols to increase their hygroscopicity and therefore cloud formation potential is not included in the model.

• *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.*
Fig.5 and 6 are revised. The left hand side of Fig.5 still uses the panels on the front and the back side of the cube – but I rather would not change the terminology into the geographic directions to avoid misunderstandings with regions on the globe.

• *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?*
Most of the oxidation of tropospheric aerosol precursors happens closer to the sources, which are located at the surface for most aerosol precursors (SO2 from anthropogenic and NOx from anthropogenic and biogenic sources). In the lower part of the troposphere the oxidation capacity is not that substantially affected by lightning as in the upper troposphere. Especially sulphate formation is dominated by aqueous phase production, and the transfer of the oxidants into the aqueous phase is affected only to a minor degree.
I have analysed the sulphate production pathways in a different simulation scenario (without dynamical feedback) and have seen changes on the order of a few percent only.

• P15; L497 - comma should go after "NH3"
corrected

• *P15; L499 - please clarify if you mean to the global aerosol nitrate burden, or the local upper troposphere*
This statement is mostly valid for the upper troposphere and this is added in revised manuscript.

• *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 CH4 + OH reaction. This may underrepresent the importance of lightning on the extratropics.*
I prefer methane lifetime, since it is less dependent on the total CH4 concentrations. Furthermore, the recycling of oxidants especially OH via various reaction pathways cannot be well represented in oxidant concentrations. The difference in the OH burden is on the order of 10% only, whereas the CH4 lifetime for the troposphere has a magnitude of more than 20%, which results from the recycling potential. Consequently, CH4 lifetime is a better estimate for the oxidation capacity.

• *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.*
The axis labels are pressure altitude on the y-Axis and aerosol diameter on the x-Axis. The statistical significance has been added to the plots such that the important changes can be easier visualised. The Figure caption is changed to include this information as well.

• *Fig. 7 - the units for the y-axis are missing (W m−2 ?)*
Both the color bar and the y-axis of the line plot depict the flux perturbation in W/m^2. This is mentioned below the color bar and at the upper edge of the y-axis.

• *Table 1 - "Differences due to LNOx emissions" is ambiguous toward its directionality. I'd recommend using "Estimated contribution from lightning emissions".*
As the difference can potentially be negative as well, I prefer the difference due to LNOx emissions. Furthermore, due to the complicated feedback, a contribution from lightning emissions might be misleading as the results can also be consequences of chemical feedback processes.