Authors response file for

Chemistry climate interactions of aerosol nitrate from lightning by H. Tost

All of the reviewer comments have been considered and replies have been formulated. These replies as well as a latexdiff track change compared to the ACPD manuscript version has been attached.
The manuscript has seen some modifications, especially most of the figures have been revised to take the statistical significance of the results into account. Some discussion has been extended and the results for the preindustrial simulations have been merged together into Appendix A.

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 CH_4 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 CH4 lifetime, the CH4 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 CH4 lifetime is mostly affected by the change in the O3 mixing ratios, which directly influence the amount of OH produced from O3P (from photolysed O3) and H2O. Furthermore, the OH recycling reaction NO + HO2 -> NO2 + 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.

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.

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Chemistry-climate interactions of aerosol nitrate from lightning

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Abstract. Lightning represents one of the dominant emission ~~source~~ sources for NO_x in the troposphere. The direct release of oxidised nitrogen in the upper troposphere does not only affect ozone formation, but also chemical and microphysical properties of aerosol particles in this region. This study investigates the direct impact of LNO_x emissions on upper tropospheric nitrate using a global chemistry climate model. The simulation results show a substantial influence of the lightning emissions on the mixing ratios of nitrate aerosol in the upper troposphere of more than 50%. In addition to the impact on nitrate, lightning substantially affects the oxidising capacity of the atmosphere with substantial implications for gas phase sulphate formation and new particle formation in the upper troposphere. In conjunction with the condensation of nitrates, substantial differences in the aerosol size distribution occur in the upper troposphere as a consequence of lightning. This has implications for the extinction properties of the aerosol particles and for the cloud optical properties. While the extinction is generally slightly enhanced due to the LNO_x emissions, the response of the clouds is ambiguous due to compensating effects in both liquid and ice clouds. Resulting shortwave flux perturbations are of $\sim -100 \mathrm{mW/m^2}$ as determined from several sensitivity scenarios, but an uncertainty range of almost 50% has to be defined due to the large internal variability of the system and the uncertainties in the multitude of involved processes. Despite the clear statistical significance of the influence of lightning on the nitrate concentrations, the robustness of the findings gradually decreases towards the determination of the radiative flux perturbations.

1 Introduction

Lightning is one of most energetic phenomenon in the Earth atmosphere. Due to the tremendous electricity, the associated temperatures allow for breaking up the stable molecular nitrogen compounds into fragments which partly recombine to nitrogen oxides (Schumann and Huntrieser, 2007). Hence lightning represents a natural atmospheric emission source for NO_x, in addition to the anthropogenic sources from industry, energy production, traffic and agriculture (e.g., Jaegle et al., 2005). Furthermore, biomass burning contributes significantly to the total NO_x emissions (e.g., van der Werf et al., 2010). The global lightning NO_x (LNO_x) production has been estimated to range between 2 and 8 Tg N/yr (Schumann and Huntrieser, 2007), hence being in a similar order of magnitude as the soil emissions (e.g., Steinkamp and Lawrence, 2011), which represent an additional important natural contribution to the total oxidised nitrogen in the atmosphere. In contrast to the other sources (with the exception of aircraft emissions), LNO_x represents an upper tropospheric source. Due to a different chemical composition and chemical reactivity compared to the boundary layer, chemical conversion into HNO_3 is relatively efficient and most important is not subject to fast removal by dry deposition. Once nitric acid is formed this can condense on existing aerosol particles, mostly thermodynamically stabilised by ammonium (NH_4^+), forming ammoniumnitrate (NH_4NO_3). ~~However, this~~ This aerosol species is considered semi-volatile such that in the lowermost atmosphere a substantial part of nitrate re-evaporates (e.g., Stelson et al., 1979). However, due to the lower temperatures in the upper troposphere, NH_4NO_3 is thermally stabilised, and remains mostly in the aerosol phase. ~~However, the~~ The total amount of nitrate aerosol does not only depend on the available nitric acid, but also neutralising cations, i.e. mostly ammonium.Furthermore, the condensation rate of nitric acid forming nitrates also depends on the available aerosol concentration, as well as competition for neutralising compounds with e.g. sulphate ions.

Experimental evidence for the occurrence of nitrate in convective outflows have recently been observed over the Amazon by aircraft measurements [1]. Furthermore, NH_3 has been observed based on satellite retrievals in the outflow of the Asian monsoon (Höpfner et al., 2016), leading to the conclusion that not all soluble compounds are completely removed by scavenging during convective lifting. Consequently, there is observational evidence for both NH_3 and NO_3^-, such that the formation of NH_4NO_3 is likely due to the low temperatures in the upper troposphere.

Being a component of the mixture of aerosol particles, the nitrate aerosol from lightning can also influence atmospheric radiation via the direct and indirect aerosol effects by a multitude of pathways. First, nitrate contributes to water uptake increasing ambient aerosol size at a given relative humidity. Both the particulate nitrate mass as well as the additional aerosol water enhance the radiative extinction properties of the particle and can even alter the single scattering albedo influencing the direct aerosol effect of nitrate particles (e.g., Adams et al., 2001).

Additionally, the overall chemical composition of the aerosol is modified by nitrate from lightning such that the cloud condensation nuclei (CCN) or ice nuclei (IN) efficiency of the particles changes with implications for the indirect aerosol effects. Consequently, ~~the~~ this indirect effect should be considered a competition between the formation of additional CCN (e.g., Makkonen et al., 2012)

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[1] J. Schneider, pers. comm., 2016

by the nitrate and a potential deactivation of IN for contact or deposition freezing by providing a hygroscopic coating. Immersion freezing is ~~only affected to a minor degree~~ also affected via the concentration of the solutions~~and hence~~ , IN deactivation and freezing point modifications.

Currently, only a few global aerosol chemistry climate models can realistically simulate aerosol nitrate and its implications for the direct and partially also indirect aerosol effect due to the complex chemical interactions and the semivolatility (e.g., Jacobson, 2001; Adams et al., 2001; Bauer et al., 2007; Bellouin et al., 2011; Xu and Penner, 2012; Makkonen et al., 2012). However, regional scale air quality models often consider the formation of nitrate and also implications of the emissions from various sources on the NO_3^- formation, e.g. Zare et al. (2014) , who investigate the role of LNO_x on PM2.5 in Europe or Allen et al. (2012) , who among other aspects analysed the influence of lightning on nitrate wet deposition in the US. The effect of LNO_x on gas phase chemistry and the oxidation capacity of the atmosphere has been previously studied and is continuously revisited (e.g., Labrador et al., 2004; Schumann and Huntrieser, 2007; Gressent et al., 2016; Finney et al., 2016a, b) .

Usually, the effect of nitrates is determined from *annihilation studies*, i.e. the effect of nitrates is completely ignored. However, this might have implications for the overall aerosol chemical composition and size distribution such that these effects are mixed with the pure impacts of nitrate. Instead of a total nitrate *annihilation scenario*, this study investigates the omission of only a fraction of the aerosol nitrate, which originates from the lightning NO_x emissions. According to our knowledge, the resulting climate impacts from lightning caused by particulate nitrate have not yet been quantified with a comprehensive chemistry climate model. Nevertheless, this scenario reveals already strong impacts due to the substantial implications of LNO_x emission on tropospheric chemistry. However, emission sensitivity studies on LNO_x would suffer from an even worse signal-to-noise ratio than found in the current study.

The results from this study are in agreement with previous studies as mentioned above with respect to the impact on ozone and the oxidation capacity, but explicitly analyse the impact of LNO_x on upper tropospheric nitrate concentrations and its implications for the climate system.

2 Model description

2.1 The EMAC modelling system

This study applies the ECHAM5/MESSy Atmospheric Chemistry (EMAC) model, which is a numerical chemistry and climate simulation system that includes sub-models describing tropospheric and middle atmosphere processes and their interaction with oceans, land and human influences (Jöckel et al., 2010). It uses the second version of the Modular Earth Submodel System (MESSy2) (Jöckel et al., 2005) to link multi-institutional computer codes. The core atmospheric model is the 5th generation European Centre Hamburg general circulation model (ECHAM5, Roeckner et al., 2006). For the present study we applied EMAC (ECHAM5 version 5.3.02, MESSy version 2.50) in the T42L31-resolution, i.e. with a spherical truncation of T42 (corresponding to a quadratic Gaussian grid of approx. 2.8 by 2.8 degrees in latitude and longitude) with 31 vertical hybrid pressure levels up to 10 hPa. The applied model setup comprised the submodels for radiation, convection, large-scale clouds and condensation and the budget (source, transport and loss processes) of chemical compounds in the gas and aerosol phase.

2.2 Aerosol climate processes within EMAC

To simulate the relevant processes mentioned above and sketched in Fig. 1, we employed the lightning ~~NOx~~ NO_x emissions scheme by Price and Rind (1992). This scheme uses convective cloud top height as simulated by the convection scheme (Tiedtke, 1989). Even though such a combination of two parameterisations is subject to large uncertainties, we have shown in the past ~~,~~ that this special combination is relatively robust and able to provide relatively realistic lightning distributions compared to satellite data from the LIS/OTD sensors (Tost et al., 2007). The emitted NO is subject to gas phase chemical transformations which are calculated with the help of the chemical model MECCA (Sander et al., 2011). The resulting nitric acid (HNO_3) can subsequently partition into the aerosol phase. The corresponding aerosol processes are simulated with the GMXE aerosol submodel (Pringle et al., 2010; Tost and Pringle, 2012) taking both the gas-aerosol phase partitioning and the interactions with other chemical compounds as well as the microphysical properties of the aerosol particles into account. The gas-aerosol phase partitioning of inorganic semivolatile compounds is calculated by the ISORROPIA2 model (Fountoukis and Nenes, 2007), which is part of the GMXE aerosol scheme. The aerosol particles are discretised in 4 lognormal size categories, and for the larger three modes a distinction is done between internally mixed hydrophilic particles and externally mixed hydrophobic particles resulting overall in 7 lognormal modes. Additional anthropogenic and natural emissions except for lightning are simulated with the submodels ONEMIS, OFFEMIS and TNUDGE (Kerkweg et al., 2006b) submodels, providing sources for other primary and secondary aerosol particles. Physical loss processes (dry and wet deposition, additionally sedimentation for aerosol processes) are explicitly considered in the schemes DRYDEP, SCAV and SEDI (Kerkweg et al., 2006a; Tost et al., 2006).

To consider aerosol radiation interactions the prognostic aerosol mass and number concentrations (including aerosol water) are used in lookup tables from Mie calculations precalculated with the LIBRADTRAN (Mayer and Kylling, 2005) to determine the radiative properties of the atmospheric aerosol (extinction, single scattering albedo, asymmetry parameter) as described in detail in Pozzer et al. (2012); de Meij et al. (2012) and Dietmüller et al. (2016). These parameters are explicitly considered in the radiation scheme native to ECHAM5 (Roeckner et al., 2003) replacing the aerosol climatology by Tanre et al. (1984) as described in Dietmüller et al. (2016). For the treatment of indirect aerosol effects, we have implemented a two moment cloud microphysics scheme (Lohmann and Hoose, 2009; Lohmann et al., 2010). The activation of aerosols is calculated with the scheme of Abdul-Razzak and Ghan (2000), which has been adapted to the simulated aerosol types. Furthermore, the interactions of aerosols in the homogeneous and heterogeneous freezing processes (Kärcher et al., 2006) are considered, including an adaption to the more comprehensive chemical composition of the aerosol simulated with GMXE. To investigate the sensitivity of the climate impacts of aerosol particles influenced by lightning caused by aerosol cloud interactions (ACI), a second set of simulations has been performed using a modified activation scheme based on a combination of the work of Abdul-Razzak and Ghan (2000) and Petters and Kreidenweis (2007), as described in detail by Chang et al. (2014). The set of simulations using the Abdul-Razzak and Ghan (2000) aerosol activation scheme will afterwards be abbreviated with ARG, for the other set of simulations following Chang et al. (2014) the KK (i.e. kappa-koehler) acronym will be used.

Both schemes include the effects of nitrate, but different approaches are followed to calculate the critical supersaturation: the ARG scheme uses parameters for the osmotic coefficient and dissociation of nitrate in the solution (which are more uncertain than for $(NH_4)_2SO_4$), whereas the KK approach uses a volume weighted κ to determine the total aerosol water uptake and hence cloud formation potential. The two approaches are therefore merely a different representation of the Raoult effect in the cloud activation. Note, that the aerosol-cloud interactions are only taken into account for large-scale clouds as the treatment of cloud microphysics in convective clouds is even further simplified.

2.3 Simulation Setup

~~We performed decadal~~ Decadal simulations including all feedback mechanisms with the EMAC model have been performed for present day and preindustrial conditions. Sea surface temperatures are prescribed by a climatology from the AMIPII database for all model configurations. To determine the effects of aerosols caused by lightning two simulations are performed for each scenario, one with and one without LNO_x emissions. Even though this annihilation scenario potentially cannot capture some compensation effects, we choose this approach due to the expected lower signal to noise ratio. Furthermore, as there is nitrate produced from anthropogenic, biomass burning and soil sources of NO_x, there is no complete annihilation of all nitrate. For the present day scenario we applied emissions from the ACCMIP emission inventory ~~(Lamarque et al., 2010)~~ (Lamarque et al., 2010, 2013) for trace gases and aerosol emissions from the AEROCOM (Dentener et al., 2006) experiment, for preindustrial conditions we followed the AEROCOM (Dentener et al., 2006) recommendation for preindustrial conditions. Biomass burning is included using the GFED data (van der Werf et al., 2010), as well as a compilation for preindustrial biomass burning. Note, that we applied prescribed aerosol emissions for dust and sea salt, not including a potential feedback of a changed circulation (wind speed and wind patterns) on aerosol sources. The model simulations have been initialised with results from previous experiments (see Jöckel et al. (2016)) eliminating spin-up effects. Due to the comprehensive feedback mechanisms we refrain from comparing individual years of the simulations, but focus on the decadal mean values and distributions.

3 Results

3.1 Lightning and associated emissions

To estimate the effects of lightning and associated emissions, we have analysed the distribution of NO_x emissions from lightning. Fig. 2 depicts in a 3D visualisation the temporal mean emitted LNO_x for the present day scenario.

The magnitude of the LNO_x emissions is displayed as an isosurface of $1 \cdot 10^{-16}$ kg/(m^3 s). A second isosurface of $3 \cdot 10^{-16}$ kg/(m^3 s), which is visible as a darker shading embedded in the first isosurface, shows that the dominant emissions take place in the upper part of the LNO_x plume. This is a consequence of the fact that the vertical emissions redistribution follows a C-shape profile according to Pickering et al. (1998), leading to enhanced emissions in the upper section of the plume. Even though more recent studies (e.g., Ott et al., 2010) suggest a different vertical emission distribution, past studies with the EMAC model have shown good agreement using the C-shape profiles (Huntrieser et al., 2007; Tost et al., 2010) .

The ~~colour coding of the isosurface displays the~~ outer emission isosurface is colour coded with the total aerosol nitrate mixing ratio (in mol/mol), depicting the amount of aerosol nitrate present ~~at~~ in the emission peaks. Furthermore, the gray shaded isosurface visualises ~~an isosurface~~ a level of 0.1 ppb$_v$ of aerosol nitrate. This shows that ~~even though the higher~~ high aerosol nitrate concentrations are located in the lower troposphere, but also enhanced mixing ratios (>100 ppt$_v$) of particulate nitrate can be found in the upper troposphere, partially concurring with the LNO_X emissions ~~. The higher~~ (compare the values of up to 100 ppt$_v$ as marked by the color coding on the emission plume surfaces).

The enhanced NO_3^- mixing ratios ~~closer to the surface~~ in the lower troposphere can be explained by the ~~higher~~ anthropogenic NO_x emissions as well as sources for neutralising cations such as NH_4^+ close to the surface.

The coloured map at the bottom of Fig. 2 depicts the mean flash frequency per ~~second~~km^2 and minute, which is relatively well confined to the tropical continents and in reasonable agreement with observations from LIS/OTD (updated data following Christian et al. (2003)). Consequently, ~~also~~ the LNO_x emissions are co-located over the same regions.

The total LNO_x emissions are 5.95 Tg N/yr with a 1σ-variability of 0.03 Tg N/yr over the simulated decade in the present day scenario (ARG), for the KK scenario a total emission strength from lightning with 6.04 ± 0.03 Tg N/yr is simulated. Note that the seasonal variability is substantially larger. ~~In the preindustrial scenario total emissions of 6.12 ± 0.03 Tg N/yr are simulated.~~ More than

80% of the emitted LNO$_x$ is placed above 500 hPa altitude in both scenarios. The difference between the two scenarios is a consequence of slightly different meteorological conditions which are caused by the feedback effects of the aerosol and cloud properties. ~~For the KK scenario again the feedback slightly alters the emissions from lightning with 6.04 ± 0.03 and 6.16 ± 0.03 Tg N/yr for present day and preindustrial conditions, respectively, with also more than 80% of the emitted above 500hPa.~~

3.2 Tropospheric nitrate from lightning

3.2.1 Chemical budgets and distribution of oxidised nitrogen compounds

The emitted NO$_x$ from lightning ~~mixes~~ along with other NO$_x$ molecules in the atmosphere ~~forming~~ forms HNO$_3$ mostly via the gas phase reaction of $NO_2 + OH \rightarrow HNO_3$ ~~.~~ on typical time scales of hours to days. Even though this reaction is much slower than the conversion between NO and NO$_2$, it represents an important end of the circular conversion of the NO$_x$ species.

The LNO$_x$ emissions for the present day scenario result in $\sim$ 40% higher nitric acid mixing ratios in the troposphere, and even up to $\sim$ 61% between 500 hPa and the tropopause, compared to simulations without lightning NO$_x$ emissions (see Tab. 1). Even though the enhancement effect for N$_2$O$_5$ is stronger in relative numbers, the overall mixing ratios for N$_2$O$_5$ are substantially smaller (almost two orders of magnitude) such that dinitrogenpentoxide plays a minor role for the highly oxidised nitrogen compounds in the troposphere.

~~During preindustrial times, this effect is even more amplified with $\sim$ 67% and $\sim$ 76% increases for in the total and upper troposphere, respectively. This stronger enhancement is due to the larger contribution of the~~ PAN is found to see a reduction by 15% for the whole troposphere and 21% in the upper troposphere when no lightning emissions are considered, therefore being less affected by the LNO$_x$ emissions ~~compared to the total release, which is mostly from anthropogenic sources in present day conditions. Enhancements for are of $\sim$ 80%, but also under these conditions is a minor contributor to the total highly oxidised nitrogen tropospheric load (less than 5%)~~due to a potential limitation of VOCs for PAN formation.

The nitric acid molecules can condense on pre-existing aerosol particles forming NO$_3^-$ ions in the deliquesced aerosol solution or solid NH$_4$NO$_3$ or NaNO$_3$ crystals. Tropospheric nitrate mixing ratios are typically a factor of two to three lower compared to HNO$_3$. Nevertheless, the changes in upper tropospheric NO$_3^-$ mixing ratios are ~~globally averaged $\sim$ 32%~~$\sim$ 32% globally averaged.

Fig. 3 shows isosurfaces of tropospheric nitrate concentration differences between the simulation with LNO$_x$ emissions to the case without this source. The ~~grey~~ gray shaded isosurface depicts a 30% difference, the blue isosurface 45% differences and the red enclosed area 60% enhanced NO$_3^-$ mixing ratios, with only statistically significant (based on a two sided t-test of annual mean data with a significance level of 90%) data shown. Even though the maximum NO$_3^-$ absolute differences occur down to the surface, the largest relative differences are apparent in the UT. Most of these differences are constrained to the tropics where the strongest LNO_x emissions are prevalent. However, the maxima in the differences are not directly co-located with the emission maxima (i.e. Central Africa, Amazonia and the maritime continent, see Fig. 2), but generally further downwind. This is a consequence of atmospheric transport during the time required for oxidation of NO_x to HNO_3 and subsequent partitioning into the aerosol phase.

To provide an estimate of the total amount of nitrate in the upper troposphere the back panel of Fig. 3 depicts the zonal mean NO_3^- mixing ratio with values between 10 and 100 ppt$_v$ for the tropical middle and upper troposphere where the impact of the LNO_x emissions is largest.

~~Additionally, the mean upper~~ The mean upper tropospheric column burden (500 hPa to the tropopause) in mg/m^2 is depicted by the coloured panel at the bottom of the plot. The turquoise isolines on this panel depict the 20%, 40% and 60% differences in the UT column burden between the simulations with and without LNO_x emissions. These are of course co-located with the regions of the isosurfaces.

~~A similar figure for preindustrial conditions~~ The figure shows that the tropical upper troposphere is a region in which substantial amounts of particulate nitrate can be found~~in the supplement. Due to the lower other sources of in the atmosphere, the importance of the emissionsis substantially increased. Therefore,~~. Both visualisations of the ~~relative differences in are larger which can be seen from the extended areas included by the respective isosurfaces and the larger areas covered by the turquoise contour lines at the floor panel of the figure. The lower overall nitrate mixing ratios are obvious from the colour scale of the column nitrate which is almost an order of magnitude lower compared to present day conditions~~differences (UT column burden and mixing ratios) show that a large contribution of the upper tropospheric NO_3^- originates from lightning emissions, but also that a sufficient amount of neutralising cations is available to stabilise the nitrate in the particulate phase. These findings are a direct consequence of the emission and conversion of NO_x to N(V), but of course are also indirectly affected by changes in the oxidation capacity of the atmosphere and hence the HNO_3 formation.

The tropospheric budget of the highly oxidised N compounds is summarised in Tab. 1, including the relative importance of the LNO_x emissions. For present day conditions, the particulate nitrate contribution to the total N(V) is ∼ 25%, whereas in the upper troposphere it is only ∼ 13%. ~~For preindustrial conditions a lower contribution of particulate nitrate to the total N(V) load with 22% for the whole and 9% for the upper troposphere is simulated. The lower contribution for the preindustrial conditions can be mostly attributed to lower emissions and consequently less ions to neutralise and thermodynamically stabilise available aerosol nitrate.~~

The neglect of the LNO_x emissions leads to a shift in the contribution of the particulate phase to the total N(V). Under these conditions 33% of the N(V) is in the form of particulate NO_3^- for the whole troposphere, whereas for the UT the fraction is higher than 21%. This is a consequence of the reduced available N(V), but a similar amount of neutralising cations, i.e. mostly NH_4^+. ~~For preindustrial conditions this enhancement of the particulate phase is even stronger to $\sim$ 34% for the whole troposphere and 17% for the UT.~~

The sensitivity simulations in the KK configuration result in almost identical values, (budget is shown in Tab. 1 of the supplement) depicting the fact that the description of cloud and cloud removal processes plays a minor role for the total budget of particulate nitrate in both model configurations.

The budget of the loss processes of N(V) compounds can be found in the supplement for both present day and preindustrial conditions. The differences in the nitrate burden are similarly present in the loss processes, with wet deposition being mostly influenced (50% change in the loss flux), but sedimentation and dry deposition also equally contribute to the change in the total loss.

3.2.2 Influences on other chemical species

The reduced NO_x burden in the simulation without lightning emissions has substantial impact on the concentrations of other species. As only a minor fraction of the total LNO_X emissions of $\sim$ 6 Tg N/yr are converted to N(V+) (e.g. 186 Gg N change in N(V) according to Tab. 1), the impact on ozone is comparable to previous studies ~~(e.g., Labrador et al., 2005)~~ (e.g., Labrador et al., 2005; Finney et al., 2016a) : the calculated values for tropospheric ozone are 362 Tg in the simulation without lightning and 444 Tg if lightning is included, which corresponds to a 22% increase in the tropospheric ozone burden ~~. This effect~~ (even though the absolute values for the tropospheric ozone burden differ substantially in each of the studies). The effect of the LNO_x emissions is again very prominent in the upper troposphere, where the O_3 load increases from 195 to 248 Tg by LNO_x emissions, i.e. an increase of 27%. Driven by the location of most of the emissions, these effects mostly occur in the low latitudes. The resulting decrease in ozone in the zonal average resulting from neglecting lightning emissions is comparable to the findings of Grewe (2007). The change in the column burdens in the tropics also agrees with the results of Martin et al. (2002), both in the geographic patterns and the amplitude of the signal. Therefore concluding, the explicit consideration of the nitrate formation has no important impact on the tropospheric ozone distribution, such that the results of impact studies of lightning NO_x (such as e.g., Banerjee et al., 2014) do not have to be revised.

However, the OH concentrations and therefore the oxidation capacity of the atmosphere are also substantially influenced by lightning (Labrador et al., 2004) via changes in O_3 and subsequent OH production via ozone photolysis and reaction of the products with water vapour. The effect on OH is displayed by a modification of the methane lifetime, as depicted in Tab. 2. The emissions of LNO_x are responsible for an increase of the tropospheric methane lifetime of $\sim$ 1.7 to 1.9 years; however in the upper troposphere (above 500 hPa up to the tropopause) an increase of the CH_4 lifetime of almost 10 and 13.2 years for present and preindustrial conditions, respectively, corresponding to almost a halving of the oxidation capacity of the ~~atmosphere~~upper troposphere.

This is especially relevant for the production of sulphate via the gas phase oxidation of SO_2, which substantially influences aerosol formation and aerosol composition in the upper troposphere. The sulphate burden for present day conditions decreases from 576 Gg S(VI) to 565 Gg. This is also prominent in the UT region, where instead of 74.6 Gg S(VI) only 70.2 Gg S(VI) are simulated. ~~For preindustrial conditions a similar reduction in the sulphate load is calculated without emissions despite the substantially lower total emissions.~~ As most of the oxidation in the upper troposphere takes place in the gas phase, the aqueous phase oxidation in the lower troposphere is affected to a minor degree.

Particulate ammonium mixing ratios are only affected to a minor degree by the LNO_x emissions: for present day conditions the NH_4^+ burden slightly decreases without lightning emissions, but increases during preindustrial times. However, the changes in the atmospheric burden are lower than $\pm 5\%$. These changes can be explained by the of lower sulphate and nitrate burdens and effects of the oxidation capacity on gaseous ammonium concentrations. A significant change due to LNO_x emissions in the sulphate to bisulphate ratio is not simulated by the model despite the capabilities of the thermodynamic equilibrium model.

The KK simulations show a very similar behaviour in the burdens, the CH_4 lifetime and the changes induced by the omission of the LNO_x emissions, such that a detailed discussion is skipped here.

3.3 Aerosol microphysical effects

The influences of the chemical composition in the upper troposphere due to lightning are also reflected in the microphysical properties of the aerosol, which are described by the size distributions at various locations. The regions in the tropics are mostly characteristic for the tropical continents, as the oceanic aerosol distributions usually contain substantially lower particle concentrations such that the distribution of those regions is dominated by the continental grid cells. The regions have been selected following the analysis of the significant changes in Sect. 3.2.1, but also to represent various conditions of typical aerosol size distributions where lightning can play an important or only a minor role.

The temporal mean size distributions of the aerosol particles for various regions (i.e. those regions covered by the plot on the map) are shown in Fig. 4 as a 2D plot with the horizontal axis depicting the ambient aerosol diameter and the vertical axis the pressure altitude; the colour coding displays the relative difference in aerosol number concentration [in %] between the simulation with and without lightning. Furthermore, the absolute values of the size distribution [in cm^{-3}] are depicted by the contour lines with the solid lines representing concentrations larger than 1 per cm^3. The figure shows the simulation results for the present day conditions (ARG case)~~, for preindustrial conditions a similar figure can be found in the supplement~~. The hatched regions in the 2D size distribution diagram mark those areas in which the changes do not exhibit statistical significance.

Over the tropical continents in the upper troposphere enhanced nucleation mode particle numbers are simulated if lightning emissions are considered, which is a result of the enhanced S(VI) concentrations and consequently enhanced new particle formation. At approximately 10 nm diameter the particle number is smaller in case of $\mathrm{LNO_x}$ emissions, due to the interaction of enhanced coagulation and condensation on the enhanced particle concentrations. Also additional condensation on existing particles and enhanced coagulation with the small particles causes the reduction of particle numbers at $\sim$ 80 nm. For larger particles a slight reduction is found as a consequence of the more efficient coagulation (due to higher particle numbers in the small mode) versus the slower condensational growth. For the lower troposphere the picture is more ambiguous and most changes are not statistically significant: in South America and Indonesia reduced nucleation takes place. However, the absolute particle concentrations are so low that this process is almost negligible, whereas in Central Africa enhanced new particle formation is caused by lightning emissions. For the larger particles the impact is rather small.

In the mid-latitudes relevant changes in the size distributions are only found in the Eastern US and China but little impact in Europe and Siberia. The first two regions are located slightly more southwards such that lightning frequencies are enhanced compared to the latter two regions (see bottom map of Fig. 2. The general pattern of the changes is comparable to the tropical continents.

In the Southern Atlantic region the changes in the size distribution profile are relatively small, as lightning emissions play a minor role for the total particle concentration as well as for the oxidation capacity of the atmosphere and therefore sulphate formation, but they are still robust. In the Central Pacific where enhanced nitrate concentrations are simulated in the upper troposphere (see Fig. 3), the changes in the aerosol size distribution are moderate with strongest signals in the middle troposphere (between 700 and 500 hPa). As lightning $\mathrm{NO_x}$ emissions are substantially smaller over the ocean, the oxidation capacity is affected to a minor degree; however nitrate and gaseous N(V) are transported downwind from the source region affecting the nitrate concentrations and particle numbers, mostly via coagulation.

~~For preindustrial conditions (see supplement) the simulation provides similar results from the tropics. However, in the mid-latitudes where a substantial reduction of other emissions is applied compared to the present day scenario the impact of lightning on the nitrogen budget as well as the size distribution becomes more important. Obviously, a similar pattern as for the tropical continents is simulated; however, due to the weaker emissions in the mid-latitudes the effects are substantially smaller compared to the tropics.~~

To analyse the impact of the $\mathrm{LNO_x}$ emissions on the aerosol water uptake, the mean growth factor (GF = ambient diameter / dry diameter) is compared. Using this parameter has the advantage ~~, that in contrast to~~ that, e.g. in contrast to the aerosol water content~~–~~, it is independent of the aerosol size and particle number concentrations. The overall impact of lightning on the GF is relatively small: for present day conditions the nucleation and aitken mode depict a slight increase in the upper troposphere (up to 2%), whereas in the accumulation and coarse mode a decrease of up to −2% is simulated caused by LNO$_x$ emissions. Due to the relatively dry conditions in the upper troposphere, where the nitrate changes are strongest, the effect on water uptake is expected to be that small. Closer to the surface the differences in the growth factor are even smaller and ambiguous in their sign, lacking statistical significance.

~~For preindustrial conditions the results are similar in their distribution with a slightly increased amplitude (up to ±5%). The sensitivity simulations are characterised by the same distribution and dependencies on the emissions by lightning. Corresponding figures can be found in the supplement.~~

3.4 Impacts on climate

3.4.1 Aerosol optical properties

As aerosol particles scatter and absorb solar and infrared radiation, the impact of the LNO$_x$ emissions is investigated with the help of Fig. 5 for the ARG present day simulation. The ~~floor panel depicts the annual mean aerosol optical depth (AOD)~~ left panel shows both the mean column AOD at 550 nm wavelength ~~with maximum values in the dust emission regions (e.g. Northern Africa) and the anthropogenic pollution centers (e.g. Eastern China~~(on the floor level) as well as the zonal mean extinction (rear panel). The ~~climatological global mean value with present day emissions is 0.122, which is close to the observations as derived from MODIS (e.g., Mao et al., 2014) . The influence of the emissions can be analysed with the help of the ceiling map in Fig. 5, which shows the percentage fractions of the~~ ceiling panel depicts statistical significant changes in the column AOD and the front panel visualises statistical significant changes in the ~~simulated AOD. Globally a slight reduction to 0.121 can be found (∼ −1%). However, in some regions even higher extinction is simulated in case of no nitrate formation from lightning, which is in contrast to the findings of Sect. 3.2.1, where no regions with enhanced nitrate have been simulated. These regions are mostly oceanic or remote with little direct impact by either lightning and primary emissions.~~ extinction. Note, that for determining the zonal mean of statistical significant changes, only the individual data points which exhibit significance are selected and the average over these (selected) points is taken.

The rear panel depicts the zonal mean of the average extinction per km~~is provided in the rear panel of Fig. 5.~~. Substantial extinction is simulated in the lower troposphere (below 700 hPa) with an additional enhancement of the optical depth between 20° and 50°N. This is a consequence of both the distribution of water vapour and hence aerosol water supporting aerosol growth and extinction in the lower troposphere of the tropics, on the other hand driven by natural sources of dust (and associated extinction) as well as anthropogenic pollution (especially from East Asia). The relative changes of aerosol extinction due to LNO$_x$ are depicted in the front panel. The most substantial enhancement of aerosol extinction is simulated in the middle to upper troposphere in the tropics with zonal mean enhancements higher than ~~10%~~20%. In the uppermost troposphere ~~also~~ reductions in extinction are also simulated as a consequence of neglecting the lightning emissions. ~~Please note, that the stratospheric influences are masked in the zonal mean views as the stratospheric circulation and processes are not well resolved with this model configuration.~~ Compared to the nitrate enhancements as analysed from Fig. 3, the maximum enhancement of aerosol extinction by $\mathrm{LNO_x}$ is located further downwards in the middle troposphere, i.e. between 400 and 600 hPa, whereas the strongest nitrate enhancement has been simulated between 200 and 400 hPa.

The ~~isosurfaces depicting the regions of enhancements and reductions of extinction in~~ column AOD (floor panel) shows maximum values in the dust emission regions (e.g. Northern Africa) and the anthropogenic pollution centers (e.g. Eastern China). The climatological global mean value with present day emissions is 0.122, which is close to the observations as derived from MODIS (e.g., Mao et al., 2014) . The influence of the $\mathrm{LNO_x}$ emissions can be analysed with the help of the ceiling map in Fig. 5, which shows the percentage fractions of the changes in the simulated AOD. Globally a slight reduction to 0.121 can be found ($\sim$ 1%). However, in some regions even higher extinction is simulated in case of no nitrate formation from lightning, which is in contrast to the findings of Sect. 3.2.1, where no regions with enhanced nitrate have been simulated. However, only in very few regions a statistical significant signal in the column AOD can be found. This is a consequence of the fact that significant changes in extinction are mostly located in the upper troposphere which contributes less to total column AOD (cf. rear panel). The right panel of Fig. 5 ~~provide further information on their locations~~depicts again the absolute values of AOD (floor panel) and extinction (rear panel), but also shows isosurfaces of the location of the regions of enhancement and reduction of extinction. The pale red depicts the $+10\%$ isosurface, whereas the embedded dark red regions mark an increase of the extinction due to $\mathrm{LNO_x}$ of more than $+20\%$. On the other hand the pale blue marks regions of a medium reduction (-10%) and dark blue regions (only at the tropopause) depict extinction reductions of more than 20%. The main enhancement of extinction by ~~lighting~~ lightning emissions occur between 400 and 600 hPa over the tropical continents and slightly further downwind. This general pattern corresponds to the enhanced nitrate mixing ratios (c.f. Fig. 3), but is more restricted to the continents. In the extra-tropics an even higher extinction is simulated if no $\mathrm{LNO_x}$ emissions are considered. This is a consequence of the secondary effects caused by the emissions, i.e. the influence on sulphur oxidation via the oxidation capacity of the atmosphere and changes in the size distribution. Comparing the pattern correlation between the changes in nitrate and extinction a relatively low value is found ($R \approx 0.3$). However, the changes in sulphate and extinction show a higher correlation value of $R \approx 0.4$.

~~For preindustrial conditions the situation is quite comparable (see supplement), despite the intensive AOD signals in the regions with anthropogenic pollution, especially East Asia. The simulated global mean column AOD at 550 nm is 0.090. Consequently, the enhancement of the extinction between 20° and 50°N is less strong. The neglect of the lightning emissions results in a reduced column AOD in most regions, resulting in a global mean value of 0.089. A reduction in column AOD~~

~~occurs only in regions of strong hydrophobic emissions (dust or BC) where the aerosol lifetime is reduced by a coating by nitrates and subsequent faster conversion from hydrophobic to hydrophilic categories, e.g. in the Northern part of the Saharan outflow or in Southern Africa. The extension of the $+10\%$ isosurface in the tropical upper troposphere is smaller, especially over Central Africa and the maritime continent. In contrast, over South America the enhancement of aerosol extinction caused by the emission is even stronger. On the other hand, also slightly more reductions in extinction are simulated in the extra-tropics and mid-latitude upper troposphere, but they hardly exceed the -10% level.~~

Analysing the PDF of the differences in AOD between the simulation with and without lightning emissions (~~not shown~~see supplement) reveals that for present day conditions larger deviations occur more often compared to the preindustrial cases. The PDFs are almost symmetric, but show a slight shift towards enhanced extinction in the simulations including LNO_x emissions.

The sensitivity simulations with the alternative warm cloud activation scheme in general show a similar distribution of the aerosol extinction with the enhancement in the middle and upper tropical troposphere (see supplement). The global mean column AOD for present day conditions is slightly lower (0.120) and the reduction in case of no LNO_x emissions even further reduced ($\sim 0.5\%$). Strongest enhancement is found over the tropical continents, i.e. the regions with maximum lightning activity, but most of the nitrate is simulated in the upper troposphere, whereas also in this configuration the extinction is enhanced in the middle to upper troposphere. For preindustrial conditions, the enhancement of the extinction by lightning is a bit stronger, but still small ($\sim 1\%$).

To analyse whether the contribution of scattering versus absorption is influenced by the LNO_x emissions, the single scattering albedo is compared in both model simulations. The zonal mean value changes by less than $\sim +1\%$ in the free troposphere and stratosphere, and is reduced by a maximum of $\sim -4\%$ in the boundary layer when lightning emissions are considered. Consequently, a change in the thermodynamic structure resulting from direct aerosol-radiation interactions is expected to be small.

3.4.2 Cloud properties

As cloud properties can be influenced indirectly by the LNO_x emissions three parameters are analysed to provide the causes for aerosol-cloud-radiation interactions. The cloud cover may hint at modifications of the cloud lifetime effect (Albrecht, 1989), whereas the combination of the liquid (ice) water content and the cloud droplet (ice crystal) numbers, which can also be expressed via the effective radii, can shed light onto the direct influence of the lightning emissions on the radiative properties of the clouds via the Twomey (1977) effect.

- Cloud cover:

  The cloud and precipitation cover changes only to a minor degree between the simulations with and without lightning for the present day scenario (ARG case). In the tropics the LNO_x

emissions lead to a small increase in the mid troposphere cloud coverage of the Northern hemisphere, whereas the dipole pattern is compensated by reduced cloudiness in the Southern hemisphere. Apart from this, only the polar latitudes show some larger (but not statistical significant) changes in cloud coverage. Consequently, the impact of the cloud lifetime effect is expected to be small.

- Liquid / ice water content:
  The patterns for cloud water and cloud ice are characterised by local dipole changes, but there are no significant modifications of the distribution of cloud water and ice, respectively. Only the tropical middle to upper troposphere of the Northern hemisphere shows an enhanced ice mixing ratio in the simulation with lightning emissions, which correlates to the increase in cloud coverage. However, the decrease in cloud coverage in the corresponding region in the Southern hemisphere is not accompanied by a substantial decrease in ice water. In the zonal average the increase in total water is less than 10%.

- Cloud droplet / ice crystal numbers:
  Compared to the other two parameters, some substantial changes in cloud droplet and especially in ice crystal number are found as a consequence of the $\mathrm{LNO_x}$ emissions. For the present day scenario, in the troposphere between 650 and 400 hPa an increase in cloud droplets is simulated in case of active lightning emissions in the tropics. Further North a decrease of cloud droplets between 900 and 600 hPa is calculated. Both are of approximately the same magnitude both in absolute and in relative terms and are a consequence of more hydrophilic particles due to nitrate coating and a reduction of lower tropospheric CCN due to reduction in lifetime and faster conversion from hydrophobic to hydrophilic conditions. However, the statistical significance of the changes is relatively low. For the ice crystal number the influence is even stronger: in the tropical upper troposphere the $\mathrm{LNO_x}$ emissions lead to an increase in the ice crystal number in a region where the highest ice crystal numbers are simulated. In the mid-latitudes a reduction of the ice crystal numbers is simulated in both hemispheres above 400 hPa. Both modifications are of the order of more than $\pm 20\%$. Due to the coarse temporal resolution of the model output it is not possible to attribute the differences to a change in regime from homogeneous to heterogeneous freezing. Nevertheless, it is possible that locally this effect can occur and has a substantial influence on the ice crystal number concentration.

By Analysing the effective radii ~~with the help of~~ (Fig. 6), some of the effects on water mixing ratios and numbers compensate, whereas others positively amplify each other. This is both valid for liquid clouds in the lower and middle troposphere ~~. For these~~ as well as ice clouds in the middle and upper troposphere. For liquid clouds, the effective radii increase in the simulation with lightning in the polewards of 40° below 800 hPa and in the tropics (0° to 20°N) up to 400 hPa. However, most of the changes are not statistically significant, such that Fig. 6 depicts only the changes in ice crystal size; a corresponding figure for the liquid clouds can be found in the supplement. The back panel depicts in colours the statistical significant zonal mean change in effective ice crystal size (in absolute units of μm). The white contours represent absolute ice crystal sizes with maximum values in the upper part of the mid-latitude storm tracks and the upper tropical troposphere. Note, that the absolute values (not marked) are of secondary importance as mean values over cloudy and non-cloudy conditions are taken), but the difference is robust. For ice clouds the effective radii also increase in the same tropical latitude range between 600 and 200 hPa. Additionally, a decrease in effective radii of similar magnitude on the other side of the ITCZ is simulated. Also in the high latitudes the effective ice crystal size mostly increases in the simulation with $\mathrm{LNO_x}$ emissions, both in the upper troposphere as well as in the boundary layer. The shape of the regions of ice crystal changes extends vertically over more than 100 hPa and is not located in the uppermost troposphere only. This indicates that the statistical significant changes are more associated to the process of immersion freezing in mixed phase clouds than to the cirrus regime with competition between homogeneous and heterogeneous freezing.

~~For preindustrial conditions the response to the emissions is comparable, but shows a slightly higher amplitude.~~ Using the KK model configuration, which applies a different cloud droplet activation scheme, results in a lower sensitivity of the liquid droplet effective radius at elevated altitude for present day conditions. However, in the lower troposphere a similar strong signal is simulated, which leads to an increase of effective droplet radius in the tropical Southern hemisphere and a decrease in the Northern counterpart. For ice crystals, mostly a reduction of effective crystal size is simulated which is a consequence of the liquid droplet freezing. The effects for preindustrial conditions are similar, with even lower sensitivity of the low clouds on the lightning emissions.

In general, the regions with positive and negative effects in the effective droplet and crystal size show ambiguous signals, such that the impact on cloud radiative properties can only be addressed in conjunction with cloud coverage and the calculation of the radiative fluxes.

3.4.3 Radiative Fluxes

The combined effects of modifications in the aerosol extinction and the cloud optical properties lead to a modification of the shortwave and longwave radiation fluxes. In Fig. 7 the mean impact on the total sky shortwave flux is depicted in the upper panel. Shown are the differences in the SW flux at the top of the atmosphere between a simulation with and one without $\mathrm{LNO_x}$ emissions (ARG configuration). Despite the relatively large internal variability of the system, several regions show a statistical significant change in the TOA short wave flux (based on a t-test with 90% significance threshold on the annual mean data), which are marked by the hatched regions. However, the regions with increased or decreased fluxes are not directly co-located with the $\mathrm{LNO_x}$ emissions. Downwind of Central Africa an increased TOA short wave flux (corresponding to a heating) is simulated, whereas in Northern Amazonia and the maritime continent a decrease of the fluxes (corresponding to a cooling) is calculated. Also in the mid-latitude storm tracks mostly a slight increase in the fluxes is simulated, whereas over the tropical oceans the model suggests decreased fluxes. In the global mean the overall effect is determined as an enhanced backward reflection of the shortwave radiation of ~ -0.1 W/m^2 for all sky conditions. However, the time series of the global mean differences (depicted in the lower panel of Fig. 7) is characterised by large internal variability with both positive and negative differences. The clear sky fluxes show a substantially weaker signal with only ~ -0.05 W/m^2 and hardly any regions with statistical significance. Furthermore, the patterns of substantial changes are clearly separated from the regions where lightning is dominant (i.e. mid-latitudes versus tropical continents). Consequently, the clear sky signal is interpreted as statistical noise.

~~For preindustrial conditions (for a graph see supplement) the situation is comparable with a slightly increased amplitude of ~ -0.14 W/m^2 and a similar pattern distribution. The clear sky signal is almost negligible with ~ -0.01 W/m^2 and no statistical significance.~~

Considering both the temporal variability of the mean and the large spatial variability, the uncertainty in the total effect has to be considered as relatively large. Using the sensitivity simulations with the alternative cloud activation scheme reveals for both present day and preindustrial conditions a shortwave flux perturbation of ~ -0.06 W/m^2 again with no significant contribution from the clear sky fluxes.

To bring these numbers into context with the total shortwave flux disturbances from anthropogenic aerosols, simulation results with both cloud activation schemes and present day and preindustrial emission scenarios are intercompared (for figures see supplement). Both model configurations show a statistical significant cooling over the regions of dominant anthropogenic aerosol pollution (Eastern US, Europe, China) for both clear sky and all sky conditions. The clear sky disturbance is ~ -0.57 for the ARG and ~ -0.50 W/m^2 for the KK configurations. All sky fluxes change by ~ -1.62 for the ARG and ~ -1.42 W/m^2 for the KK simulation setups, respectively. The overall anthropogenic aerosol effect is relatively large in the simulations compared to the results published in the latest IPCC report (IPCC, 2013) and therefore an overestimated sensitivity to the aerosol disturbance by lightning $\mathrm{NO_x}$ nitrate cannot be ruled out completely.

The effect on the longwave radiation via both aerosol extinction and cloud effects is relatively small. Especially, the clear sky flux disturbances are almost negligible, such that the total effect is dominated by ACI.

Consequently, as the differences between both cloud activation schemes indicate an uncertainty of almost 50% the quantitative estimate of -100 mW/m^2 should also be used with a comparable uncertainty range of 50 mW/m^2.

4 Conclusions

The overall impact of chemically produced aerosol nitrate from lightning NO_x emissions is analysed with a global chemistry climate model. Based on a total emission of 6 Tg N/yr, the contribution of LNO_x to the concentrations of upper tropospheric oxidised nitrogen by more than 57% for present day and 75% for preindustrial conditions. Given a sufficient amount of neutralising cations, especially NH_3, substantial amounts of aerosol nitrate are formed with the increases in the upper tropospheric burden exceeding 30% for present day and 50% for preindustrial conditions. Therefore, lightning is also a major contributor to the aerosol nitrate burden in the upper troposphere. The concentration enhancements are not uniformly distributed, but follow the regions of maximum emissions, and further downwind transport.

Besides the well known impacts of lightning on O_3, ~~also~~ the impact on the chemical oxidation capacity of the atmosphere is highly important for the upper tropospheric aerosol ~~load~~loading. Because of a decrease of the oxidation potential (as represented by an increase in the methane lifetime), the sulphate formation via the gas phase reaction pathway is reduced in case of neglecting LNO_x emissions. Hence new particle formation is suppressed, efficiently modifying the small part of the aerosol size distribution. In addition to the condensation of nitrate on the aerosol particles, the combination of a reduced particle number and the coagulation of the particles, substantial effects on the particle size distribution in the upper troposphere are simulated.

The changes in aerosol size, chemical composition and number concentrations have implications on climate via both aerosol-radiation and aerosol-cloud interactions. Lightning and its consequences (nitrate formation, size distribution changes) causes mostly an increase in the total aerosol extinction with pronounced increase maxima in the upper troposphere. On the other hand, both liquid and ice phase cloud optical properties are modified by the LNO_x emissions, represented by e.g. effective droplet and ice crystal size. However, in contrast to the nitrate concentration increases the cloud effects resulting from lightning are ambiguous.

The resulting shortwave flux perturbations caused by LNO_x emissions are quantified to be ~ -100 mW/m^2. The resulting effect is caused mostly by the aerosol-cloud interactions, whereas the direct aerosol-radiation interactions are of secondary importance. However, an uncertainty range of almost ~ 50 mW/m^2 has to be assumed due to large internal variability and uncertainties in the process description (mostly in the aerosol-cloud interactions).

Nevertheless, an increase in lightning activity in the future or impacts of a more efficient transformation of the LNO_x emissions into nitrate under future climate conditions might have a non-negligible impact on the radiation balance of the atmosphere. On the other hand, aerosol nitrate formed from lightning offers further possibilities to address the feedback and (potentially compensating) impacts of combined chemistry-aerosol-climate interactions.

Appendix A: Preindustrial conditions

The results from the sensitivity simulations for preindustrial conditions are summarised in this section.

- Emissions:
  In the preindustrial scenario total LNO_x emissions of 6.12 ± 0.03 Tg N/yr are simulated for the ARG scenario and 6.16 ± 0.03 Tg N/yr for the KK simulations.

- Nitrate enhancement:
  During preindustrial times, an increase in the HNO_3 burden with $\sim 67\%$ and $\sim 76\%$ in the total and upper troposphere is even stronger compared to present day conditions. This stronger enhancement is due to the larger contribution of the LNO_x emissions compared to the total NO_x release, which is mostly from anthropogenic sources in present day conditions. Enhancements for N_2O_5 are of $\sim 80\%$, but also under these conditions N_2O_5 is a minor contributor to the total highly oxidised nitrogen tropospheric load (less than 5%). A similar figure as Fig. 3 for preindustrial conditions can be found in the supplement. Due to the lower other sources of NO_x in the atmosphere, the importance of the LNO_X emissions is substantially increased. Therefore, the relative differences in NO_3^- are larger which can be seen from the extended areas included by the respective isosurfaces and the larger areas covered by the turquoise contour lines at the floor panel of the figure. The lower overall nitrate mixing ratios are obvious from the colour scale of the column nitrate which is almost an order of magnitude lower compared to present day conditions. A lower contribution of particulate nitrate to the total N(V) load with 22% for the whole and 9% for the upper troposphere is simulated, which can be mostly attributed to lower NH_3 emissions and consequently less NH_4^+ ions to neutralise and thermodynamically stabilise available aerosol nitrate. However, the enhancement of the particulate phase due to LNO_x is even stronger compared to present day conditions to $\sim$ 34% for the whole troposphere and 17% for the UT.

- Methane lifetime and sulphate:
  The lifetime increase is even stronger without anthropogenic emissions, such that for the upper troposphere almost a doubling of the CH_4 lifetime occurs (see lower part of Tab. 2). Nevertheless, this results in a similar reduction of the sulphate burden compared to present day without LNO_x emissions despite the substantially lower total SO_2 emissions.

- Size distributions:
  For preindustrial conditions (see supplement) the simulation provides similar results from the tropics. However, in the mid-latitudes where a substantial reduction of other NO_x emissions is applied compared to the present day scenario the impact of lightning on the nitrogen budget as well as the size distribution becomes more important. Obviously, a similar pattern as for

the tropical continents is simulated; however, due to the weaker $\mathrm{LNO_x}$ emissions in the midlatitudes the effects are substantially smaller compared to the tropics.

- Growth factors:

  The results for the GF are similar in their distribution compared to the present day scenario with a slightly increased amplitude (up to $\pm 5\%$). The sensitivity simulations *KK* are characterised by the same distribution and dependencies on the emissions by lightning. Corresponding figures can be found in the supplement.

- Extinction and AOD:

  For preindustrial conditions the situation is quite comparable (see supplement), despite the intensive AOD signals in the regions with anthropogenic pollution, especially East Asia. The simulated global mean column AOD at 550 nm is 0.090. Consequently, the enhancement of the extinction between 20° and 50°N is less strong. The neglect of the lightning emissions results in a reduced column AOD in most regions, resulting in a global mean value of 0.089. A reduction in column AOD occurs only in regions of strong hydrophobic emissions (dust or BC) where the aerosol lifetime is reduced by a coating by nitrates and subsequent faster conversion from hydrophobic to hydrophilic categories. However, also in this case the statistical significance is relatively small. With respect to the zonal mean extinction the changes are slightly larger compared to present day conditions.

- Cloud response:

  For preindustrial conditions the response to the $\mathrm{LNO_x}$ emissions is comparable, but shows a slightly higher amplitude.

- Radiative fluxes:

  For preindustrial conditions (for a graph see supplement) the situation is comparable with a slightly increased amplitude of ~ -0.14 W/m^2 and a similar pattern distribution. The clear sky signal is almost negligible with ~ -0.01 W/m^2 and no statistical significance.

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References

Abdul-Razzak, H. and Ghan, S.: A parameterization of aerosol activation: 2. Multiple aerosol types, Journal of Geophysical Research: Atmospheres, 105, 6837–6844, 2000.

Adams, P. J., Seinfeld, J. H., Koch, D., Mickley, L., and Jacob, D.: General circulation model assessment of direct radiative forcing by the sulfate-nitrate-ammonium-water inorganic aerosol system, Journal of Geophysical Research: Atmospheres, 106, 1097–1111, 2001.

Albrecht, B. A.: Aerosols, Cloud Microphysics, and Fractional Cloudiness, Science, 245, 1227–1230, 1989.

Allen, D. J., Pickering, K. E., Pinder, R. W., Henderson, B. H., Appel, K. W., and Prados, A.: Impact of lightning-NO on eastern United States photochemistry during the summer of 2006 as determined using the CMAQ model, Atmospheric Chemistry and Physics, 12, 1737–1758, doi:10.5194/acp-12-1737-2012, 2012.

Banerjee, A., Archibald, A. T., Maycock, A. C., Telford, P., Abraham, N. L., Yang, X., Braesicke, P., and Pyle, J. A.: Lightning $\mathrm{NO_x}$, a key chemistry–climate interaction: impacts of future climate change and consequences for tropospheric oxidising capacity, Atmospheric Chemistry and Physics, 14, 9871–9881, doi:10.5194/acp-14-9871-2014, 2014.

Bauer, S. E., Koch, D., Unger, N., Metzger, S. M., Shindell, D. T., and Streets, D. G.: Nitrate aerosols today and in 2030: a global simulation including aerosols and tropospheric ozone, Atmospheric Chemistry and Physics, 7, 5043–5059, 2007.

Bellouin, N., Rae, J., Jones, A., Johnson, C., Haywood, J., and Boucher, O.: Aerosol forcing in the Climate Model Intercomparison Project (CMIP5) simulations by HadGEM2-ES and the role of ammonium nitrate, Journal of Geophysical Research: Atmospheres, 116, n/a–n/a, 2011.

Chang, D. Y., Tost, H., Steil, B., and Lelieveld, J.: Aerosol–cloud interactions studied with the chemistry-climate model EMAC, Atmospheric Chemistry and Physics Discussions, 14, 21 975–22 043, 2014.

Christian, H. J., Blakeslee, R. J., Boccippio, D. J., Boeck, W. L., Buechler, D. E., Driscoll, K. T., Goodman, S. J., Hall, J. M., Koshak, W. J., Mach, D. M., and Stewart, M. F.: Global frequency and distribution of lightning as observed from space by the Optical Transient Detector, J. Geophys. Res., 108, 4005, doi:10.1029/2002JD002347, 2003.

de Meij, A., Pozzer, A., Pringle, K. J., Tost, H., and Lelieveld, J.: EMAC model evaluation and analysis of atmospheric aerosol properties and distribution with a focus on the Mediterranean region, Atmospheric Research, 114–115, 38 – 69, doi:10.1016/j.atmosres.2012.05.014, 2012.

Dentener, F., Kinne, S., Bond, T., Boucher, O., Cofala, J., Generoso, S., Ginoux, P., Gong, S., Hoelzemann, J. J., Ito, A., Marelli, L., Penner, J. E., Putaud, J.-P., Textor, C., Schulz, M., van der Werf, G. R., and Wilson, J.: Emissions of primary aerosol and precursor gases in the years 2000 and 1750 prescribed data-sets for AeroCom, Atmospheric Chemistry and Physics, 6, 4321–4344, doi:10.5194/acp-6-4321-2006, 2006.

Dietmüller, S., Jöckel, P., Tost, H., Kunze, M., Gellhorn, C., Brinkop, S., Frömming, C., Ponater, M., Steil, B., Lauer, A., and Hendricks, J.: A new radiation infrastructure for the Modular Earth Submodel System (MESSy, based on version 2.51), Geoscientific Model Development Discussions, 2016, 1–21, doi:10.5194/gmd-2015-277, 2016.

Finney, D. L., Doherty, R. M., Wild, O., and Abraham, N. L.: The impact of lightning on tropospheric ozone chemistry using a new global lightning parametrisation, Atmospheric Chemistry and Physics, 16, 7507–7522, doi:10.5194/acp-16-7507-2016, 2016a.

Finney, D. L., Doherty, R. M., Wild, O., Young, P. J., and Butler, A.: Response of lightning NOx emissions and ozone production to climate change: Insights from the Atmospheric Chemistry and Climate Model Intercomparison Project, Geophysical Research Letters, 43, 5492–5500, doi:10.1002/2016GL068825, 2016b.

Fountoukis, C. and Nenes, A.: ISORROPIA II: a computationally efficient thermodynamic equilibrium model for K^+, Ca^{2+}, Mg^{2+}, $NH_4{}^+$, Na^+, $SO_4{}^{2-}$, $NO_3{}^-$, Cl^-, H_2O aerosols, Atmospheric Chemistry and Physics, 7, 4639–4659, doi:10.5194/acp-7-4639-2007, 2007.

Gressent, A., Sauvage, B., Cariolle, D., Evans, M., Leriche, M., Mari, C., and Thouret, V.: Modeling lightning-NO_x chemistry on a sub-grid scale in a global chemical transport model, Atmospheric Chemistry and Physics, 16, 5867–5889, doi:10.5194/acp-16-5867-2016, 2016.

Grewe, V.: Impact of climate variability on tropospheric ozone, Science of The Total Environment, 374, 167–181, doi:http://dx.doi.org/10.1016/j.scitotenv.2007.01.032, 2007.

Höpfner, M., Volkamer, R., Grabowski, U., Grutter, M., Orphal, J., Stiller, G., von Clarmann, T., and Wetzel, G.: First detection of ammonia (NH_3) in the Asian summer monsoon upper troposphere, Atmospheric Chemistry and Physics, 16, 14 357–14 369, doi:10.5194/acp-16-14357-2016, 2016.

Huntrieser, H., Schlager, H., Roiger, A., Lichtenstern, M., Schumann, U., Kurz, C., Brunner, D., Schwierz, C., Richter, A., and Stohl, A.: Lightning-produced NO_x over Brazil during TROCCINOX: airborne measurements in tropical and subtropical thunderstorms and the importance of mesoscale convective systems, Atmospheric Chemistry and Physics, 7, 2987–3013, doi:10.5194/acp-7-2987-2007, 2007.

IPCC: Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change, Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA, doi:10.1017/CBO9781107415324, www.climatechange2013.org, 2013.

Jacobson, M. Z.: Global direct radiative forcing due to multicomponent anthropogenic and natural aerosols, Journal of Geophysical Research: Atmospheres, 106, 1551–1568, 2001.

Jaegle, L., Steinberger, L., Martin, R. V., and Chance, K.: Global partitioning of NOx sources using satellite observations: Relative roles of fossil fuel combustion, biomass burning and soil emissions, Faraday Discuss., 130, 407–423, 2005.

Jöckel, P., Sander, R., Kerkweg, A., Tost, H., and Lelieveld, J.: Technical Note: The Modular Earth Submodel System (MESSy) - a new approach towards Earth System Modeling, Atmos. Chem. Phys., 5, 433–444, 2005.

Jöckel, P., Kerkweg, A., Pozzer, A., Sander, R., Tost, H., Riede, H., Baumgaertner, A., Gromov, S., and Kern, B.: Development cycle 2 of the Modular Earth Submodel System (MESSy2), Geoscientific Model Development, 3, 717–752, doi:10.5194/gmd-3-717-2010, 2010.

Jöckel, P., Tost, H., Pozzer, A., Kunze, M., Kirner, O., Brenninkmeijer, C. A. M., Brinkop, S., Cai, D. S., Dyroff, C., Eckstein, J., Frank, F., Garny, H., Gottschaldt, K.-D., Graf, P., Grewe, V., Kerkweg, A., Kern, B., Matthes, S., Mertens, M., Meul, S., Neumaier, M., Nützel, M., Oberländer-Hayn, S., Ruhnke, R., Runde, T., Sander, R., Scharffe, D., and Zahn, A.: Earth System Chemistry integrated Modelling (ESCiMo) with the Modular Earth Submodel System (MESSy) version 2.51, Geoscientific Model Development, 9, 1153–1200, doi:10.5194/gmd-9-1153-2016, 2016.

Kärcher, B., Hendricks, J., and Lohmann, U.: Physically based parameterization of cirrus cloud formation for use in global atmospheric models, J. Geophys. Res., 111, D01 206, doi:10.1029/2005JD006219, 2006.

Kerkweg, A., Buchholz, J., Ganzeveld, L., Pozzer, A., Tost, H., and Jöckel, P.: Technical Note: An implementation of the dry removal processes DRY DEPosition and SEDImentation in the Modular Earth Submodel System (MESSy), Atmos. Chem. Phys., 6, 4617–4632, 2006a.

Kerkweg, A., Sander, R., Tost, H., and Jöckel, P.: Technical Note: Implementation of prescribed (OFFLEM), calculated (ONLEM), and pseudo-emissions (TNUDGE) of chemical species in the Modular Earth Submodel System (MESSy), Atmos. Chem. Phys., 6, 3603–3609, 2006b.

Labrador, L. J., v. Kuhlmann, R., and Lawrence, M. G.: Strong sensitivity of the global mean OH concentration and the tropospheric oxidizing efficiency to the source of $\mathrm{NO_x}$ from lightning, Geophys. Res. Lett., 31, L06 102, doi:10.1029/2003GL019229, 2004.

Labrador, L. J., v. Kuhlmann, R., and Lawrence, M. G.: The effects of lightning-produced $\mathrm{NO_x}$ and its vertical distribution on atmospheric chemistry: sensitivity simulations with MATCH-MPIC, Atmos. Chem. Phys., 5, 1815–1834, 2005.

Lamarque, J.-F., Bond, T. C., Eyring, V., Granier, C., Heil, A., Klimont, Z., Lee, D., Liousse, C., Mieville, A., Owen, B., Schultz, M. G., Shindell, D., Smith, S. J., Stehfest, E., Van Aardenne, J., Cooper, O. R., Kainuma, M., Mahowald, N., McConnell, J. R., Naik, V., Riahi, K., and van Vuuren, D. P.: Historical (1850–2000) gridded anthropogenic and biomass burning emissions of reactive gases and aerosols: methodology and application, Atmospheric Chemistry and Physics, 10, 7017–7039, doi:10.5194/acp-10-7017-2010, 2010.

Lamarque, J.-F., Shindell, D. T., Josse, B., Young, P. J., Cionni, I., Eyring, V., Bergmann, D., Cameron-Smith, P., Collins, W. J., Doherty, R., Dalsoren, S., Faluvegi, G., Folberth, G., Ghan, S. J., Horowitz, L. W., Lee, Y. H., MacKenzie, I. A., Nagashima, T., Naik, V., Plummer, D., Righi, M., Rumbold, S. T., Schulz, M., Skeie, R. B., Stevenson, D. S., Strode, S., Sudo, K., Szopa, S., Voulgarakis, A., and Zeng, G.: The Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP): overview and description of models, simulations and climate diagnostics, Geoscientific Model Development, 6, 179–206, doi:10.5194/gmd-6-179-2013, 2013.

Lohmann, U. and Hoose, C.: Sensitivity studies of different aerosol indirect effects in mixed-phase clouds, Atmospheric Chemistry and Physics, 9, 8917–8934, 2009.

Lohmann, U., Rotstayn, L., Storelvmo, T., Jones, A., Menon, S., Quaas, J., Ekman, A. M. L., Koch, D., and Ruedy, R.: Total aerosol effect: radiative forcing or radiative flux perturbation?, Atmospheric Chemistry and Physics, 10, 3235–3246, 2010.

Makkonen, R., Romakkaniemi, S., Kokkola, H., Stier, P., Räisänen, P., Rast, S., Feichter, J., Kulmala, M., and Laaksonen, A.: Brightening of the global cloud field by nitric acid and the associated radiative forcing, Atmospheric Chemistry and Physics, 12, 7625–7633, 2012.

Mao, K., Ma, Y., Xia, L., Chen, W. Y., Shen, X., He, T., and Xu, T.: Global aerosol change in the last decade: An analysis based on {MODIS} data, Atmospheric Environment, 94, 680 – 686, 2014.

Martin, R. V., Jacob, D. J., Logan, J. A., Bey, I., Yantosca, R. M., Staudt, A. C., Li, Q., Fiore, A. M., Duncan, B. N., Liu, H., Ginoux, P., and Thouret, V.: Interpretation of TOMS observations of tropical tropospheric ozone with a global model and in situ observations, Journal of Geophysical Research: Atmospheres, 107, ACH 4–1–ACH 4–27, doi:10.1029/2001JD001480, 4351, 2002.

Mayer, B. and Kylling, A.: Technical note: The libRadtran software package for radiative transfer calculations - description and examples of use, Atmospheric Chemistry and Physics, 5, 1855–1877, 2005.

Ott, L. E., Pickering, K. E., Stenchikov, G. L., Allen, D. J., DeCaria, A. J., Ridley, B., Lin, R.-F., Lang, S., and Tao, W.-K.: Production of lightning NOx and its vertical distribution calculated from three-dimensional cloud-scale chemical transport model simulations, Journal of Geophysical Research: Atmospheres, 115, doi:10.1029/2009JD011880, 2010.

Petters, M. D. and Kreidenweis, S. M.: A single parameter representation of hygroscopic growth and cloud condensation nucleus activity, Atmospheric Chemistry and Physics, 7, 1961–1971, doi:10.5194/acp-7-1961-2007, 2007.

Pickering, K. E., Wang, Y., Tao, W.-K., Price, C., and Müller, J.-F.: Vertical distribution of lightning $\mathrm{NO_x}$ for use in regional and chemical transport models, J. Geophys. Res., 103, 31 203–31 216, 1998.

Pozzer, A., de Meij, A., Pringle, K. J., Tost, H., Doering, U. M., van Aardenne, J., and Lelieveld, J.: Distributions and regional budgets of aerosols and their precursors simulated with the EMAC chemistry-climate model, Atmospheric Chemistry and Physics, 12, 961–987, doi:10.5194/acp-12-961-2012, 2012.

Price, C. and Rind, D.: A simple Lightning Parametrization for Calculating Global Lightning Distributions, J. Geophys. Res., 97, 9919–9933, 1992.

Pringle, K. J., Tost, H., Metzger, S., Steil, B., Giannadaki, D., Nenes, A., Fountoukis, C., Stier, P., Vignati, E., and Lelieveld, J.: Description and evaluation of GMXe: a new aerosol submodel for global simulations (v1), Geoscientific Model Development, 3, 391–412, doi:10.5194/gmd-3-391-2010, 2010.

Roeckner, E., Bäuml, G., Bonaventura, L., Brokopf, R., Esch, M., Giorgetta, M., Hagemann, S., Kirchner, I., Kornblue, L., Manzini, E., Rhodin, A., Schleese, U., Schulzweida, U., and Tompkins, A.: The atmospheric general circulation model ECHAM5: Part 1, Tech. Rep. 349, Max-Planck-Institut für Meteorologie, 2003.

Roeckner, E., Brokopf, R., Esch, M., Giogetta, M., Hagemann, S., Kornblueh, L., Manzini, E., Schleese, U., and Schulzweida, U.: Sensitivity of simulated climate to horizontal and vertical resolution in the ECHAM5 atmosphere model, J. Clim., 19, 3771–3791, 2006.

Sander, R., Baumgaertner, A., Gromov, S., Harder, H., Jöckel, P., Kerkweg, A., Kubistin, D., Regelin, E., Riede, H., Sandu, A., Taraborrelli, D., Tost, H., and Xie, Z.-Q.: The atmospheric chemistry box model CAABA/MECCA-3.0, Geoscientific Model Development, 4, 373–380, doi:10.5194/gmd-4-373-2011, 2011.

Schumann, U. and Huntrieser, H.: The global lightning-induced nitrogen oxides source, Atmos. Chem. Phys., 7, 3823–3907, doi:10.5194/acp-7-3823-2007, 2007.

Steinkamp, J. and Lawrence, M. G.: Improvement and evaluation of simulated global biogenic soil NO emissions in an AC-GCM, Atmospheric Chemistry and Physics, 11, 6063–6082, 2011.

Stelson, A., Friedlander, S., and Seinfeld, J.: A note on the equilibrium relationship between ammonia and nitric acid and particulate ammonium nitrate, Atmospheric Environment (1967), 13, 369 – 371, 1979.

Tanre, D., Geleyn, J.-F., and Slingo, J. M.: Aerosols and Their Climatic Effects, chap. First results of the introduction of an advanced aerosol-radiation interaction in the ecmwf low resolution global model, A. Deepak, Hampton, Va., 1984.

Tiedtke, M.: A Comprehensive Mass Flux Scheme for Cumulus Parametrization in Large-Scale Models, Mon. Weather Rev., 117, 1779–1800, 1989.

Tost, H. and Pringle, K. J.: Improvements of organic aerosol representations and their effects in large-scale atmospheric models, Atmospheric Chemistry and Physics, 12, 8687–8709, doi:10.5194/acp-12-8687-2012, 2012.

Tost, H., Jöckel, P., Kerkweg, A., Sander, R., and Lelieveld, J.: Technical Note: A new comprehensive SCAVenging submodel for global atmospheric chemistry modelling, Atmos. Chem. Phys., 6, 565–574, 2006.

Tost, H., Jöckel, P., and Lelieveld, J.: Lightning and convection parameterisations - uncertainties in global modelling, Atmos. Chem. Phys., 7, 4553–4568, 2007.

Tost, H., Lawrence, M. G., Brühl, C., Jöckel, P., GABRIEL-Team, and SCOUT-O3-DARWIN/ACTIVE-Team: Uncertainties in atmospheric chemistry modelling due to convection parameterisations and subsequent scavenging, Atmospheric Chemistry and Physics, 10, 1931–1951, doi:10.5194/acp-10-1931-2010, 2010.

Twomey, S.: The Influence of Pollution on the Shortwave Albedo of Clouds, Journal of the Atmospheric Sciences, 34, 1149–1152, doi:10.1175/1520-0469(1977)034<1149:TIOPOT>2.0.CO;2, 1977.

van der Werf, G. R., Randerson, J. T., Giglio, L., Collatz, G. J., Mu, M., Kasibhatla, P. S., Morton, D. C., DeFries, R. S., Jin, Y., and van Leeuwen, T. T.: Global fire emissions and the contribution of deforestation, savanna, forest, agricultural, and peat fires (1997–2009), Atmospheric Chemistry and Physics, 10, 11 707–11 735, doi:10.5194/acp-10-11707-2010, 2010.

Xu, L. and Penner, J. E.: Global simulations of nitrate and ammonium aerosols and their radiative effects, Atmospheric Chemistry and Physics, 12, 9479–9504, 2012.

Zare, A., Christensen, J. H., Gross, A., Irannejad, P., Glasius, M., and Brandt, J.: Quantifying the contributions of natural emissions to ozone and total fine PM concentrations in the Northern Hemisphere, Atmospheric Chemistry and Physics, 14, 2735–2756, doi:10.5194/acp-14-2735-2014, 2014.

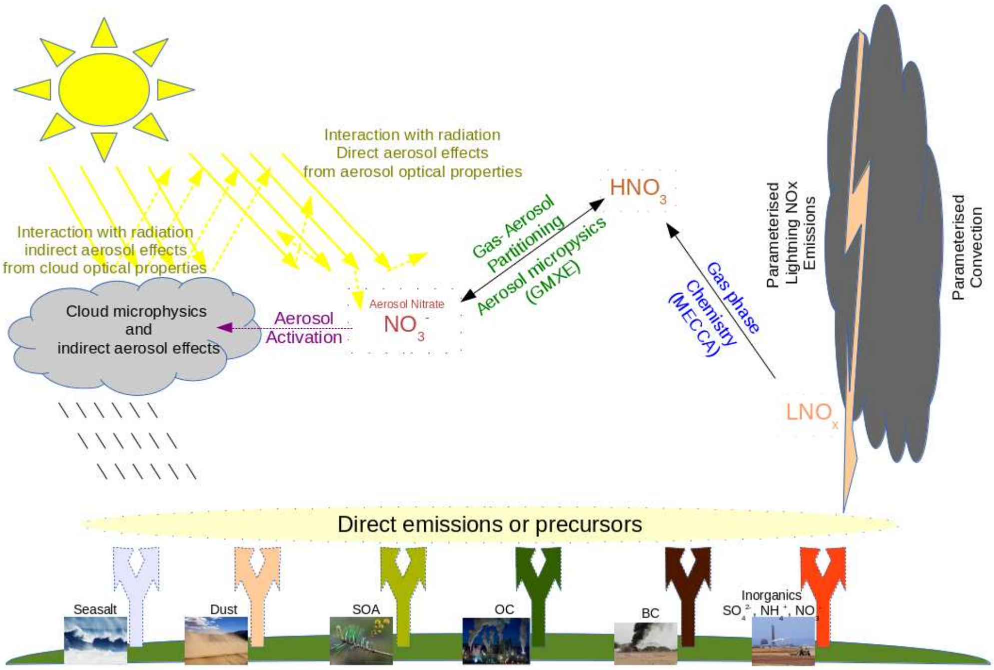

Figure 1. Sketch of the simulated processes from the emission of $\mathrm{NO_x}$ molecules by lightning over gas phase conversion to $\mathrm{HNO_3}$, gas aerosol partitioning to particulate $\mathrm{NO_3^-}$ and implications for the direct and indirect aerosol effects.

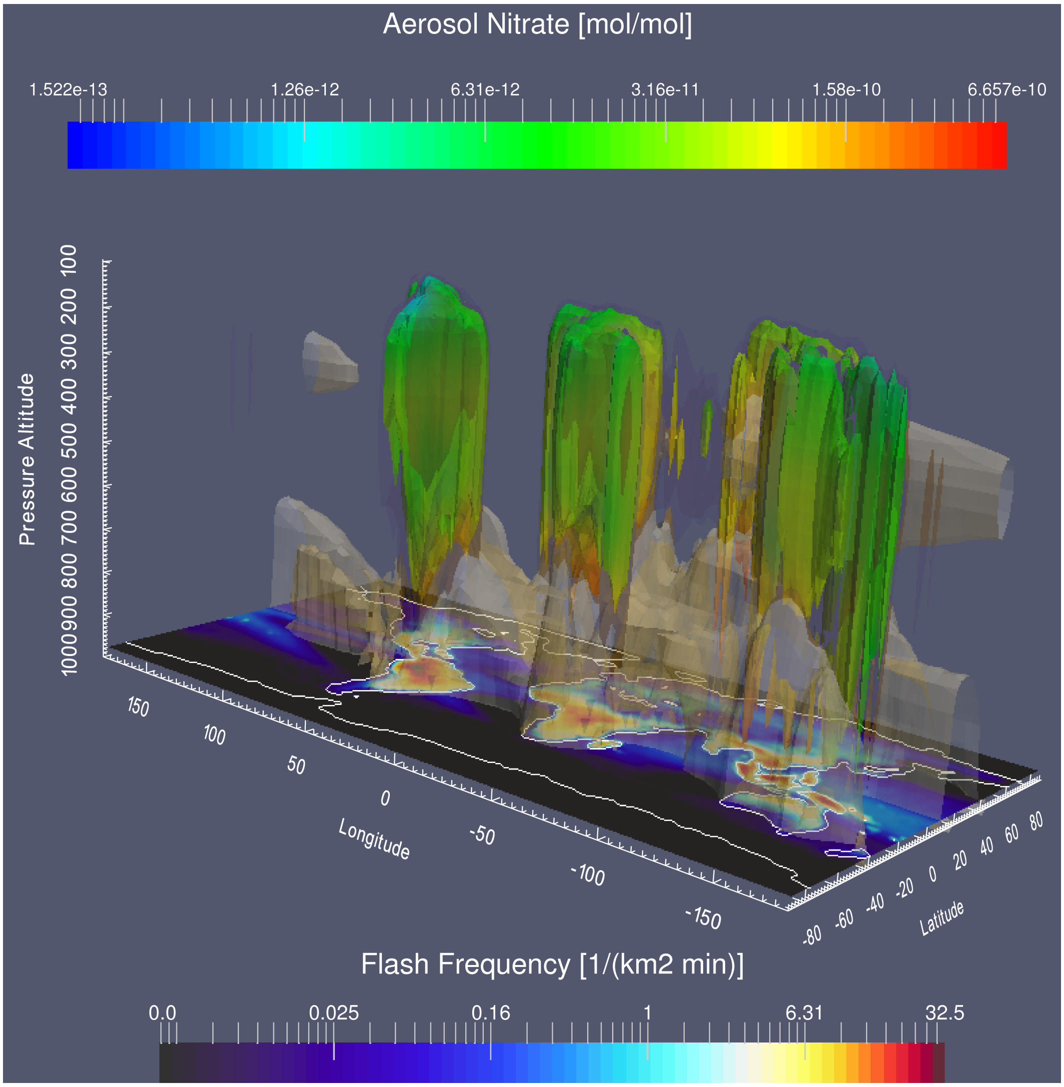

Figure 2. 3D visualisation of LNO$_x$ emissions (coloured isosurface of $1 \cdot 10^{-16}$ kg/(m^{3}s) and darker shaded isosurface of $3 \cdot 10^{-16}$ kg/(m^{3}s)) and the total aerosol nitrate mixing ratios (~~grey~~ gray isosurface of 0.1 ppb$_v$). Additionally, the mean flash rate in 1/~~s~~ (km^2 min) is depicted by the 2D slice at the bottom. Note the logarithmic scaling of both colour bars.

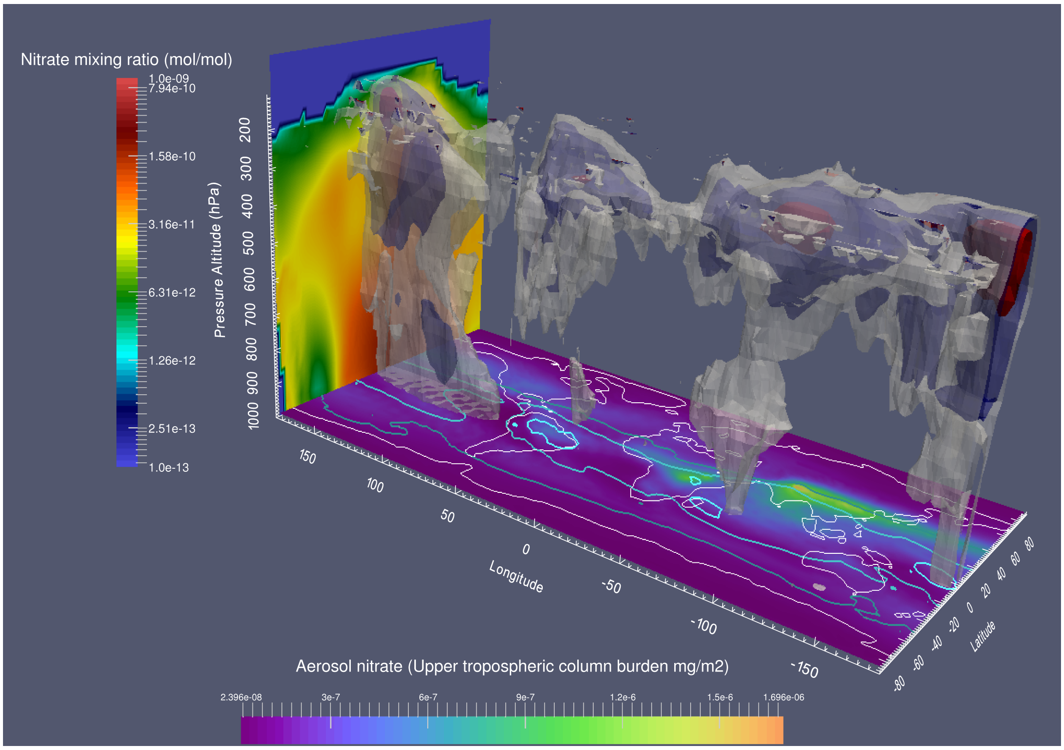

Figure 3. 3D visualisation of the relative differences in tropospheric aerosol nitrate mixing ratios between the simulations with and without LNO_x emissions to the simulation including LNO_x emissions. The ~~white~~ gray isosurface depicts a relative difference of 30%, the blue isosurface of 45%, and the red isosurface of 60%. Additionally, the upper tropospheric aerosol nitrate column burden (in mg/m^2) between 500 hPa and the tropopause is depicted by the coloured panel at the bottom of the graph. The turquoise contour lines depict relative differences of 20%, 40% and 60% difference in this column burden between the two simulations.

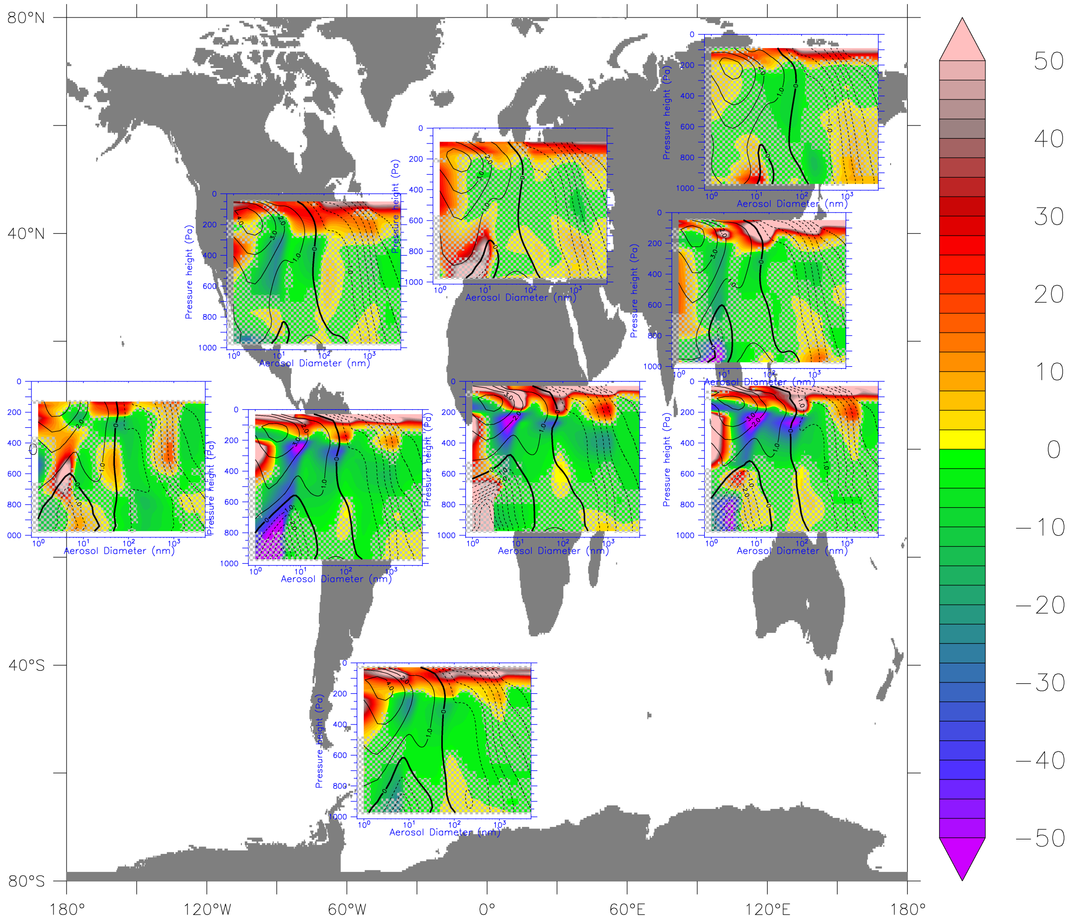

Figure 4. Map of vertically resolved percentage differences (including $\mathrm{LNO_x}$ emissions as the reference case) in the aerosol size distributions as spatial and regional average (for the respective regions). In each small frame the y-axis depicts pressure altitude and the x-axis the aerosol diameter from 1nm to 10 μm. Overlayed are the contours of the absolute values of the size distributions, i.e. the absolute particle numbers calculated from the overlaying of the individual modes, determined from the spatial and temporal mean in particles/cm^3. The solid lines depict 1, 10, 100, 1000 particles, whereas the dashed lines represent 10^{-1}, 10^{-2}, etc. particles. The figure depicts the present day conditions. The respective regions are: Central Pacific (150W:110W, 20S:0), Amazonia (80W:35W, 20S:10N), Eastern US (100W:75W, 30N:45N), South Atlantic (50W:10W, 80S:60S), Central Africa (5E:45E, 10S:10N), Europe (10W:25E, 40N:60N), Indonesia (95E:155E, 12S:10N), East Asia (105E:125E, 20N:42N), Siberia (60E:100E, 50N:70N). The hatched regions mark areas without statistical significance.

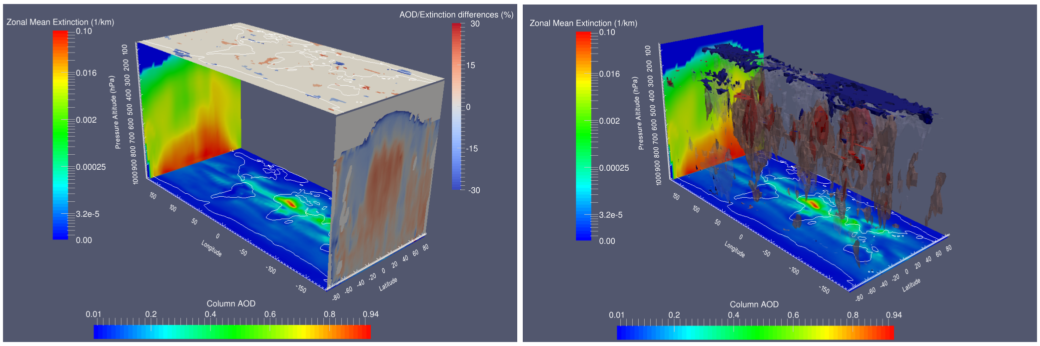

Figure 5. 3D visualisation of aerosol extinction and the influence of $\mathrm{LNO_x}$ emissions. The ~~floor~~ left panel shows a map of the vertically integrated column AOD (at 550 nm) when lightning emissions are included (floor plane). The ceiling depicts the relative differences of the integrated column AOD between the simulation with lightning emissions minus the simulation without lightning $\mathrm{LNO_x}$, with the full setup serving as reference. The back panel displays the zonal average aerosol extinction (in 1/km at 550 nm) of the full simulations (Please, note the logarithmic scale.). ~~Additionally, the~~ The front panel depicts ~~again~~ relative percentage differences due $\mathrm{NO_x}$ emissions from lightning. White areas mark regions without statistical significance.

The right panel also shows AOD (floor) and zonal mean extinction (rear panel), and additionally 3D isosurfaces in the center of the box represent the $+10\%$ (pale red) and $+20\%$ (dark red) of the enhanced extinction due to active $\mathrm{LNO_x}$ emissions, whereas the -10% (pale blue) and -20% (dark blue) isosurfaces mark regions, in which the emissions result in a reduction of the extinction.

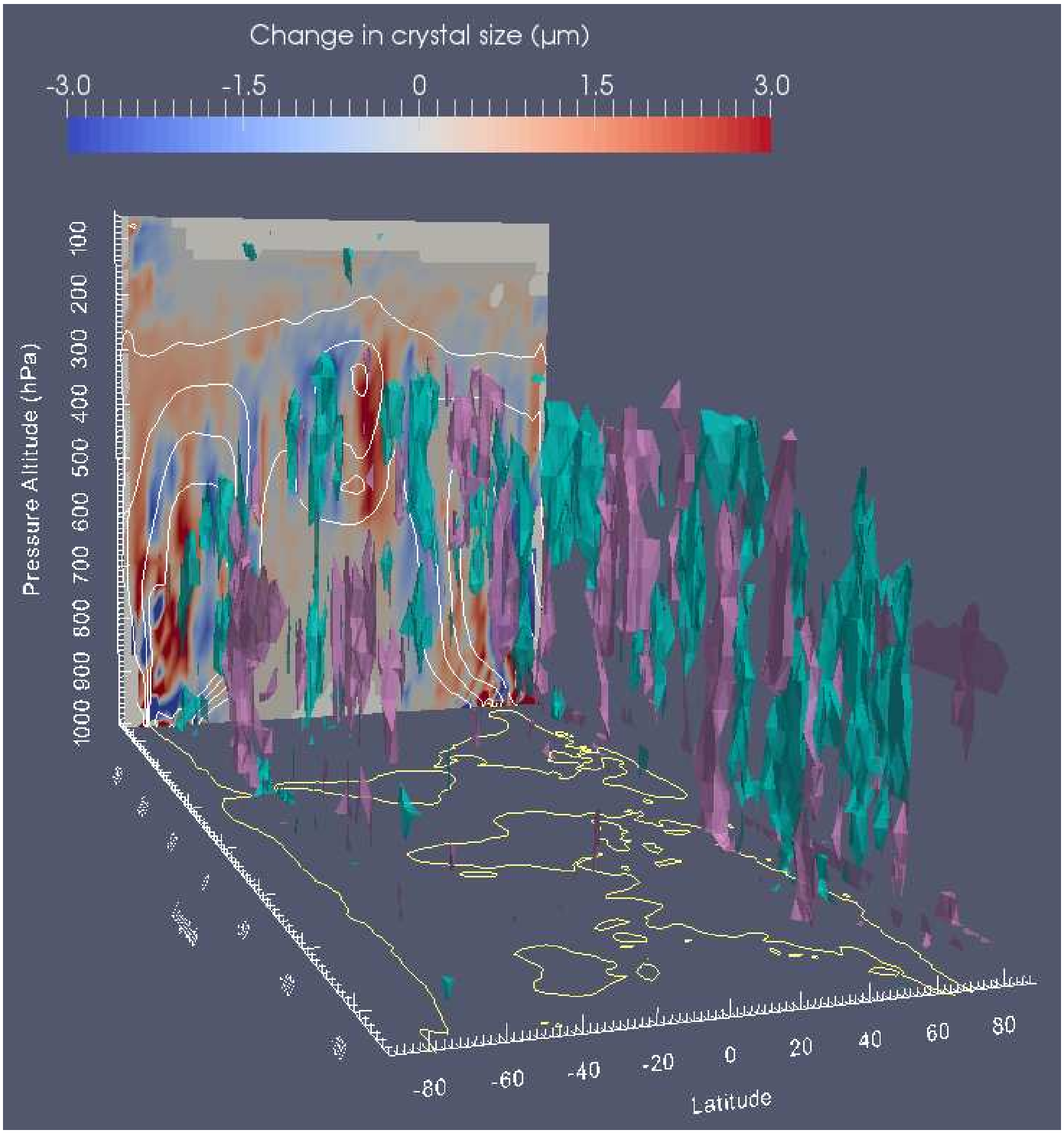

Figure 6. Visualisation of changes in the effective radius of ~~liquid water droplets and~~ ice crystals. The back panel displays the ~~zonal average liquid droplet effect radius (white contours) and the~~ statistically significant absolute changes ~~due to the emissions. The front panel depicts similarly the~~ in zonal average ice crystal ~~effective~~ size ~~(~~in μm. The white contours ~~)~~ depict a mean ice crystal size and ~~the absolute change due to the lightning emissions~~therefore mark regions with large and smaller crystals. Additionally, the isosurfaces represent the regions for substantial absolute changes for the effective ~~radius for water droplets (blue negative, red positive) and effective~~ ice crystal size (turquoise for negative and purple for positive) due the $\mathrm{LNO_x}$ emissions.

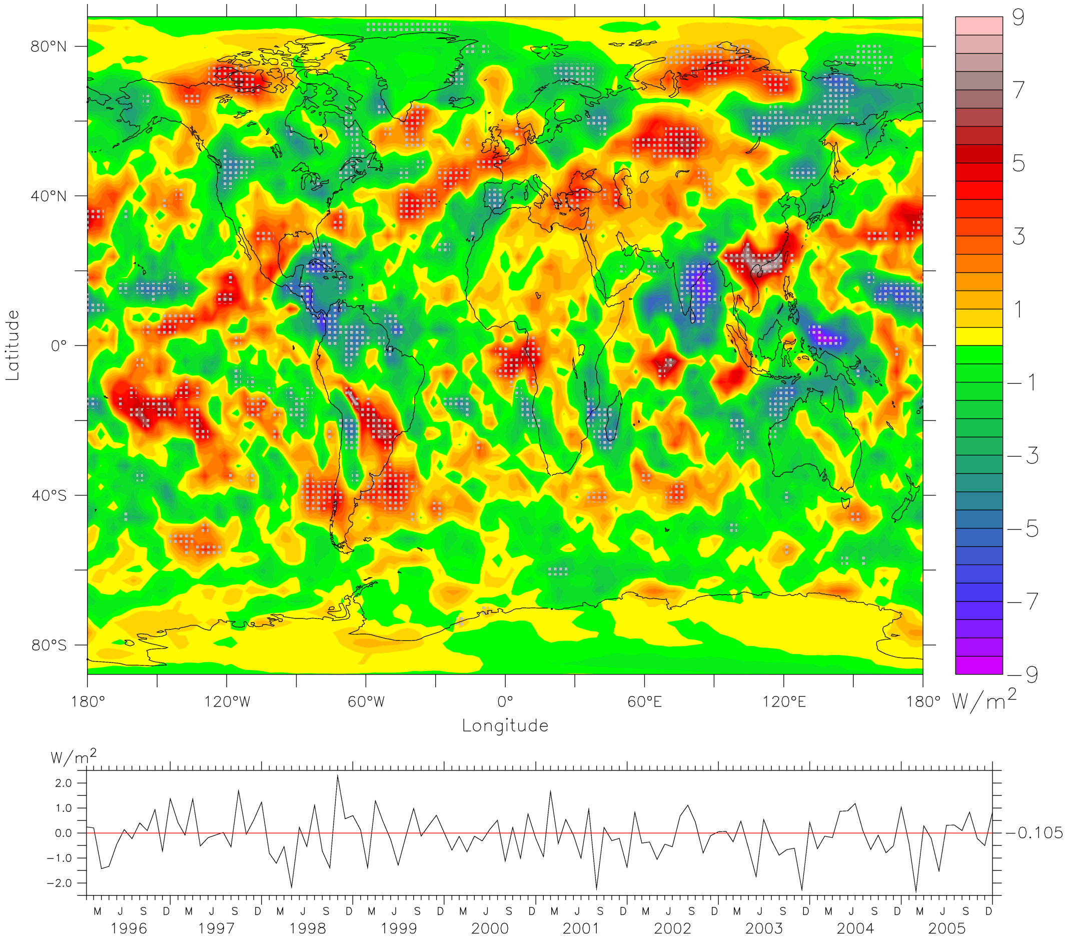

Figure 7. Absolute changes in the shortwave all-sky fluxes at the top of the atmosphere. The upper panel depicts the long term time average of the difference in the simulation with $\mathrm{LNO_x}$ emissions minus the fluxes without lightning emissions. The hatches mark regions with a statistical significant signal (compared to the internal interannual variability). The bottom panel depicts the time series of the monthly mean global mean differences with the red line marking a 0 W/m^2 change.

Table 1. Tropospheric and upper tropospheric burden in the ARG simulation of the important highly oxidised nitrogen species, i.e. gaseous HNO_3, gaseous N_2O_5, aerosol NO_3^- and the sum of those three compounds. All values are given in Gg N (except for the relative differences which are provided in %) and are globally and vertically integrated over the whole and the upper troposphere (500hPa up to the tropopause).

	HNO_3	N_2O_5	NO_3^-	Total N(V+)
Present day				
Absolute values (with LNO_x emissions):				
Tropospheric Column burden	403	8.0	143	554
UT Column burden	174	5.0	27.8	207
Absolute differences due to LNO_x emissions:				
Tropospheric Column burden	162	3.9	20.0	186
UT Column burden	107	3.5	9.0	119
Relative differences in (%) due to LNO_x emissions:				
Tropospheric Column burden	40.1	48.4	14.0	33.5
UT Column burden	61.3	70.3	32.3	57.7
Preindustrial conditions				
Absolute values (with LNO_x emissions):				
Tropospheric Column burden	238	4.5	67.8	310
UT Column burden	143	4.1	15.0	161
Absolute differences due to LNO_x emissions:				
Tropospheric Column burden	158	3.7	25.8	188
UT Column burden	110	3.5	8.1	121
Relative differences in (%) due to LNO_x emissions:				
Tropospheric Column burden	66.6	82.6	38.1	60.6
UT Column burden	76.9	86.0	54.3	75.1

Table 2. CH_4 tropospheric and upper tropospheric (500 hPa to tropopause) lifetime and absolute ~~increase~~ change due to ~~the neglect of~~ LNO_x emissions.

	CH_4 lifetime [years]	Increase of the CH_4 lifetime [years]
Present day		
troposphere	7.4	1.7
UT only	14.0	9.7
Preindustrial conditions		
troposphere	8.9	1.9
UT only	14.1	13.2

The CH_4 lifetime is calculated with the help of the actual methane and OH concentrations, calculating a pseudo-first order loss rate. The corresponding temperature dependent reaction rate is identical to the one used in the online chemistry calculations, using also the temperature output of the simulations. Calculating the lifetime for each individual cell, average values are determined using grid mass weighted factors for certain regions or the whole troposphere.