Reply by the authors to Referee #2's comments on
"Quantifying the impact of global nitrate aerosol on tropospheric composition fields and its production from lightning NOx" (https://doi.org/10.5194/egusphere-2024-1363-RC2)

Anonymous Referee #2 (RC2)

We are grateful to the Referee for their time to review our manuscript and making a number of important points. In the following, we provide our responses to these comments (the Referee's comments are shown in blue).

Summary

This manuscript uses the UKCA chemistry-climate model (CCM) to evaluate the impact of lighting NOx emissions on particulate nitrate chemistry (recently implemented in the UKCA model), and thereby, its influence on climate.

Overall Comment

I see no overall flaws with the study as performed and reported.

Response: Thank you.

However, the manuscript as written oversells its originality and its accuracy. Many global atmospheric chemistry models — including those cited in the introduction — have had nitrate aerosol chemistry for decades, e.g., GEOS-Chem (Park et al., 2004), GISS (Bauer et al., 2007), GFDL (Paulot et al., 2016; and ref. therein).

Response: Our intention was not to oversell the paper's originality or accuracy, and we now add suitable caveats and references as suggested by the Referee.

In Introduction when we say "*Despite the above, few global chemistry-climate models include nitrate aerosol, and usually its effects are completely ignored (Tost, 2017), with the main reason being the chemical complexity of nitrate formation and the semi-volatile nature of ammonium nitrate. To give an example, of the ten global Earth system models that participated in the Aerosol and Chemistry Model Intercomparison Project (AerChemMIP) of the Coupled Model Intercomparison Project Phase 6 (CMIP6), aimed at understanding the effects of reactive gases and aerosols on Earth's climate, only two had an interactive stratospheric and tropospheric gas-phase and aerosol- chemistry scheme together with an explicit treatment of nitrate aerosol (Thornhill et al., 2021)*" those two models are indeed GISS and GFDL (GEOS-Chem is a chemical transport model, not a chemistry-climate model, so not included). We have revised the text to read:

"Although nitrate aerosol has been included in some global models, such as the chemical transport model GEOS-Chem (e.g., Park et al., 2004) and the chemistry-climate models GISS (e.g., Bauer et al., 2007) and GFDL (e.g., Paulot et al., 2016), it is often ignored in global chemistry-climate models (Tost, 2017). This may be partly due to the computational cost of simulating nitrate, combined with the chemical complexity of its formation and the semi-volatile nature of ammonium nitrate, which can reevaporate into the atmosphere (e.g., Stelson et al., 1979). In fact, out of the ten global Earth system models with atmospheric chemistry that participated in the Aerosol and

Chemistry Model Intercomparison Project (AerChemMIP) under the Coupled Model Intercomparison Project Phase 6 (CMIP6), which aims to assess the effects of reactive gases and aerosols on Earth's climate, only the GISS and GFDL models explicitly treated nitrate aerosol along with an interactive stratospheric and tropospheric chemistry scheme (Thornhill et al., 2021)."

Therefore, any of the many publications that have used these models to look at the impact of lightning on atmospheric composition and climate have included the impact of nitrate particles. The main reason that it has not been highlighted in the manuscripts is because it was noticed be negligible compared to other impacts and/or the nitrate simulation was evaluated to have too poor a skill in reproducing observational constraints, precluding meaningful conclusions.

Response: We believe all current global chemistry-climate models include lightning-generated emissions of NO_x (LNO_x) irrespective of whether nitrate aerosol is included or not. The lightning-nitrate relationship exists at least implicitly in studies that have used the models listed by the Referee, even if it has not been explored or quantified.

From the Referee's point of view, it may well have been the case that the impact of LNO_x on nitrate aerosol was noticed to be negligible compared to anthropogenic sources or compared to other impacts (which we assume is what the Referee is stating), but the Referee does not point out any references that support their assertion that "it has not been highlighted … because it was noticed be negligible" nor their assertion about "poor a skill". We would gladly cite any references that attest to either of these assertions, but the Referee has not provided references.

We have not seen any modelling study reported in the peer-reviewed literature that investigates the impact (significant or otherwise) of lightning on nitrate concentrations, other than that by Tost (2017). Similarly, we have not come across any study that examines how the modelled tropospheric composition is impacted when nitrate is accounted for.

We modify the relevant paragraph in Introduction to read:

"The area of quantifying the role of LNO_x production on aerosol, particularly with nitrate aerosol included, has only received very limited attention compared to its role on gaseous atmospheric composition, and this could be due to reasons such as the inference that the low LNO_x emission (~ 12%) compared to anthropogenic and biomass burning NO_x sources contributes negligibly to nitrate concentrations. To our knowledge, the global modelling by Tost (2017), which involves a modal aerosol scheme with nitrate included, is the only study to explicitly examine the impact of LNO_x on aerosol. It shows that LNO_x (parameterised via the Price and Rind (1992) (PR92) scheme described below) is a significant source of nitrate in the upper troposphere and influences the aerosol size distribution and radiation. It is reported that chemical conversion of LNO_x into HNO_3 is more favourable in the middle to upper troposphere, where lightning NO_x mostly occurs, as compared to within the atmospheric boundary layer (where the dominant NO_x and NH_3 sources are located) due to differences in chemical composition, chemical reactivity, and loss processes (Tost, 2017). Tost (2017) points to observational support for the occurrence of both NH_3 and NO_3 aerosol in convective outflows so that the formation of NH_4NO_3 is likely because of the low temperatures in the upper troposphere. Therefore, LNO_x can change the spatial distribution of nitrate concentrations and concomitant climate impacts. Given the emerging importance of nitrate as sulfate concentrations wane it is important to assess the relative importance of all nitrate sources. At the same time, how modelled global atmospheric composition is impacted when nitrate is accounted needs to be quantified."

The UKCA implementation of nitrate as reported in Jones et al. (2021) similarly shows very poor skill in reproducing surface nitrate observations over the United States and Europe.

Response: The UKCA nitrate scheme compares very well to other AeroCom models and Earth system models (e.g. Table 3, Jones et al, 2021) on a global basis, and showed significant skill over the United States (e.g., R = 0.92 and 0.43 for NH_4 and NO_3, respectively, Figure 5, Jones et al., 2021), although the Referee is correct that the skill over Europe was at the time low. Jones et al (2021) attributed this to uncertainties in the NH_3 emissions inventory, as also found by Drugé et al. (2019) (cited in our paper). Recent tests with UKCA nitrate in the Met Office's AQUM (Air Quality in the Unified Model) have shown significant skill in predicting air quality episodes (https://www.ukca.ac.uk/images/5/59/PA_UKCA_Dec2022.pdf). In short, we have no reservations over the UKCA nitrate scheme.

I would support this manuscript's publication if it is recast as a theoretical exercise assuming the chemistry is correct (the lightning chemistry-climate interactions via nitrate and cloud microphysics are very interesting!) or if a proper evaluation of the in situ nitrate and aerosol optical depths were included to provide confidence in the simulation.

Response: In this paper, we use a state-of-the-art GCM with comprehensive stratospheric and tropospheric chemistry (StratTrop 1.0, used in UKESM2, Archibald et al., 2020 (cited)) as outlined in Section 3, with chemical emissions following the CMIP6 protocol, i.e., based on observations. We believe, it is incorrect therefore to describe this paper as a theoretical exercise when it is clearly designed to be as close to reality as possible. The only "theoretical" aspect of the methodology is the use of constant year 2000 conditions, which is used to remove the transient aspect of the emissions over the 20-year simulation period whilst maintaining a realistic geospatial emissions inventory. Rather, our paper can be considered as a sensitivity study. The results provide an important and useful quantification of the variation of the various fields considered with respect to the variation in LNO_x and to whether or not nitrate is considered.

We add the following caveat to Conclusions:

"Nitrate concentrations are sensitive to precursor emissions, and in this study, we used constant emissions forcings representative of the year 2000 following CMIP6 protocol. A more comprehensive study could include transient emissions to investigate recent trends in ammonium nitrate concentrations."

We add the following to Introduction:

"Our modelling can be considered as a study of sensitivity of global fields of interest to changes in lightning NO_x without and with nitrate aerosol."

And in Conclusions we already state:

"In this sensitivity focused study, we have essentially addressed two problems through the use of a global chemistry-climate model, UM-GA8.0-UKCA: 1) quantifying the impact of including nitrate aerosol on tropospheric composition, AOD and radiation, and 2) the dependency of these effects on lightning generated NO_x."

A comparison with MODIS derived AOD has been done by Jones et al. (2021) for UM-UKCA without and with nitrate. We find that the global change in AOD for the LNO_x range considered (0–5.2 Tg N yr^{-1}) is ~ 0.0015, which is ten times smaller than the increase of ~ 0.0154 in AOD when nitrate is included (see our Section 4.5 and Figure 13). AOD is a bulk aerosol quantity, and we

believe that its measurements are unlikely to discern the magnitude of changes in the modelled AOD in response to changes in LNO_x and nitrate, particularly LNO_x. Indeed, for example, Levy et al. (2013, https://doi.org/10.5194/amt-6-2989-2013) note that the uncertainty in MODIS global AOD cannot be reduced below ± 0.03, or 15–20% of global mean AOD. This uncertainty in observations is much greater than the above modelled variation in AOD in response to changes in LNO_x and is also larger than the increase in AOD when nitrate is included. Therefore, an evaluation using AOD measurements would prove indecisive as the modelled AOD differences will most likely be within the observational uncertainty.

Essentially, to do a targeted evaluation of the impact of LNO_x on aerosol, we need new data, e.g. from aircraft measurements in the tropics, coupled with some high-resolution regional chemical transport modelling, which is beyond the scope of the present study.

We agree with the Referee that the lightning chemistry-climate interactions via nitrate and cloud microphysics are very interesting. We believe more research is needed in this area, but for the present we include the following plot (which can go in either section 4.5 as Figure 15 or Supplement S2 where CDNC is discussed in more detail) on the impact of nitrate and LNO_x on the modelled global-mean cloud droplet number concentration (CDNC). We state "We also examined the impact of nitrate on the modelled tropospheric cloud droplet number concentration (CDNC) (see Supplement S2) and found that compared to the no-LNOx case, there is a 3.1% increase in the mean tropospheric CDNC when LNO_x is considered (via the Lu21 scheme). The incorporation of nitrate in the model causes an average increase of ~ 3.5% in CDNC for any given LNO_x value. The average increase in CDNC per Tg N yr^{-1} of LNO_x is 0.035 cm^{-3}.

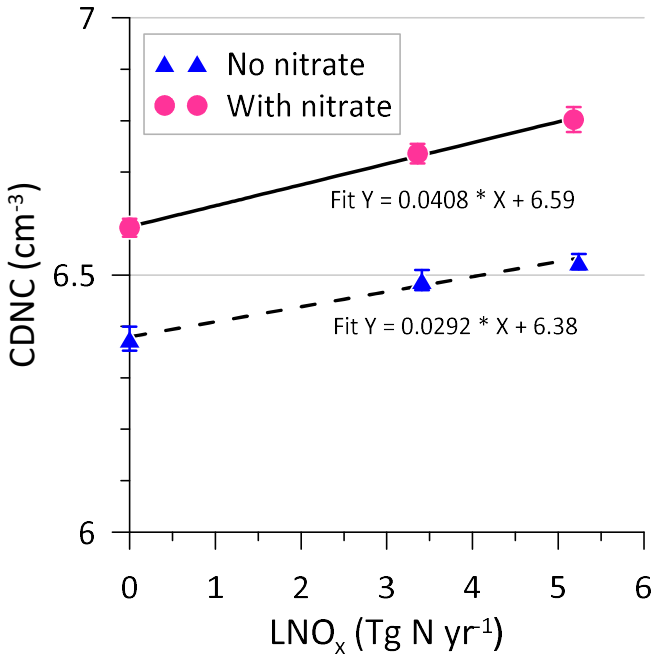

Figure 15: Modelled annual-mean tropospheric cloud droplet number concentration (CDNC), as a function of lightning-generated NO_x. The lines are linear least squares fits. The error bars correspond to a 1-sigma standard deviation.