Supplement of

Quantifying the impact of global nitrate aerosol on tropospheric composition fields and its production from lightning NO$_x$

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Contents

S1. Impact on tropospheric sulfate with the inclusion of nitrate

We also looked at how tropospheric sulfate is impacted by the inclusion of nitrate. Figure S1 shows the differences in the
column mass burdens of sulfate between the Lu21 simulation with nitrate and that without nitrate for various aerosol modes.
The main feature is that with nitrate on, sulfate mass is transferred from the Aitken (soluble) mode to accumulation mode (via
aerosol growth with ammonium sulfate formation) and also to some extent to the coarse model. The mean global reduction in
sulfate mass burden in the Aitken (soluble) mode is 4.3% of the mean total global sulfate burden and this reduction is mainly
concentrated between 0–60° N in the lower troposphere. The corresponding increase in sulfate mass burden in the accumulation
mode is 3.6%, concentration in the same region. The small decrease in the nucleation mode is primarily confined to regions
between 15° S – 50° N extending from the upper troposphere to the top of lower troposphere (plot not shown). The small
increase in the coarse mode sulfate is mainly near the surface, between 15°–60° N. The total difference in the last plot of Figure
S1 suggests a small overall decrease (∼ 0.5%) in the sulfate aerosol mass.
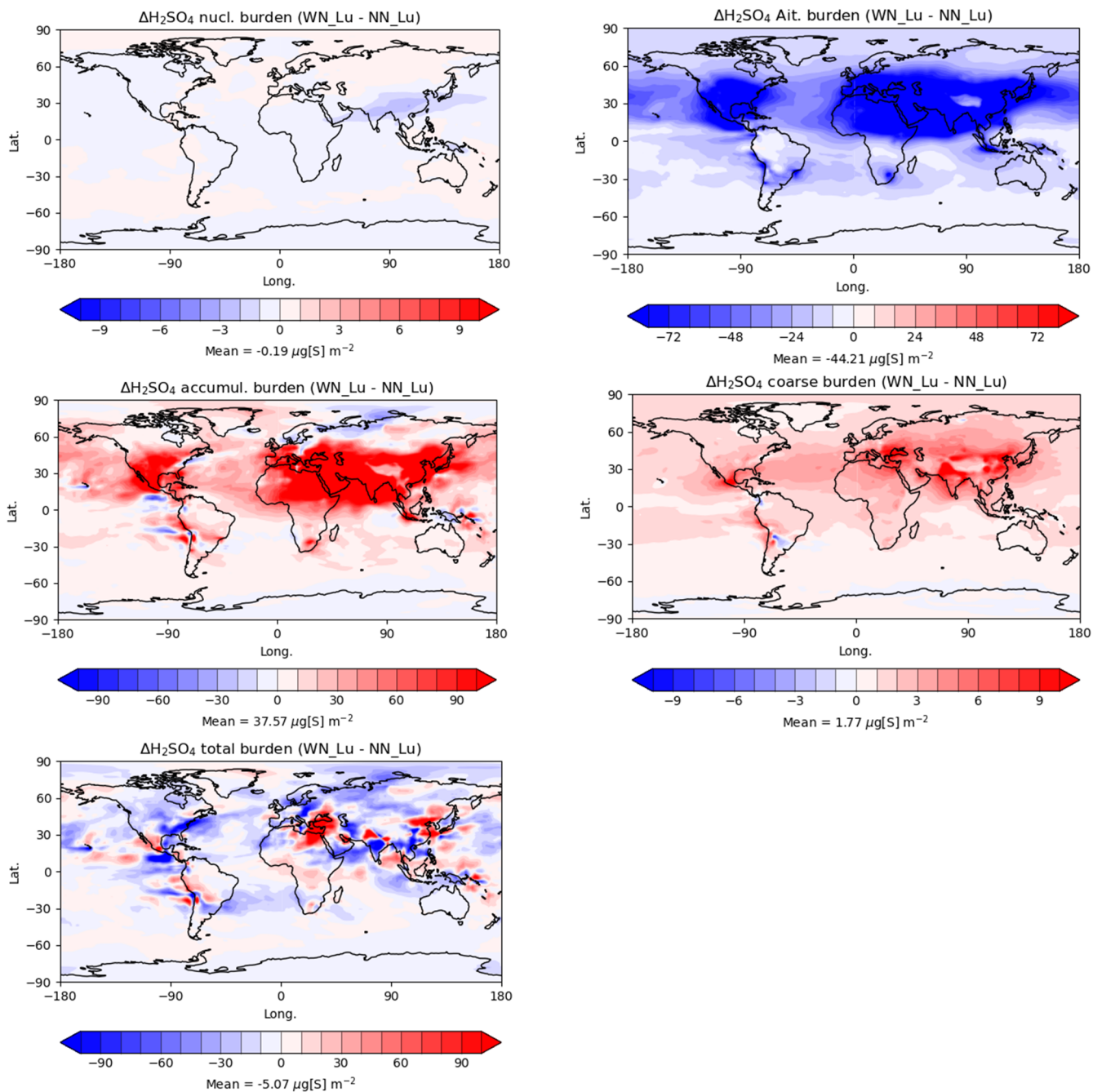
Figure S1: Differences in the annual-mean mass burdens of tropospheric sulfate aerosol between the Lu21 simulation with nitrate and that without nitrate for various aerosol modes: (a) nucleation soluble, (b) Aitken soluble, (c) accumulation soluble, (d) coarse soluble, and (e) the total difference.

S2. Impact on cloud droplet number concentration (CDNC)

We also examine modelled cloud droplet number concentration (CDNC) (one could also look at CCN or CN, but such output was not available from the model runs made). CDNC can be used as a proxy for CCN (the latter is a measure of the potential to form cloud droplets at the bottom of the cloud. Typical cloud droplet is 20 µm in diameter. Apart from modifying the aerosol size distribution, nitrate also modifies the chemical composition of the aerosol which can change the cloud condensation nuclei (CCN) efficiency of the particles with ramifications for the indirect aerosol effects (Tost, 2017). In Figure S2 (left), the zonal mean tropospheric CDNC distribution obtained using the Lu21 scheme with nitrate shows that droplets are mostly confined to the bottom layers of the lower troposphere, with greater concentrations in the tropics. Compared to the PR92 scheme, there is a small (~ 1%) increase in the mean tropospheric CDNC (Figure S2 (middle)), and it is apparent that most of the increase is in the Southern Hemisphere and parts of the northern tropic. 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), and this increase is mostly in the tropics and the Southern Hemisphere (Figure S2 (right)). (This is consistent with Tost (2017) who find that in the troposphere, an increase in cloud droplets is simulated in case of active lightning emissions in the tropics.)
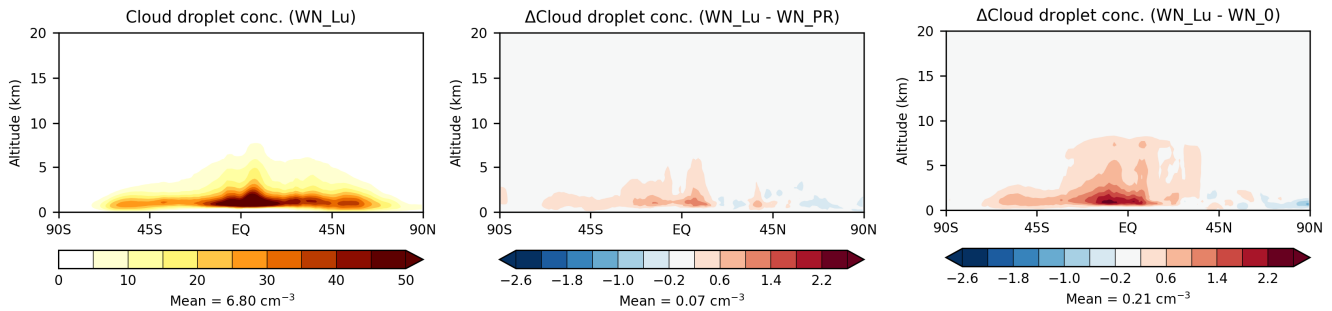
Figure S2: Zonal mean tropospheric cloud droplet number concentration (CDNC) from the Lu21 simulation (left), the difference between the Lu21 and PR92 simulations (Lu21 – PR92) (middle), and the difference between the Lu21 and no-LNOₓ simulations (Lu21 – no-LNOₓ) (right). All simulations with nitrate on.

The difference between the zonal mean tropospheric CDNC from the Lu21 simulation with and without the nitrate scheme presented in Figure S3 indicates that the incorporation of nitrate in the model causes increases in CDNC in the lower troposphere, particularly within the tropics and between 30°–60° N, with a mean tropospheric increase of 4.2%. It is clear that there are greater changes when nitrate is considered compared to changes caused by changes in LNO_x scheme.
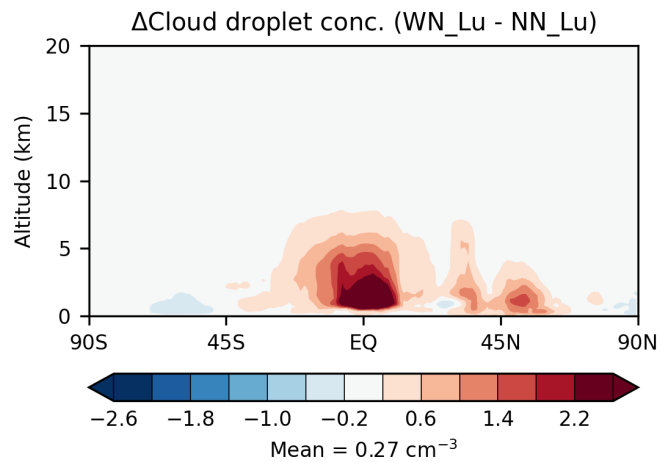
Figure S3: Difference between the zonal mean tropospheric cloud droplet number concentration (CDNC) from the Lu21 simulation with and without the nitrate scheme.

60

References

Tost, H.: Chemistry–climate interactions of aerosol nitrate from lightning, Atmos. Chem. Phys., 17, 1125–1142, https://doi.org/10.5194/acp-17-1125-2017, 2017.