



Supplement of

Exploring the sensitivity of atmospheric nitrate concentrations to nitric acid uptake rate using the Met Office's Unified Model

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S1 Unified Model configuration

The Unified Model (UM)'s dynamical core, Even Newer Dynamics for General atmospheric modelling of the environment (ENDGame), is a semi-implicit and semi-Lagrangian solver for the non-hydrostatic deep-atmosphere equations of motion (Wood *et al.*, 2014). Mass conservation of moist tracers is achieved using the Optimised Conservative Filter scheme (Zerroukat and Allen, 2015). Atmospheric radiative transfer is modelled assuming the two-stream approximation, with 6 wavebands in the shortwave spectrum and 9 wavebands in the longwave spectrum (Manners *et al.*, 2015). Large-scale cloud is modelled with the Prognostic Cloud fraction and Prognostic Condensate (PC2) scheme (Wilson *et al.*, 2008) and cloud microphysics uses the single-moment scheme based on Wilson and Ballard (1999) with extensive modifications. Full details of GA7.1/GL7.0 are provided in Walters *et al.* (2019) and Mulcahy *et al.* (2018).

The United Kingdom Aerosol and Chemistry (UKCA) model configuration utilised for this study, StratTrop1.0, simulates the chemical cycles of O_x, HO_x, NO_x, and halogenic compounds; the oxidation of various organic compounds; and heterogenous chemistry on Polar Stratospheric Cloud (PSCs), Nitric Acid Trihydrate (NAT) and tropospheric aerosols, with 84 species and 291 chemical reactions. Interactive photolysis in UKCA is modelled with the Fast-JX scheme, which covers the wavelength range 177-750 nm (Telford *et al.*, 2013). Gaseous wet deposition follows the effective Henry's law approximation described in Giannakopoulou *et al.* (1999) while dry deposition is treated with a resistance type model (O'Connor *et al.*, 2014).

In terms of the GLOMAP-mode aerosol scheme (Mann *et al.*, 2010), cloud condensation nuclei are converted to cloud droplets using UKCA-Activate which is based on the Abdul-Razzak and Ghan (2000) parameterization, while autoconversion of cloud droplets to rainwater follows Khairoutdinov and Kogan (2000) (Boutle *et al.*, 2014; West *et al.*, 2014; Mulcahy *et al.*, 2018). Aerosol processes in GLOMAP-mode include binary homogeneous SO₄ nucleation, mode-merging, cloud processing, condensational uptake of H₂SO₄ gas and secondary organic vapour from UKCA, and coagulation within and between modes. Aerosol removal processes treated by UKCA-mode include dry deposition and sedimentation, in-cloud scavenging by the autoconversion of cloud to rainwater; beneath cloud scavenging by rain and snow impaction; and convective plume scavenging (Mann *et al.*, 2010, Mulcahy *et al.*, 2020).

S2 Primary Ammonium Nitrate Production

S2.1 Determine the equilibration constant (K_p) of the ammonium-nitrate system

Free ammonia is used for the neutralization of nitric acid to ammonium nitrate aerosol following the equilibrium reaction:



The equilibrium constant (K_p) of (1) strongly depends on relative humidity and temperature. The parameterization used for this dependence is based on Mozurkewich (1993). Firstly, the deliquescence relative humidity (DRH , %) is calculated based on Seinfeld and Pandis (1998):

$$DRH = \exp\left(\frac{723.7}{T} + 1.6954\right) \quad [S2]$$

where T is the air temperature (K). For relative humidities lower than DRH, $K_p = K_{pd}$ and is calculated with:

$$K_{pd} = \exp\left[118.87 - \frac{24084}{T} - 6.025 \ln(T)\right] \quad [S3]$$

For relative humidities higher than DRH, $K_p = K_{ph}$ and depends on both temperature and relative humidity (RH , %) and is calculated based on:

$$K_{ph} = K_{pd} (p_1 - p_2 RH_1 + p_3 RH_1^2) RH_1^{1.75} \quad [S4]$$

With RH_1 defined as $(1-RH/100)$ and p_1 , p_2 , and p_3 provided by:

$$p_1 = \exp\left[-135.94 + \frac{8763}{T} + 19.12 \ln(T)\right] \quad [S5a]$$

$$p_2 = \exp\left[-122.65 + \frac{9969}{T} + 16.22 \ln(T)\right] \quad [S5b]$$

$$p_3 = \exp\left[-182.61 + \frac{13875}{T} + 24.46 \ln(T)\right] \quad [S5c]$$

S2.2 Determine the equilibration time scale (τ) for the ammonium-nitrate system

Ammonium nitrate formation is primarily a condensation process rather than nucleation process (Ackerman *et al.*, 1995) and therefore depends on the concentration and properties of existing atmospheric aerosol. In UKCA-mode, each lognormal mode is represented by an overall particle number mixing ratio (N in eq-kg/kg(air)) and individual constituent mass mixing ratios (M in kg/kg(air)). Each constituent (e.g. dust, sulphate, etc) is associated with a density (ρ) and molar mixing ratio. Each mode has its own fixed width parameter (σ). The geometric median diameter for the mode (D_p) can be calculated thus:

$$D_{p,r} = \left[\frac{6}{\pi e^{4.5 \ln^2 \sigma_r}} \times \frac{k_B}{R} \times \frac{1}{N_r} \sum_x \frac{M_{x,r}}{\rho_x} \right]^{1/3} \quad [S6]$$

The indices r and x represent the mode and constituent respectively. $R = 287.05 \text{ J Kg}^{-1} \text{ K}^{-1}$ is the specific gas constant of dry air and $k_B = 1.3804 \times 10^{-23} \text{ J K}^{-1}$ is the Boltzmann constant. Using properties of the lognormal distribution, the modal mean diameter D_μ is then:

$$D_{\mu,r} = D_{p,r} e^{0.5 \ln^2 \sigma_r} \quad [S7]$$

UKCA-mode explicitly calculates hygroscopic growth using the Zdanovskii-Stokes-Robinson (ZSR) algorithm, with aerosol-associated water volume a diagnostic rather than a prognostic. As the UKCA emissions scheme precedes UKCA-mode within the UKCA calling sequence, the water content and concomitantly the hygroscopic growth factor is not readily available to the nitrate production scheme. Instead we grow the particles using a relative humidity dependent growth parameterisation based on ammonium sulphate (Gerber *et al.*, 1985).

$$D_{w,r} = 2 \times \left[\frac{0.6628 D_{\mu,r}^{3.082}}{1.1658 \times 10^{-13} D_{\mu,r}^{-1.428} - \log_{10} \frac{RH}{100}} + D_{\mu,r}^3 \right]^{1/3} \quad [S8]$$

To test the Gerber parameterisation, we utilise annual-mean climatology from an atmosphere-only integration of the MetUM (N96). Annual-mean aerosol wet diameters from UKCA for aerosol mode (calculated using the ZSR scheme every time-step) are compared to wet diameters from applying the Gerber parameterisation to annual-mean dry diameters. Every grid-cell below the tropopause (i.e. the bottom-most 49 levels) is treated equally irrespective of aerosol number concentration and the results presented for Aitken and accumulation modes in Figure S1. The Gerber parameterisation over-estimates hygroscopic growth compared to ZSR for many gridcells in which the actual diameter $> 0.25 \mu\text{m}$ owing probably to the differing aerosol compositions which is not accounted for in Eq. S8.

We follow Makar *et al.* (1998) in expressing the equilibration time scale (τ) as the reciprocal of the product of a condensation rate (k_{HNO_3}) and the number concentration of aerosol particles (N). The Schwartz (1986) first-order uptake scheme is utilised to determine k_{HNO_3} for each mode (Eqs S9-S12).

$$D_g = \frac{3}{8A_c \rho_a d_a^2} \left[\frac{m_a R_a T}{2\pi} \times \frac{m_a + m_{HNO_3}}{m_{HNO_3}} \right]^{\frac{1}{2}} \quad [S9]$$

$$\lambda = \frac{3D_g}{v} = \frac{3D_g}{\sqrt{\frac{8R_a T}{\pi m_{HNO_3}}}} \quad [S10]$$

$$K_{n,r} = \frac{2\lambda}{D_{w,r}} \quad [S11]$$

$$k_{HNO_3,r} = \frac{2\pi D_{w,r} D_g}{1 + \frac{4K_{n,r}}{3\gamma} \times \left(1 - \frac{0.47\gamma}{1 + K_{n,r}}\right)} \quad [S12]$$

Equations S9-S12 determine the molecular diffusivity coefficient (D_g , m^2s^{-1}), the mean free path (λ , m), the Knudsen number (K_n), and the modal condensation rate ($k_{HNO_3,r}$, m^3s^{-1}). Constants in the algorithm include the Avogadro constant $A_c = 6.022 \times 10^{23} \text{ mol}^{-1}$, the gas constant of dry air $R_a = 8.314 \text{ J mol}^{-1} \text{ K}^{-1}$, the molar mass of dry air $m_a = 0.029 \text{ kg mol}^{-1}$, the molar mass of HNO_3 $m_{HNO_3} = 0.063 \text{ kg mol}^{-1}$, the molecular diameter of dry air $d_a = 4.5 \times 10^{-10} \text{ m}$, and the reactive uptake coefficient (γ) for HNO_3 which we assume to be invariant at 0.193 (Feng and Penner, 2007). Variables in Eqs S9-S12 include the temperature T (K) and air density ρ_a (kg m^{-3}). In Eq S10, v is the mean molecular speed (m s^{-1}).

Equations S9-S12 are also applied to CLASSIC dust bins 1-3 (Woodward *et al.*, 2001), with representative diameters ($D_{w,r}$ in Eqs S11 and S12) of 0.088 μm , 0.28 μm and 0.88 μm for Bins 1-3 respectively. The RH-dependent uptake coefficient for dust (γ) follows Fairlie *et al.* (2010). Finally, multiplying the rate coefficients (k) by particle number concentrations (N , m^{-3}) gives the rate at which HNO_3 gas is transferred to the aerosol phase (Nk , s^{-1}).

$$Nk_{AIT} = k_{HNO_3,ait} \times N_{ait} + k_{dust,bin1} \times N_{dust,bin1} \quad [S13]$$

$$Nk_{ACC} = k_{HNO_3,acc} \times N_{acc} + k_{dust,bin2} \times N_{dust,bin2} + 0.5 \times k_{dust,bin3} \times N_{dust,bin3} \quad [S14]$$

The equilibrium timescale (τ) is then:

$$\tau = \frac{1}{Nk_{AIT} + Nk_{ACC}} \quad [S15]$$

The same annual-mean fields as utilised for the Gerber approximation (Fig.S1) are used to investigate the equilibrium timescale (Fig. S2) and the accumulation to Aitken production ratio (Fig.S4). Over key ammonium nitrate regions such as Europe, South East Asia and Eastern North America, the equilibrium timescale is less than 10 minutes up to a nominal altitude of 2 km (Fig. S2). This is commensurate with the typical timescales in Ackerman *et al.* (1995) for particles with radii between 0.1 and 0.5 μm . At the surface, ammonium nitrate production is, on a global-mean basis, a factor of 8 greater than Aitken production, which reduces to a factor of 1.5 at 3 km altitude owing to the preponderance of smaller aerosols at higher altitudes (Fig. S4).

S2.3 Determine the ammonium sulphate neutralization state

Mass mixing ratios from UKCA [X] are firstly converted to molar concentrations $\{X\} = \frac{[X]\rho_a}{M_X}$, where ρ_a is the air density in (kg m^{-3}) and M_X is the molar weight of substance X in (kg mol^{-1}). Modal

concentrations are denoted by $\{X\}_r$ where the subscript $r = 1, \dots, 3$ denotes the mode (Aitken, accumulation and coarse respectively). Total concentrations are denoted by $\{X\}$ where $\{X\} = \sum_{r=1,3} \{X\}_r$. Output quantities are denoted $\{\dot{X}\}$.

Total ammonia, nitrate and sulphate molar concentrations use a different notation, following Hauglustaine *et al.* (2014).

$$T_N = \{\text{NO}_3\} + \{\text{HNO}_3\} \quad [S16a]$$

$$T_A = \{\text{NH}_4\} + \{\text{NH}_3\} \quad [S16b]$$

$$T_S = \{\text{SO}_4\} \quad [S16c]$$

The sulphate equilibrium state is determined using the concentrations of ammonia (T_A) and sulphate (T_S) following Metzger *et al.* (2002):

$$\Gamma_{\text{SO}_4} = \begin{cases} 2 & 2 T_S < T_A \\ 1.5 & T_S < T_A < 2 T_S \\ 1 & T_A < T_S \end{cases} \quad \begin{array}{l} 2\text{NH}_3 + \text{H}_2\text{SO}_4 \rightarrow (\text{NH}_4)_2\text{SO}_4 \\ 3\text{NH}_3 + 2\text{H}_2\text{SO}_4 \rightarrow (\text{NH}_4)_3\text{H}(\text{SO}_4)_2 \\ \text{NH}_3 + \text{H}_2\text{SO}_4 \rightarrow (\text{NH}_4)\text{HSO}_4 \end{array} \quad [S17a]$$

The total number of moles of ammonia that remain after SO_4 neutralization equals:

$$T_A^* = \text{Max}(0, T_A - \Gamma_{\text{SO}_4} \times T_S) \quad [S18]$$

However, the aim is to neutralise SO_4 mode by mode (starting with the smallest mode) to determine the existing NH_4 mixing ratio associated with SO_4 , which is achieved using the following algorithm:

Initial conditions (total available ammonia + ammonium for neutralisation):

$$T_{A,\text{rem}} = T_A \quad [S19a]$$

For $r = 1, \dots, 3$ where r denotes the size mode (1=Aitken, 2=Accumulation, 3=Coarse), the moles of NH_4 associated with existing SO_4 equals:

$$\{\text{NH}_4(\dot{\text{SO}}_4)\}_r = \text{Min}(T_{A,\text{rem}}, \Gamma_{\text{SO}_4} \times \{\text{SO}_4\}_r) \quad [S19b]$$

$$T_{A,\text{rem}} = \text{Max}(0, T_{A,\text{rem}} - \{\text{NH}_4(\dot{\text{SO}}_4)\}_r) \quad [S19c]$$

It can be shown that:

$$T_A^* = \text{Max}(0, T_A - \Gamma_{\text{SO}_4} \times T_S) \equiv \text{Max}\left(0, T_A - \sum_{r=1,3} \{\text{NH}_4(\dot{\text{SO}}_4)\}_r\right) \quad [S20]$$

The overlying dot in $\{\text{NH}_4(\dot{\text{SO}}_4)\}_r$ is to differentiate output quantities from input. Also, let $\{\text{NH}_4(\dot{\text{SO}}_4)\} = \sum_{r=1,3} \{\text{NH}_4(\dot{\text{SO}}_4)\}_r$ denote the total NH_4 concentration associated with sulphate. The next step will also require the concentration of NH_3 gas used to neutralise SO_4 :

$$\{\text{NH}_3(\dot{\text{SO}}_4)\} = \text{Max}(0, \{\text{NH}_4(\dot{\text{SO}}_4)\} - \{\text{NH}_4\}) \quad [S21]$$

S2.4 Produce ammonium nitrate if enough ammonium remains

If $T_A^*T_N > K_p$, then ammonium nitrate forms from the condensation of ammonia and nitric acid onto existing aerosol particles. Firstly, the number of moles of $\{\text{NH}_4\dot{\text{NO}}_3\}$ in equilibrium is calculated according to Seinfeld and Pandis (1998):

$$\{\text{NH}_4\dot{\text{NO}}_3\} = \frac{1}{2} \left[T_A^* + T_N - \sqrt{(T_A^* + T_N)^2 - 4(T_N T_A^* - K_p)} \right] \quad [S22]$$

Before calculating the time-dependent concentration of ammonium nitrate, it is first necessary to update the NH_3 concentration for any neutralisation of SO_4 that might have occurred:

$$\{\text{NH}_3\}_u = \text{Max}(0, \{\text{NH}_3\} - \{\text{NH}_3\dot{(\text{SO}_4)}\}) \quad [S23]$$

The equilibrium concentrations of HNO_3 and NH_3 are calculated thus:

$$\{\text{NH}_3\}_{eq} = \text{Max}(0, T_A^* - \{\text{NH}_4\dot{\text{NO}}_3\}) \quad [S24a]$$

$$\{\text{HNO}_3\}_{eq} = \text{Max}(0, T_N - \{\text{NH}_4\dot{\text{NO}}_3\}) \quad [S24b]$$

The equilibrium timescale (τ) from Section S2.2 (Eq. S15) is now introduced. The time-dependent concentrations of HNO_3 and NH_3 (accounting for the model time-step Δt) are now calculated:

$$\{\dot{\text{NH}}_3\} = \text{Max} \left(0, \{\text{NH}_3\}_u - \left(1 - e^{-\frac{\Delta t}{\tau}} \right) \times (\{\text{NH}_3\}_u - \{\text{NH}_3\}_{eq}) \right) \quad [S25a]$$

$$\{\dot{\text{HNO}}_3\} = \text{Max} \left(0, \{\text{HNO}_3\}_{eq} - \left(1 - e^{-\frac{\Delta t}{\tau}} \right) \times (\{\text{HNO}_3\}_{eq} - \{\text{HNO}_3\}) \right) \quad [S25b]$$

The time-dependent total molar concentrations of NO_3 and HNO_3 are re-calculated:

$$\{\dot{\text{NH}}_4\} = \text{Max}(0, T_A^* - \{\dot{\text{NH}}_3\}) \quad [S26a]$$

$$\{\dot{\text{NO}}_3\} = \text{Max}(0, T_N - \{\dot{\text{HNO}}_3\}) \quad [S26b]$$

The quantities in Eq S26 represent total molar concentrations. In order to speciate the concentration between modes, the following algorithm (Eq S27) is utilised. Note that the variables N_r and k_r in Eq 27b are the modal number concentrations and uptake coefficients from Section S2.2 (Eqs S13 and S14) where $N_r = k_r = 0$ for $r = 3$ (the coarse mode).

Initial conditions (NH_4 available for NO_3 association):

$$T_{\text{NH}_4,\text{rem}} = \{\dot{\text{NH}}_4\} \quad [S27a]$$

For $r = 1, \dots, 3$ where r denotes the size mode (1=Aitken, 2=Accumulation, 3=Coarse), the moles of NH_4 associated with existing NO_3 equals:

$$\frac{\Delta\{\text{NO}_3\}_r}{\Delta t} = \frac{(\{\dot{\text{NO}}_3\} - \{\text{NO}_3\})}{\Delta t} \times \frac{N_r k_r}{\sum_r N_r k_r} \quad [S27b]$$

$$\{\text{NH}_4\dot{(\text{NO}_3)}\}_r = \text{Min} \left(T_{\text{NH}_4,\text{rem}}, \{\text{NO}_3\}_r + \frac{\Delta\{\text{NO}_3\}_r}{\Delta t} \times \Delta t \right) \quad [S27c]$$

$$T_{\text{NH}_4,\text{rem}} = \text{Max} (0, T_{\text{NH}_4,\text{rem}} - \{\text{NH}_4\dot{(\text{NO}_3)}\}_r) \quad [S27d]$$

Any remaining NH₄ (i.e. $T_{\text{NH}_4,\text{rem}} > 0$) is added to the Aitken mode.

S2.5 Disassociate existing ammonium nitrate if $T_A^* T_N \leq K_p$

In low concentrations of ammonia and nitrate, any existing ammonium nitrate aerosol disassociates, i.e. for modes $r = 1, \dots, 3$:

$$\frac{\Delta\{\text{NO}_3\}_r}{\Delta t} = -\frac{\{\text{NO}_3\}_r}{\Delta t} \quad [S28]$$

$$\{\text{NH}_4\dot{(\text{NO}_3)}\}_r = 0 \quad [S29]$$

S2.6 Calculate ammonium, ammonia and nitric acid tendencies

The ammonium tendency is the difference between the accumulated ammonium associated with sulphate and nitrate (as diagnosed above) and the input concentration of ammonium (Eq. S30). The nitric acid and ammonia tendencies are determined from the total nitrate and ammonium tendencies respectively (Eqs S31 and S32). All tendencies are the inverted to mass mixing ratios using $[X] = \frac{\{X\} M_X}{\rho_a}$ where M_X is the molar weight of substance X and ρ_a is the air density.

$$\frac{\Delta\{\text{NH}_4\}_r}{\Delta t} = \frac{\{\text{NH}_4\dot{(\text{NO}_3)}\}_r + \{\text{NH}_4\dot{(\text{SO}_4)}\}_r - \{\text{NH}_4\}_r}{\Delta t} \quad [S30]$$

$$\frac{\Delta\{\text{HNO}_3\}}{\Delta t} = -\sum_r \frac{\Delta\{\text{NO}_3\}_r}{\Delta t} \quad [S31]$$

$$\frac{\Delta\{\text{NH}_3\}}{\Delta t} = -\sum_r \frac{\Delta\{\text{NH}_4\}_r}{\Delta t} \quad [S32]$$

Checks are then made to ensure that HNO₃ and NH₃ mass concentrations are not less than zero at the end of the timestep and that number is also removed if, by dissociation (Section S2.4), total aerosol mass in a mode is zeroed. NH₃ and HNO₃ concentrations are updated at the end of the module.

S3 Coarse mode nitrate production

Nitric acid irreversibly condenses onto dust and sea-salt particles to form calcium nitrate and sodium nitrate respectively (Wang and Laskin, 2014). This reaction is slower than ammonium nitrate production, therefore numerically ammonium nitrate production is solved first. In the new UKCA-mode nitrate scheme, coarse nitrate is present in the accumulation and coarse soluble modes. Although functionality has been added for NO_3 uptake on CLASSIC 2 bin dust and the UKCA 2 insoluble mode dust scheme, only the CLASSIC 6-bin dust scheme (default in the N96 configuration at the time of writing) is described here.

S3.1 Calculate sea-salt number concentration and modal diameter

For every gridcell, the scheme first determines the dry median diameter ($D_{SS,r}$) for the accumulation and coarse soluble modes using Eq. S6 and then, under the assumption that sea-salt is externally mixed, the number concentration of sea-salt particles ($N_{SS,r}$ in m^{-3}) using the sea-salt mass mixing ratio ($M_{SS,r}$), modal width (σ_r), sea-salt density (ρ_{SS}), and air density (ρ_a):

$$N_{SS,r} = \frac{M_{SS,r}\rho_a}{\rho_{SS} \frac{\pi D_{SS,r}^3}{6} e^{4.5 \ln^2 \sigma_r}} \quad [\text{S33}]$$

S3.2 Calculate dust number concentration

The CLASSIC dust scheme utilises 6 size bins with mass logarithmically spaced with diameter. Only nitrate condensing on bins 2-6 (i.e. accumulation and coarse modes) is considered by the scheme. The volume median diameters ($D_{du,b}$ where index $b = 2, \dots, 6$ represents the bin) are 0.306, 0.966, 3.06, 9.66, and 30.6 μm . The dust number concentration per bin ($N_{du,b}$ in m^{-3}) is then determined from the dust mass mixing ratio ($M_{du,b}$), dust density (ρ_{du}), and air density (ρ_a):

$$N_{du,b} = \frac{M_{du,b}\rho_a}{\rho_{du} \frac{\pi D_{du,b}^3}{6}} \quad [\text{S34}]$$

S3.3 Determine the RH-dependent reactive uptake coefficient for dust and sea-salt

The reactive uptake coefficients for dust (γ_{du}) and sea-salt (γ_{SS}) are linearly interpolated from observationally constrained values with relative humidity [Fairlie *et al.*, 2010]. For the relative humidity values [15%, 30%, 70%, 80%], the γ_{du} values are [$1 \times 10^{-5}, 1 \times 10^{-4}, 6 \times 10^{-4}, 1.05 \times 10^{-3}$] and the γ_{SS} values are a factor of 100 greater at [$1 \times 10^{-3}, 1 \times 10^{-2}, 6 \times 10^{-2}, 1.05 \times 10^{-1}$], with no extrapolation at RH extrema (e.g. for RH > 80%, $\gamma(\text{RH}=80\%)$ is used).

S3.4 Determine the condensation rates for each sea-salt mode and dust bin

The first-order reactive uptake approximation (Fuchs and Sutugin, 1970; Schwartz, 1986) is used to determine condensation rates for each sea-salt mode and dust bin. The variables and constants are described in Section S2.2 (Eqs S9-S12) and the formulae again provided below.

$$D_g = \frac{3}{8A_c\rho_a d_a^2} \left[\frac{m_a R_a T}{2\pi} \times \frac{m_a + m_{HNO_3}}{m_{HNO_3}} \right]^{\frac{1}{2}} \quad [S35]$$

$$\lambda = \frac{3D_g}{v} = \frac{3D_g}{\sqrt{\frac{8R_a T}{\pi m_{HNO_3}}}} \quad [S36]$$

$$K_n = \frac{2\lambda}{D} \quad [S37]$$

$$k_{HNO_3} = \frac{2\pi D D_g}{1 + \frac{4K_n}{3\gamma} \times \left(1 - \frac{0.47\gamma}{1 + K_n}\right)} \quad [S38]$$

For sea-salt, the diameter (D) in Eqs S37 and S38 refers to the dry modal-mean diameter $D_{SS,r} e^{0.5 \ln^2 \sigma_r}$ multiplied by a relative-humidity dependent growth factor which ranges from 1.442 at RH=40% to 2.876 at RH=95%. For dust, the diameter (D) in Eqs S37 and S38 refers to the geometric mean diameter for each bin ($D_{du,b} = 0.28, 0.88, 2.8, 8.8, 28 \mu\text{m}$ for bins $b = 2, \dots, 6$ respectively). For sea-salt, $\gamma = \gamma_{SS}$ in Eq. S38, while for dust $\gamma = \gamma_{du}$.

The above algorithm delivers condensation rates for sea-salt ($k_{SS,r}$ for $r = 2,3$) and dust ($k_{du,b}$ for $b = 2, \dots, 6$). A Ca²⁺ limitation is now applied as in Hauglustaine *et al.* (2014), where the assumption is made that the Ca²⁺ content of dust is invariant at 5%.

S3.5 Map NO₃ uptake to accumulation and coarse soluble modes

The existing UKCA dust scheme utilises the CLASSIC dust emissions scheme and the mapping between the 6 CLASSIC bins and 2 UKCA modes is such that mass from bin 2 and half of bin 3 is mapped to the accumulation mode, while half of bin 3 and bins 4 and 5 are mapped to the coarse mode. The same mapping ratios are utilised here, with the exception that NO₃ uptake on bin 6 (super coarse dust sizes) is also mapped to the coarse soluble mode. The Nk rates (s⁻¹) defined in Section S2.2 (Eqs S13-S14) are determined for dust, sea-salt and then a composite.

$$Nk_{du,ACC} = N_{du,2} k_{du,2} + 0.5 \times N_{du,3} k_{du,3} \quad [S39]$$

$$Nk_{du,COA} = 0.5 \times N_{du,3} k_{du,3} + N_{du,4} k_{du,4} + N_{du,5} k_{du,5} + N_{du,6} k_{du,6} \quad [S40]$$

$$Nk_{SS,ACC} = N_{SS,1} k_{SS,1} \quad [S41]$$

$$Nk_{SS,COA} = N_{SS,2} k_{SS,2} \quad [S42]$$

$$Nk_{ACC} = Nk_{du,ACC} \times \frac{m_{Ca(NO_3)_2}}{m_{HNO_3}} + Nk_{SS,ACC} \times \frac{m_{NaNO_3}}{m_{HNO_3}} \quad [S43]$$

$$Nk_{COA} = Nk_{du,COA} \times \frac{m_{Ca(NO_3)_2}}{m_{HNO_3}} + Nk_{SS,COA} \times \frac{m_{NaNO_3}}{m_{HNO_3}} \quad [S44]$$

The molar masses in Eqs S43-S44 are $m_{HNO_3} = 0.063 \text{ kg mol}^{-1}$, $m_{NaNO_3} = 0.084 \text{ kg mol}^{-1}$ and $m_{Ca(NO_3)_2} = 0.164 \text{ kg mol}^{-1}$.

The total coarse mode nitrate mass tendency (using the notation of $[X]$ to represent mass concentration of substance X) is thus defined as:

$$\frac{\Delta[\text{coarseNO}_3]}{\Delta t} = (Nk_{ACC} + Nk_{COA}) \times [\text{HNO}_3] \quad [S45]$$

The nitric acid tendency is now defined as:

$$\frac{\Delta[\text{HNO}_3]}{\Delta t} = - \left(2 \times (Nk_{du,ACC} + Nk_{du,COA}) + (Nk_{SS,ACC} + Nk_{SS,COA}) \right) \times [\text{HNO}_3] \quad [S46]$$

Note that the dust rates are doubled in Eq. S46 to account for the 2 moles of HNO_3 required to produce one mole of $\text{Ca}(\text{NO}_3)_2$.

At this point in the algorithm $\frac{\Delta[\text{coarseNO}_3]}{\Delta t}$ is adjusted (if necessary) to ensure that HNO_3 concentrations after the timestep remain above or equal to zero. The modal nitrate mass tendency is then determined:

$$\frac{\Delta[\text{coarseNO}_3]_{ACC}}{\Delta t} = \frac{\Delta[\text{coarseNO}_3]}{\Delta t} \times \frac{Nk_{ACC}}{Nk_{ACC} + Nk_{COA}} \quad [S48]$$

$$\frac{\Delta[\text{coarseNO}_3]_{COA}}{\Delta t} = \frac{\Delta[\text{coarseNO}_3]}{\Delta t} \times \frac{Nk_{COA}}{Nk_{ACC} + Nk_{COA}} \quad [S49]$$

S3.6 Determine sea-salt mass tendency and tidy up

The modal sea-salt mass tendency (for modes $r = 2,3$ or the accumulation and coarse soluble modes) is now calculated. Currently, the chlorine ion (Cl^-) associated with Na^+ is simply removed from the aerosol and replaced with NO_3^- .

$$\frac{\Delta[\text{SS}]_r}{\Delta t} = -N_{SS,r} k_{SS,r} \times \frac{m_{\text{NaCl}}}{m_{HNO_3}} \times [\text{HNO}_3] \quad [S50]$$

At present, UKCA does not allow 2-way feedback with non-UKCA UM fields, thus the CLASSIC dust concentrations cannot be altered in a similar manner to sea-salt (Eq. S50). However, the UKCA-mode dust scheme will eventually supersede CLASSIC dust and will readily allow for such feedback.

As with the ammonium nitrate production scheme, NH_3 and HNO_3 mass concentrations are updated at the end of the module.

S4 UKCA-mode emissions inventory

This section provides further detail on UKCA-mode gas and aerosol emissions used in the MetUM integrations performed for this study. All emissions are based on constant year 2000 conditions. Global totals are provided in Table S4 for each species emitted via ancillary files.

Oceanic DMS emissions utilise the monthly sea-water concentration climatology of Kettle *et al.* (1999) coupled to the sea-air exchange scheme of Liss and Merlivat (1986), while land DMS emissions follow Spiro *et al.* (1992). On an annual mean basis, total DMS emissions amount to 29 Tg[S] yr⁻¹. Anthropogenic SO₂ emissions from the CEDS inventory are divided into ‘chimney-top’ high-level emissions at 0.5 km altitude and surface-level emissions [Sellier *et al.*, 2020]. A fixed fraction of SO₂ emissions (2.5%) are emitted as primary SO₄ aerosol following AeroCom recommendations (Dentener *et al.*, 2006). Natural SO₂ emissions from passive volcanic eruptions also follow Dentener *et al.* (2006). In total, global SO₂ emissions amount to 140 Tg yr⁻¹, of which 74% is anthropogenic.

Primary sea-salt emissions are driven by the simulated surface-wind speed at each time-step using the Gong-Monahan sea-spray source function (Gong, 2003). On an annual mean basis, total sea-salt emissions amount to ~4040 Tg yr⁻¹ (of which 4016 Tg yr⁻¹ is in the coarse mode), while primary SO₄ aerosol emissions amount to 1.6 Tg[S] yr⁻¹. Primary marine OM emissions follow the parameterisation of Gantt *et al.* (2011) based on surface-wind speed and chlorophyll, while secondary OM is produced from the atmospheric oxidation of monoterpenes from land-sources (Mulcahy *et al.*, 2020). In total, global BC emissions amount to 9.8 Tg[C] yr⁻¹ while OM production amounts to 54 Tg[C] yr⁻¹. Secondary SO₄ aerosol is produced in the atmosphere via condensational uptake of H₂SO₄ aerosol on pre-existing aerosol (25 Tg[S] yr⁻¹ on a global basis from these simulations), aqueous phase SO₂ oxidation by H₂O₂ and O₃ in cloud droplets (23.7 Tg[S] yr⁻¹), and by binary homogeneous nucleation (0.4 Tg[S] yr⁻¹).

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| Reaction type | Reaction | Net global tropospheric HNO ₃ production (Tg[N] yr ⁻¹) | |
|-------------------|--|---|-------|
| | | FAST | SLOW |
| Bimolecular | $N_2O_5 + H_2O \rightarrow 2HNO_3$ | 0.78 | 0.84 |
| Bimolecular | $NO_3 + EtCHO \rightarrow HNO_3 + EtCO_3$ | 0.007 | 0.007 |
| Bimolecular | $NO_3 + HCHO \rightarrow HNO_3 + HO_2 + CO$ | 0.13 | 0.13 |
| Bimolecular | $NO_3 + MGLY \rightarrow HNO_3 + MeCO_3 + CO$ | 0.029 | 0.031 |
| Bimolecular | $NO_3 + Me_2CO \rightarrow HNO_3 + MeCOCH_2OO$ | 0.017 | 0.018 |
| Bimolecular | $NO_3 + MeCHO \rightarrow HNO_3 + MeCO_3$ | 0.061 | 0.064 |
| Bimolecular | $HNO_3 + OH \rightarrow NO_3 + H_2O$ | -6.2 | -6.3 |
| Bimolecular | $DMS + NO_3 \rightarrow SO_2 + HNO_3 + MeOO$ | 2.3 | 2.3 |
| Termolecular | $OH + NO_2 \rightarrow HNO_3 + m$ | 31.9 | 32.3 |
| Photolysis | $HNO_3 + h\nu \rightarrow NO_2 + OH$ | -3.4 | -3.4 |
| Het. PSC/NAT | $ClONO_2 + H_2O \rightarrow HNO_3 + HCl$ | 0.86 | 0.84 |
| Het. PSC/NAT | $ClONO_2 + HCl \rightarrow HNO_3 + 2Cl$ | 0.11 | 0.11 |
| Het. PSC/NAT | $N_2O_5 + HCl \rightarrow HNO_3 + NO_2 + Cl$ | 0.001 | 0.001 |
| Het. PSC/NAT/Aero | $N_2O_5 + H_2O \rightarrow 2HNO_3$ | 17.6 | 17.3 |
| Total | | 44.15 | 44.18 |

Table S1: Chemical reactions in UKCA StratTrop1.0 that involve nitric acid (HNO₃), and net annual and global tropospheric HNO₃ production (Tg[N] yr⁻¹) in the FAST and SLOW simulations. See Archibald *et al.* (2020) for details of the chemical species

| Wavelength λ (μm) | $n(\lambda)$ | $k(\lambda)$ |
|---|--------------|--------------|
| 0.1051 | 1.427 | 0.3985 |
| 0.106 | 1.439 | 0.3975 |
| 0.1069 | 1.4535 | 0.3935 |
| 0.1078 | 1.4635 | 0.3875 |
| 0.1088 | 1.4715 | 0.376 |
| 0.1097 | 1.4775 | 0.363 |
| 0.1107 | 1.4825 | 0.347 |
| 0.1117 | 1.483 | 0.3345 |
| 0.1127 | 1.4795 | 0.3305 |
| 0.1137 | 1.4795 | 0.3395 |
| 0.1148 | 1.4765 | 0.355 |
| 0.1159 | 1.484 | 0.346 |
| 0.117 | 1.5175 | 0.3495 |
| 0.1181 | 1.5565 | 0.3565 |
| 0.1192 | 1.602 | 0.345 |
| 0.1204 | 1.6295 | 0.30255 |
| 0.1216 | 1.642 | 0.2332 |
| 0.1228 | 1.6405 | 0.1898 |
| 0.124 | 1.6165 | 0.16375 |
| 0.1252 | 1.5955 | 0.154276 |
| 0.1265 | 1.577 | 0.144013 |
| 0.1278 | 1.5685 | 0.13375 |
| 0.1292 | 1.5585 | 0.1271 |
| 0.1305 | 1.5545 | 0.11375 |
| 0.1319 | 1.5495 | 0.1036 |
| 0.1333 | 1.54 | 0.0901 |
| 0.1348 | 1.5295 | 0.0714 |
| 0.1362 | 1.513 | 0.058895 |
| 0.1378 | 1.492 | 0.045915 |
| 0.1393 | 1.472 | 0.040195 |
| 0.1409 | 1.46 | 0.0458 |
| 0.1425 | 1.4515 | 0.04975 |
| 0.1442 | 1.4495 | 0.0464 |

| | | |
|--------|--------|----------|
| 0.1459 | 1.446 | 0.03965 |
| 0.1476 | 1.4405 | 0.033905 |
| 0.1494 | 1.4265 | 0.02473 |
| 0.1512 | 1.412 | 0.02157 |
| 0.1531 | 1.3955 | 0.018235 |
| 0.155 | 1.379 | 0.0149 |
| 0.1569 | 1.3545 | 0.015265 |
| 0.159 | 1.33 | 0.01768 |
| 0.161 | 1.2995 | 0.02548 |
| 0.1631 | 1.2735 | 0.03367 |
| 0.1653 | 1.2465 | 0.04965 |
| 0.1675 | 1.218 | 0.07143 |
| 0.1698 | 1.19 | 0.09385 |
| 0.1722 | 1.156 | 0.13415 |
| 0.1746 | 1.1455 | 0.17445 |
| 0.1771 | 1.137 | 0.2282 |
| 0.1797 | 1.144 | 0.299 |
| 0.1823 | 1.167 | 0.3565 |
| 0.1851 | 1.199 | 0.4045 |
| 0.1879 | 1.239 | 0.4375 |
| 0.1907 | 1.3015 | 0.46385 |
| 0.1937 | 1.362 | 0.471 |
| 0.1968 | 1.425 | 0.4946 |
| 0.2 | 1.4575 | 0.46865 |
| 0.2033 | 1.5055 | 0.466 |
| 0.2066 | 1.549 | 0.4565 |
| 0.2101 | 1.5985 | 0.4487 |
| 0.2138 | 1.6685 | 0.4281 |
| 0.2175 | 1.7375 | 0.3837 |
| 0.2214 | 1.787 | 0.29235 |
| 0.2254 | 1.804 | 0.170785 |
| 0.2296 | 1.766 | 0.174 |
| 0.2339 | 1.712 | 0.0806 |
| 0.2384 | 1.6615 | 0.0358 |
| 0.2431 | 1.624 | 0.0212 |

| | | |
|--------|----------|----------|
| 0.248 | 1.5925 | 0.0109 |
| 0.253 | 1.572 | 0.0028 |
| 0.2583 | 1.55 | 0.007121 |
| 0.2638 | 1.5365 | 0.003526 |
| 0.2695 | 1.52 | 0.00239 |
| 0.2755 | 1.5095 | 0.001996 |
| 0.2818 | 1.502 | 0.001926 |
| 0.2883 | 1.498 | 0.001941 |
| 0.2952 | 1.492 | 0.002063 |
| 0.3024 | 1.489 | 0.00237 |
| 0.31 | 1.4875 | 0.002933 |
| 0.32 | 1.488986 | 0.004039 |
| 0.33 | 1.490471 | 0.005925 |
| 0.34 | 1.491957 | 0.00797 |
| 0.35 | 1.493442 | 0.009556 |
| 0.36 | 1.494928 | 0.010146 |
| 0.37 | 1.496413 | 0.008971 |
| 0.38 | 1.497899 | 0.005179 |
| 0.39 | 1.499384 | 0.002114 |
| 0.4 | 1.50087 | 0.000979 |
| 0.405 | 1.49953 | 0.00069 |
| 0.41 | 1.49826 | 0.000466 |
| 0.415 | 1.49702 | 0.000266 |
| 0.42 | 1.49583 | 9.22E-05 |
| 0.44 | 1.49148 | 1.00E-08 |
| 0.46 | 1.48766 | 1.00E-08 |
| 0.48 | 1.48432 | 1.00E-08 |
| 0.5 | 1.48139 | 1.00E-08 |
| 0.52 | 1.47876 | 1.00E-08 |
| 0.535 | 1.47699 | 1.00E-08 |
| 0.5876 | 1.465193 | 1.00E-08 |
| 0.656 | 1.45655 | 1.00E-08 |
| 0.706 | 1.456074 | 1.48E-08 |
| 0.8 | 1.457231 | 1.06E-07 |
| 1 | 1.459694 | 3.15E-06 |

| | | |
|------|----------|----------|
| 1.07 | 1.459327 | 1.57E-06 |
| 1.15 | 1.458942 | 8.06E-06 |
| 1.2 | 1.458725 | 1.08E-05 |
| 1.3 | 1.458331 | 1.34E-05 |
| 1.4 | 1.457978 | 0.000151 |
| 1.5 | 1.457524 | 0.000199 |
| 1.58 | 1.457128 | 0.00011 |
| 1.6 | 1.45703 | 0.000958 |
| 1.7 | 1.456525 | 6.82E-05 |
| 1.8 | 1.456 | 0.000123 |
| 1.9 | 1.455475 | 0.00126 |
| 2 | 1.45495 | 0.00108 |
| 2.17 | 1.453634 | 0.00275 |
| 2.22 | 1.453247 | 0.00567 |
| 2.27 | 1.45286 | 0.00191 |
| 2.3 | 1.452627 | 0.00588 |
| 2.33 | 1.452395 | 0.00196 |
| 2.38 | 1.452008 | 0.001 |
| 2.44 | 1.451543 | 0.00309 |
| 2.5 | 1.451079 | 0.00317 |
| 2.56 | 1.450446 | 1.00E-08 |
| 2.63 | 1.449707 | 0.000884 |
| 2.7 | 1.448969 | 0.00113 |
| 2.78 | 1.448125 | 0.00951 |
| 2.86 | 1.447279 | 0.024 |
| 2.94 | 1.446379 | 0.0553 |
| 3.03 | 1.445367 | 0.209 |
| 3.12 | 1.444355 | 0.459 |
| 3.2 | 1.443455 | 0.507 |
| 3.25 | 1.442893 | 0.477 |
| 3.27 | 1.442668 | 0.418 |
| 3.32 | 1.442105 | 0.425 |
| 3.36 | 1.441485 | 0.416 |
| 3.37 | 1.44131 | 0.358 |
| 3.39 | 1.440959 | 0.311 |

| | | |
|------|----------|--------|
| 3.43 | 1.440257 | 0.28 |
| 3.45 | 1.439906 | 0.274 |
| 3.47 | 1.439555 | 0.239 |
| 3.51 | 1.438853 | 0.209 |
| 3.55 | 1.438151 | 0.17 |
| 3.57 | 1.4378 | 0.15 |
| 3.61 | 1.437115 | 0.123 |
| 3.64 | 1.436602 | 0.101 |
| 3.7 | 1.435575 | 0.0658 |
| 3.77 | 1.434378 | 0.0516 |
| 3.88 | 1.432311 | 0.0343 |
| 3.97 | 1.430281 | 0.0278 |
| 3.98 | 1.430056 | 0.0279 |
| 4.03 | 1.428928 | 0.0357 |
| 4.07 | 1.428026 | 0.0341 |
| 4.14 | 1.426448 | 0.0566 |
| 4.17 | 1.425757 | 0.074 |
| 4.18 | 1.425485 | 0.104 |
| 4.2 | 1.424941 | 0.0575 |
| 4.26 | 1.423309 | 0.0298 |
| 4.31 | 1.421949 | 0.0185 |
| 4.39 | 1.419773 | 0.0112 |
| 4.55 | 1.415361 | 0.0108 |
| 4.78 | 1.406344 | 0.0206 |
| 4.85 | 1.4036 | 0.0274 |
| 4.9 | 1.40164 | 0.0211 |
| 4.95 | 1.399679 | 0.0257 |
| 5.11 | 1.391782 | 0.0433 |
| 5.15 | 1.389623 | 0.0437 |
| 5.26 | 1.383685 | 0.0393 |
| 5.46 | 1.369257 | 0.0484 |
| 5.56 | 1.361898 | 0.0613 |
| 5.62 | 1.355779 | 0.062 |
| 5.65 | 1.35272 | 0.0773 |
| 5.68 | 1.349661 | 0.29 |

| | | |
|-------|----------|--------|
| 5.73 | 1.344563 | 0.108 |
| 5.8 | 1.337425 | 0.141 |
| 5.882 | 1.329063 | 0.0044 |
| 6.061 | 1.29893 | 0.0063 |
| 6.25 | 1.258789 | 0.0098 |
| 6.452 | 1.188639 | 0.018 |
| 6.667 | 1.048479 | 0.043 |
| 6.711 | 0.993446 | 0.058 |
| 6.757 | 0.918412 | 0.086 |
| 6.803 | 0.808378 | 0.18 |
| 6.812 | 0.783371 | 0.24 |
| 6.821 | 0.763365 | 0.28 |
| 6.831 | 0.748357 | 0.34 |
| 6.84 | 0.73835 | 0.4 |
| 6.849 | 0.733344 | 0.46 |
| 6.859 | 0.728336 | 0.51 |
| 6.868 | 0.72333 | 0.56 |
| 6.873 | 0.723326 | 0.59 |
| 6.878 | 0.723322 | 0.61 |
| 6.882 | 0.723319 | 0.64 |
| 6.887 | 0.728315 | 0.66 |
| 6.892 | 0.733312 | 0.68 |
| 6.897 | 0.738308 | 0.71 |
| 6.901 | 0.753305 | 0.73 |
| 6.906 | 0.768301 | 0.76 |
| 6.911 | 0.768298 | 0.78 |
| 6.916 | 0.753294 | 0.8 |
| 6.92 | 0.738291 | 0.82 |
| 6.925 | 0.728287 | 0.84 |
| 6.93 | 0.723283 | 0.86 |
| 6.935 | 0.72328 | 0.89 |
| 6.944 | 0.718273 | 0.93 |
| 6.969 | 0.718254 | 1.1 |
| 6.993 | 0.718237 | 1.2 |
| 7.018 | 0.718218 | 1.3 |

| | | |
|-------|----------|--------|
| 7.042 | 0.7232 | 1.4 |
| 7.067 | 0.728182 | 1.5 |
| 7.092 | 0.733163 | 1.7 |
| 7.117 | 0.743144 | 1.8 |
| 7.143 | 0.753125 | 2 |
| 7.194 | 0.783087 | 2.4 |
| 7.246 | 0.853048 | 3 |
| 7.299 | 1.048009 | 4.1 |
| 7.326 | 1.332989 | 5 |
| 7.353 | 2.147969 | 6.1 |
| 7.38 | 3.762949 | 4.8 |
| 7.407 | 3.697929 | 2 |
| 7.435 | 3.212908 | 0.95 |
| 7.463 | 2.887887 | 0.56 |
| 7.519 | 2.507845 | 0.27 |
| 7.576 | 2.297803 | 0.16 |
| 7.634 | 2.15776 | 0.11 |
| 7.692 | 2.062717 | 0.078 |
| 7.813 | 1.932627 | 0.047 |
| 7.937 | 1.852534 | 0.031 |
| 8.065 | 1.797439 | 0.023 |
| 8.197 | 1.752341 | 0.017 |
| 8.333 | 1.72224 | 0.013 |
| 9.091 | 1.636676 | 0.0053 |
| 10 | 1.44 | 0.0126 |
| 11.11 | 1.4345 | 0.0142 |
| 12.59 | 1.427 | 0.0163 |
| 14.29 | 1.416 | 0.0193 |
| 16.67 | 1.3975 | 0.0238 |
| 20 | 1.363 | 0.0315 |
| 20.83 | 1.353 | 0.0338 |
| 21.74 | 1.341 | 0.0365 |
| 22.73 | 1.3265 | 0.0398 |
| 23.81 | 1.3095 | 0.0437 |
| 25 | 1.288 | 0.0487 |

| | | |
|-------|--------|--------|
| 25.64 | 1.275 | 0.0518 |
| 26.32 | 1.2605 | 0.0552 |
| 27.03 | 1.2445 | 0.0593 |
| 27.78 | 1.226 | 0.064 |
| 28.57 | 1.2045 | 0.0697 |
| 29.41 | 1.1795 | 0.0766 |
| 30.3 | 1.15 | 0.0852 |
| 31.25 | 1.1145 | 0.0962 |
| 32.26 | 1.0705 | 0.111 |
| 33.33 | 1.0165 | 0.131 |
| 34.48 | 0.947 | 0.16 |
| 35.71 | 0.8575 | 0.206 |
| 37.04 | 0.742 | 0.288 |
| 38.46 | 0.6105 | 0.456 |
| 40 | 0.521 | 0.758 |
| 41.67 | 0.522 | 1.15 |
| 43.48 | 0.654 | 1.65 |
| 44.44 | 0.8375 | 1.97 |
| 45.45 | 1.2745 | 2.35 |
| 45.98 | 1.746 | 2.56 |
| 46.51 | 2.4935 | 2.75 |
| 47.06 | 3.1275 | 2.83 |
| 47.62 | 3.3195 | 2.68 |
| 48.19 | 3.3275 | 2.24 |
| 48.78 | 3.218 | 1.71 |
| 50 | 2.8475 | 1.01 |
| 51.28 | 2.514 | 0.763 |
| 52.63 | 2.2655 | 0.778 |
| 54.05 | 2.1215 | 0.987 |
| 55.56 | 2.1505 | 1.25 |
| 57.14 | 2.316 | 1.21 |
| 58.82 | 2.3805 | 0.834 |
| 60.61 | 2.3115 | 0.514 |
| 62.5 | 2.2105 | 0.333 |
| 66.67 | 2.033 | 0.179 |

| | | |
|-------|--------|-------|
| 71.43 | 1.89 | 0.126 |
| 76.92 | 1.7575 | 0.112 |
| 80 | 1.6885 | 0.115 |
| 83.33 | 1.611 | 0.127 |
| 86.96 | 1.5205 | 0.151 |
| 90.91 | 1.4065 | 0.201 |
| 100 | 1.037 | 0.625 |
| 102.6 | 0.9375 | 0.952 |
| 105.3 | 0.8955 | 1.39 |
| 108.1 | 0.9425 | 1.9 |
| 111.1 | 1.1535 | 2.38 |
| 114.3 | 1.61 | 2.4 |
| 117.6 | 1.9265 | 1.62 |
| 121.2 | 1.897 | 0.913 |
| 125 | 1.7875 | 0.548 |
| 133.3 | 1.6265 | 0.262 |
| 142.9 | 1.55 | 0.162 |
| 153.8 | 1.5385 | 0.118 |

| | | |
|-------|--------|---------|
| 166.7 | 1.6295 | 0.0958 |
| 173.9 | 1.782 | 0.0888 |
| 181.8 | 2.183 | 0.0836 |
| 190.5 | 3.437 | 0.0798 |
| 200 | 4.6385 | 0.0769 |
| 210.5 | 4.1645 | 0.0748 |
| 222.2 | 3.733 | 0.0733 |
| 250 | 3.263 | 0.0714 |
| 285.7 | 3.0235 | 0.0704 |
| 333.3 | 2.8815 | 0.0694 |
| 400 | 2.791 | 0.0674 |
| 500 | 2.7325 | 0.0631 |
| 578 | 2.735 | 0.0162 |
| 666.7 | 2.696 | 0.0551 |
| 751.9 | 2.715 | 0.0102 |
| 1000 | 2.6755 | 0.0418 |
| 1075 | 2.625 | 0.00618 |

Table S2: Refractive indices for sodium nitrate (NaNO_3). See paper for references.

| Wavelength λ (μm) | n(λ) | k(λ) |
|---|--------------------------------|--------------------------------|
| 0.1 | 1.611 | 1.00E-08 |
| 0.3 | 1.611 | 1.00E-08 |
| 0.41 | 1.611 | 1.00E-08 |
| 0.54 | 1.611 | 1.00E-08 |
| 0.59 | 1.611 | 1.00E-08 |
| 0.66 | 1.611 | 1.00E-08 |
| 0.71 | 1.611 | 1.48E-08 |
| 0.8 | 1.611 | 1.06E-07 |
| 1 | 1.611 | 3.15E-06 |
| 1.07 | 1.611 | 1.57E-06 |
| 1.15 | 1.611 | 8.06E-06 |
| 1.2 | 1.611 | 1.08E-05 |
| 1.3 | 1.611 | 1.34E-05 |
| 1.4 | 1.611 | 0.000151 |
| 1.5 | 1.611 | 0.000199 |
| 1.58 | 1.611 | 0.00011 |
| 1.6 | 1.611 | 0.000958 |
| 1.7 | 1.611 | 6.82E-05 |
| 1.8 | 1.611 | 0.000123 |
| 1.9 | 1.611 | 0.00126 |
| 2 | 1.611 | 0.00108 |
| 2.17 | 1.56 | 0.00275 |
| 2.22 | 1.55 | 0.00567 |
| 2.27 | 1.55 | 0.00191 |
| 2.3 | 1.55 | 0.00588 |
| 2.33 | 1.55 | 0.00196 |
| 2.38 | 1.54 | 0.001 |
| 2.44 | 1.53 | 0.00309 |
| 2.5 | 1.53 | 0.00317 |
| 2.56 | 1.51 | 1.00E-08 |
| 2.63 | 1.5 | 0.000884 |
| 2.7 | 1.48 | 0.00113 |
| 2.78 | 1.44 | 0.00951 |

| | | |
|------|------|--------|
| 2.86 | 1.4 | 0.024 |
| 2.94 | 1.31 | 0.0553 |
| 3.03 | 1.25 | 0.209 |
| 3.12 | 1.35 | 0.459 |
| 3.2 | 1.55 | 0.507 |
| 3.25 | 1.66 | 0.477 |
| 3.27 | 1.69 | 0.418 |
| 3.32 | 1.72 | 0.425 |
| 3.36 | 1.81 | 0.416 |
| 3.37 | 1.83 | 0.358 |
| 3.39 | 1.82 | 0.311 |
| 3.43 | 1.81 | 0.28 |
| 3.45 | 1.82 | 0.274 |
| 3.47 | 1.83 | 0.239 |
| 3.51 | 1.83 | 0.209 |
| 3.55 | 1.83 | 0.17 |
| 3.57 | 1.82 | 0.15 |
| 3.61 | 1.82 | 0.123 |
| 3.64 | 1.81 | 0.101 |
| 3.7 | 1.78 | 0.0658 |
| 3.77 | 1.75 | 0.0516 |
| 3.88 | 1.71 | 0.0343 |
| 3.97 | 1.68 | 0.0278 |
| 3.98 | 1.68 | 0.0279 |
| 4.03 | 1.67 | 0.0357 |
| 4.07 | 1.66 | 0.0341 |
| 4.14 | 1.64 | 0.0566 |
| 4.17 | 1.64 | 0.074 |
| 4.18 | 1.64 | 0.104 |
| 4.20 | 1.7 | 0.0575 |
| 4.26 | 1.68 | 0.0298 |
| 4.31 | 1.66 | 0.0185 |
| 4.39 | 1.65 | 0.0112 |
| 4.55 | 1.61 | 0.0108 |
| 4.78 | 1.57 | 0.0206 |

| | | |
|------|------|--------|
| 4.85 | 1.57 | 0.0274 |
| 4.9 | 1.56 | 0.0211 |
| 4.95 | 1.55 | 0.0257 |
| 5.11 | 1.53 | 0.0433 |
| 5.15 | 1.53 | 0.0437 |
| 5.26 | 1.51 | 0.0393 |
| 5.46 | 1.46 | 0.0484 |
| 5.56 | 1.44 | 0.0613 |
| 5.62 | 1.38 | 0.062 |
| 5.65 | 1.33 | 0.0773 |
| 5.68 | 1.35 | 0.29 |
| 5.73 | 1.49 | 0.108 |
| 5.80 | 1.45 | 0.141 |
| 5.85 | 1.43 | 0.145 |
| 5.92 | 1.44 | 0.144 |
| 6.02 | 1.42 | 0.13 |
| 6.13 | 1.39 | 0.129 |
| 6.25 | 1.33 | 0.134 |
| 6.37 | 1.27 | 0.155 |
| 6.49 | 1.17 | 0.2 |
| 6.67 | 1 | 0.34 |
| 6.76 | 0.85 | 0.465 |
| 6.85 | 0.84 | 0.849 |
| 6.99 | 1.05 | 1.17 |
| 7.04 | 1.16 | 1.26 |
| 7.14 | 1.5 | 1.67 |
| 7.17 | 1.64 | 1.68 |
| 7.19 | 1.77 | 1.69 |
| 7.34 | 2.38 | 1.31 |
| 7.46 | 2.56 | 0.925 |
| 7.69 | 2.45 | 0.457 |
| 7.81 | 2.37 | 0.34 |
| 8.20 | 2.18 | 0.194 |
| 8.47 | 2.11 | 0.159 |
| 8.62 | 2.09 | 0.14 |

| | | |
|-------|------|----------|
| 8.93 | 2.04 | 0.122 |
| 9.1 | 2.03 | 0.115 |
| 9.35 | 2.02 | 0.105 |
| 9.45 | 2.02 | 0.0749 |
| 9.6 | 2.03 | 0.0894 |
| 9.62 | 2.02 | 0.0717 |
| 9.80 | 1.98 | 0.0642 |
| 10 | 1.95 | 0.0636 |
| 10.2 | 1.95 | 0.0649 |
| 10.42 | 1.94 | 0.0616 |
| 10.6 | 1.94 | 0.0484 |
| 10.66 | 1.93 | 0.0459 |
| 11.11 | 1.91 | 0.038 |
| 11.36 | 1.88 | 0.027 |
| 11.63 | 1.85 | 0.0277 |
| 11.9 | 1.62 | 0.0512 |
| 12.05 | 1.57 | 0.259 |
| 12.16 | 2.72 | 0.966 |
| 12.2 | 2.54 | 0.167 |
| 12.24 | 2.46 | 0.0313 |
| 12.5 | 1.98 | 0.00525 |
| 13.51 | 1.89 | 0.0287 |
| 13.74 | 1.85 | 0.0351 |
| 13.89 | 1.87 | 0.0721 |
| 13.95 | 1.88 | 0.104 |
| 14.08 | 1.94 | 0.0238 |
| 14.29 | 1.9 | 0.0181 |
| 14.71 | 1.89 | 1.00E-08 |
| 16.13 | 1.85 | 0.0136 |
| 17.24 | 1.84 | 0.0145 |
| 19.23 | 1.81 | 0.0244 |
| 20 | 1.82 | 0.0424 |
| 22.2 | 1.85 | 0.0136 |
| 25 | 1.85 | 0.0136 |
| 28.6 | 1.85 | 0.0136 |

| | | |
|------|------|--------|
| 33.3 | 1.85 | 0.0136 |
| 40 | 1.85 | 0.0136 |

| | | |
|--------|---|----------|
| 100000 | 1 | 1.00E-08 |
|--------|---|----------|

Table S3: Refractive indices for ammonium nitrate (NH_4NO_3). See paper for references.

| Species | Source | Time | Domain | Units | Emissions |
|-------------------------------|--------------------|-------------|---------------|------------------------|------------------|
| BC | Biofuel | Monthly | Surface | Tg[C] yr ⁻¹ | 3.1 |
| BC | Fossil fuel | Monthly | Surface | Tg[C] yr ⁻¹ | 2.9 |
| BC | Biomass high level | Monthly | Surface | Tg[C] yr ⁻¹ | 1.2 |
| BC | Biomass surface | Monthly | Surface | Tg[C] yr ⁻¹ | 2.6 |
| C ₂ H ₆ | Biogenic | Monthly | Surface | Tg yr ⁻¹ | 31.1 |
| C ₂ H ₆ | Oceanic | Monthly | Surface | Tg yr ⁻¹ | 2.4 |
| C ₂ H ₆ | Anthropogenic | Monthly | Surface | Tg yr ⁻¹ | 13.8 |
| C ₂ H ₆ | Biomass | Monthly | Surface | Tg yr ⁻¹ | 10.7 |
| C ₃ H ₈ | Biogenic | Monthly | Surface | Tg yr ⁻¹ | 15.6 |
| C ₃ H ₈ | Oceanic | Monthly | Surface | Tg yr ⁻¹ | 2.8 |
| C ₃ H ₈ | Anthropogenic | Monthly | Surface | Tg yr ⁻¹ | 9.2 |
| C ₃ H ₈ | Biomass | Monthly | Surface | Tg yr ⁻¹ | 5.2 |
| C ₅ H ₈ | Biogenic | Monthly | Surface | Tg yr ⁻¹ | 568 |
| CH ₄ | Biomass | Monthly | Surface | Tg yr ⁻¹ | 18.8 |
| CO | Biogenic | Monthly | Surface | Tg yr ⁻¹ | 88.6 |
| CO | Oceanic | Monthly | Surface | Tg yr ⁻¹ | 19.6 |
| CO | Anthropogenic | Monthly | Surface | Tg yr ⁻¹ | 594 |
| CO | Biomass | Monthly | Surface | Tg yr ⁻¹ | 401 |
| DMS | Land | Monthly | Surface | Tg[S] yr ⁻¹ | 0.9 |
| DMS | Biomass | Monthly | Surface | Tg[S] yr ⁻¹ | 0.01 |
| HCHO | Biogenic | Monthly | Surface | Tg yr ⁻¹ | 4.6 |
| HCHO | Anthropogenic | Monthly | Surface | Tg yr ⁻¹ | 2.3 |
| HCHO | Biomass | Monthly | Surface | Tg yr ⁻¹ | 5.5 |
| Me ₂ CO | Biogenic | Monthly | Surface | Tg yr ⁻¹ | 37.4 |
| Me ₂ CO | Anthropogenic | Monthly | Surface | Tg yr ⁻¹ | 2.6 |
| Me ₂ CO | Biomass | Monthly | Surface | Tg yr ⁻¹ | 3.5 |
| MeCHO | Biogenic | Monthly | Surface | Tg yr ⁻¹ | 21.5 |
| MeCHO | Anthropogenic | Monthly | Surface | Tg yr ⁻¹ | 1.7 |
| MeCHO | Biomass | Monthly | Surface | Tg yr ⁻¹ | 8.2 |
| Monoterpene | Biogenic | Monthly | Surface | Tg yr ⁻¹ | 94.7 |
| NH ₃ | Oceanic | Single time | Surface | Tg yr ⁻¹ | 9.8 |
| NH ₃ | Anthropogenic | Monthly | Surface | Tg yr ⁻¹ | 50.5 |

| | | | | | |
|-----------------|--------------------------|-------------|---------|-------------------------|------|
| NH ₃ | Biomass | Monthly | Surface | Tg yr ⁻¹ | 4.7 |
| NO _x | Soil | Monthly | Surface | Tg[NO] yr ⁻¹ | 11.8 |
| NO _x | Anthropogenic | Monthly | Surface | Tg[NO] yr ⁻¹ | 76.8 |
| NO _x | Biomass | Monthly | Surface | Tg[NO] yr ⁻¹ | 16.1 |
| NO _x | Aircraft | Monthly | Levels | Tg[NO] yr ⁻¹ | 1.6 |
| NVOC | Biogenic | Monthly | Surface | Tg[C] yr ⁻¹ | 225 |
| NVOC | Anthropogenic | Monthly | Surface | Tg[C] yr ⁻¹ | 6 |
| NVOC | Biomass | Monthly | Surface | Tg[C] yr ⁻¹ | 16.6 |
| OC | Biofuel | Monthly | Surface | Tg[C] yr ⁻¹ | 9.3 |
| OC | Fossil fuel | Monthly | Surface | Tg[C] yr ⁻¹ | 5.3 |
| OC | Biomass high level | Monthly | Surface | Tg[C] yr ⁻¹ | 15.2 |
| OC | Biomass surface | Monthly | Surface | Tg[C] yr ⁻¹ | 18.8 |
| SO ₂ | Natural | Single time | Levels | Tg yr ⁻¹ | 28.8 |
| SO ₂ | Anthropogenic high level | Monthly | Surface | Tg yr ⁻¹ | 75 |
| SO ₂ | Anthropogenic surface | Monthly | Surface | Tg yr ⁻¹ | 36.6 |

Table S4: Prescribed UKCA aerosol and gas emissions for the MetUM simulations with temporal and domain specifications and global and annual total emissions in given units

| | | | | FAST | INSTANT |
|----------------|----------|---------------------------|------------------------|-------------|----------------|
| HNO_3 | Source | Gas phase | Tg[N] yr ⁻¹ | 35.2 | 35.2 |
| | | Aerosol phase | | 18.6 | 18.6 |
| | | Total | | 53.8 | 53.7 |
| | Loss | Gas phase | Tg[N] yr ⁻¹ | 9.7 | 9.6 |
| | | Fine nitrate | | 6.4 | 6.5 |
| | | Coarse nitrate | | 16.6 | 16.5 |
| | | Dry deposition | | 6 | 6 |
| | | Wet deposition | | 14.9 | 14.7 |
| | | Total | | 53.4 | 53.3 |
| | | Wet fraction | % | 71.4 | 71.2 |
| | Burden | | Tg[N] | 0.48 | 0.48 |
| | | | days | 3.2 | 3.2 |
| NO_3 | Source | Fine nitrate | Tg[N] yr ⁻¹ | 6.3 | 6.5 |
| | | Coarse nitrate | | 16.6 | 16.5 |
| | | Total | | 22.9 | 23 |
| | Loss | Dry deposition | Tg[N] yr ⁻¹ | 8.9 | 8.9 |
| | | Wet deposition | | 14.3 | 14.4 |
| | | Total | | 21.3 | 23.2 |
| | | Wet fraction | % | 61.7 | 61.9 |
| | Burden | Fine nitrate | Tg[N] | 0.11 | 0.11 |
| | | Coarse nitrate | | 0.09 | 0.09 |
| | | Total | | 0.2 | 0.21 |
| | Lifetime | Fine nitrate | days | 6.2 | 6.2 |
| | | Coarse nitrate | | 2 | 2 |
| | | Total | | 3.2 | 3.2 |
| NH_3 | Source | Emissions | Tg[N] yr ⁻¹ | 53.5 | 53.5 |
| | | Gas phase | Tg[N] yr ⁻¹ | - | - |
| | Loss | NH ₄ formation | | 30.4 | 30.4 |
| | | Dry deposition | | 17.4 | 17.3 |
| | | Wet deposition | | 5.7 | 5.6 |
| | | Total | | 53.4 | 53.4 |
| | | Wet fraction | % | 24.6 | 24.5 |
| | Burden | | Tg[N] | 0.04 | 0.04 |
| | | | days | 0.28 | 0.27 |
| NH_4 | Source | NH_3 conversion | Tg[N] yr ⁻¹ | 30.4 | 30.4 |
| | Loss | Dry deposition | Tg[N] yr ⁻¹ | 5.7 | 5.7 |
| | | Wet deposition | | 24.9 | 24.9 |
| | | Total | | 30.5 | 30.6 |

| | | | | |
|----------|--------------|-------|------|------|
| | Wet fraction | % | 81.4 | 81.4 |
| Burden | | Tg[N] | 0.42 | 0.42 |
| Lifetime | | days | 5 | 5 |

Table S5: Global and annual-mean metrics for nitric acid (HNO_3), nitrate (NO_3), ammonia (NH_3) and ammonium (NH_4) in the FAST and INSTANT simulations

| Region name | Abbreviation | Longitude range (°) | Latitude range (°) |
|-------------------------|---------------------|----------------------------|---------------------------|
| Northern Europe | NEU | 10.5W – 27.5E | 47N – 70N |
| Mediterranean Basin | MED | 10.5W – 37.5E | 30N – 47N |
| Western Africa | WAF | 20.5W – 20.5E | EQR – 18N |
| South Equatorial Africa | SQF | 0.5E – 55.5E | 26S – EQR |
| Central North America | CAN | 103.5W – 85.5W | 30N – 50N |
| South Asia | SAS | 65.5E – 104.5E | 5N – 30N |
| East Asia | EAS | 104.5E – 140.5E | 20N – 47N |
| Central Asia | CAS | 37.5E – 80.5E | 30N – 47N |
| Southeast Asia | SEA | 100.5E – 150.5E | 10S – 20N |

Table S6: Nine ‘Giorgi’ land region definitions used in this study (Giorgi, 2006)

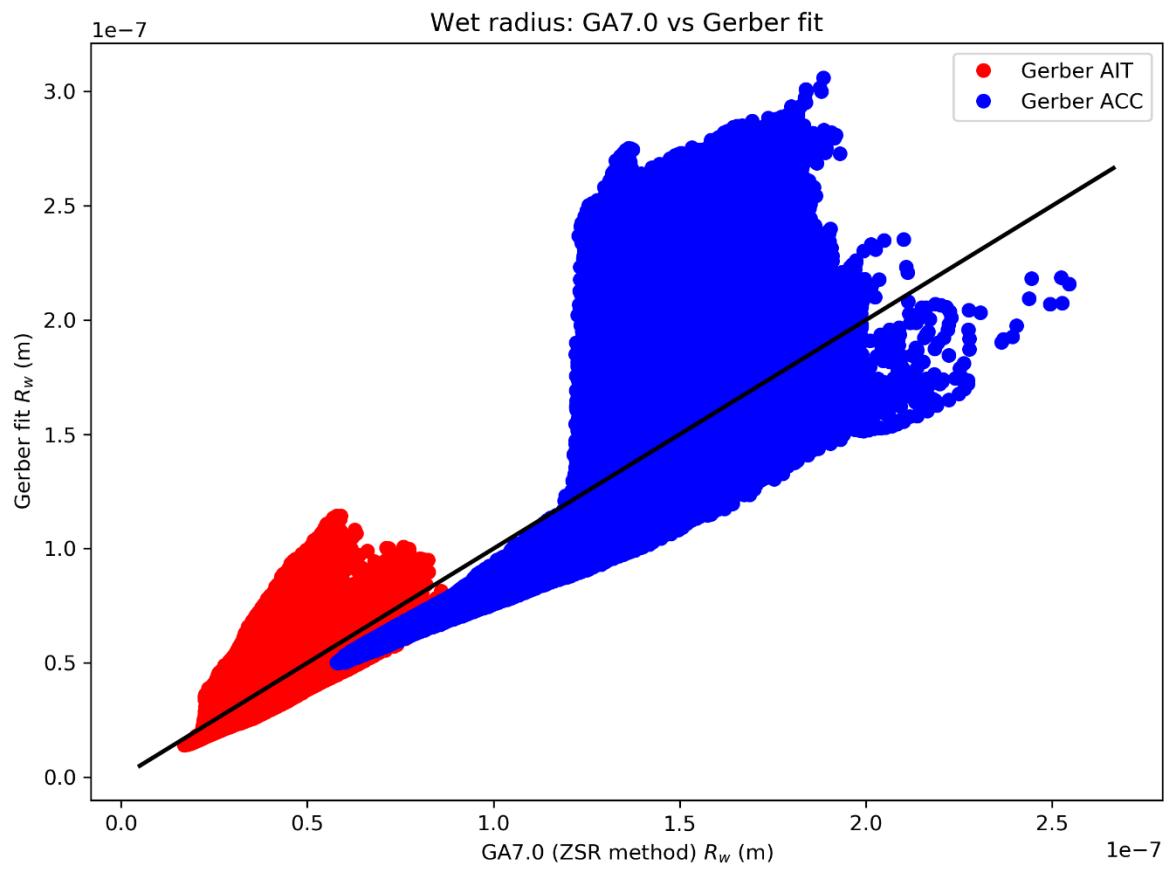


Figure S1: Comparison of wet radii calculated using Gerber's parameterisation applied to annual-mean dry radii, and annual-mean wet diameters using UKCA-mode's ZSR scheme. Results are shown for all grid-cells below the tropopause for a single year of a GA7.0 Met-UM11.5 N96 simulation (year 2000 conditions)

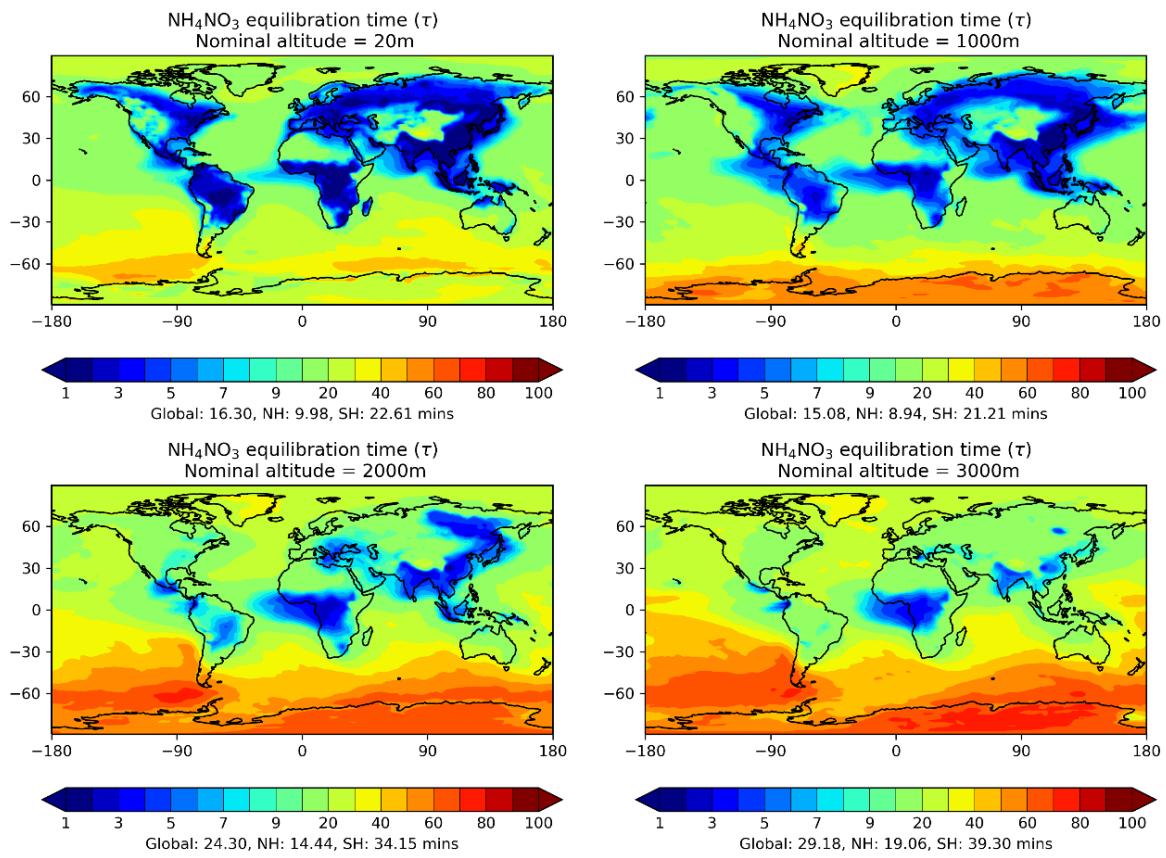


Figure S2: Timescale (τ) to reach equilibrium for 4 nominal altitudes determined using output for a single year of a GA7.0 Met-UM11.7 N96 simulation (year 2000 conditions)

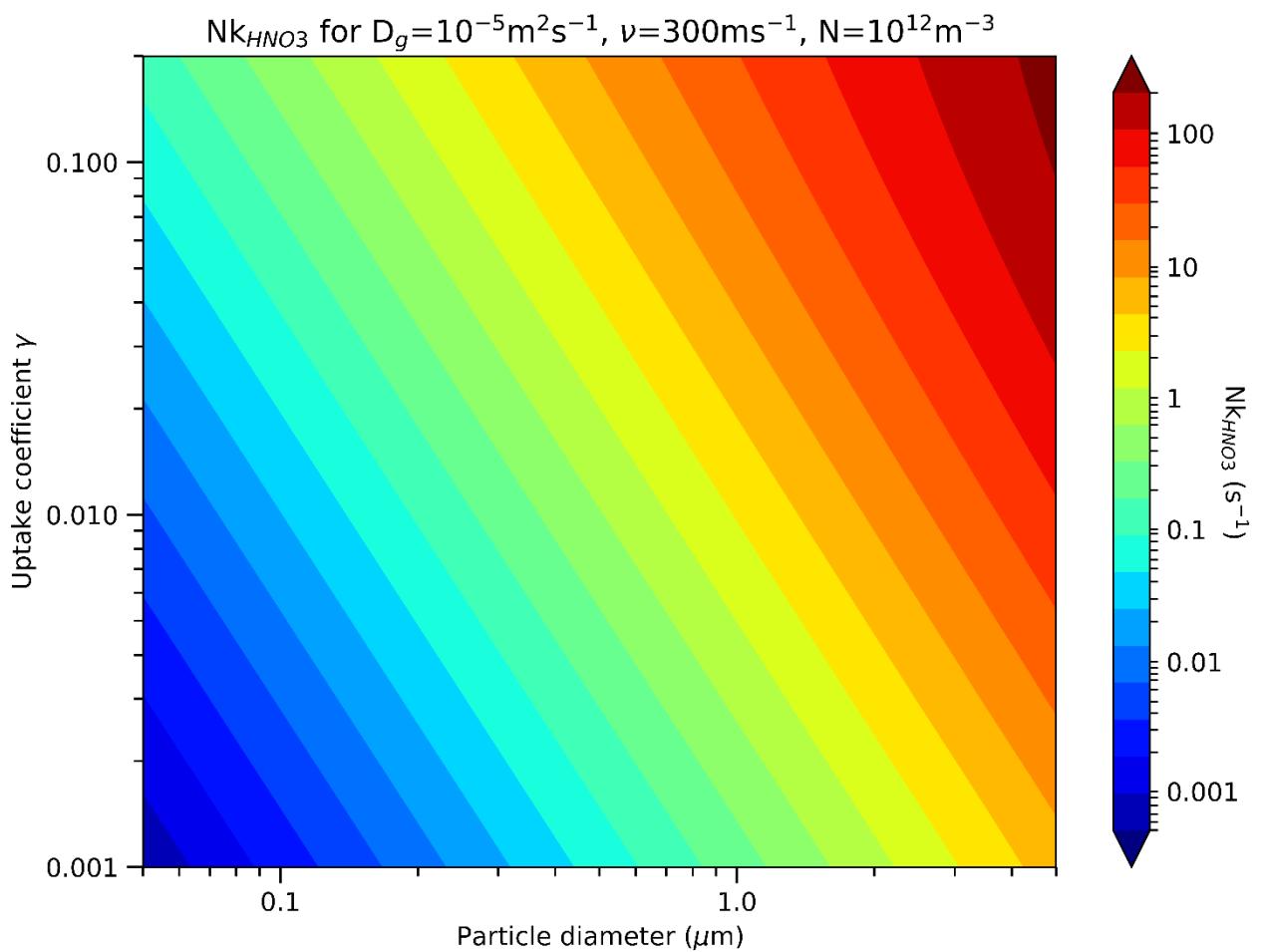


Figure S3: Sensitivity of the uptake rate (k_{HNO_3}) in standard atmospheric conditions to the uptake coefficient (γ , ranging from 0.001 to 0.2) and the particle diameter (D , ranging from 0.05 to 0.5 μm), normalised by a particle number concentration of $N = 10^{12} \text{ m}^{-3}$. k_{HNO_3} is determined using the first order uptake parameterisation (Eqs S9-S12).

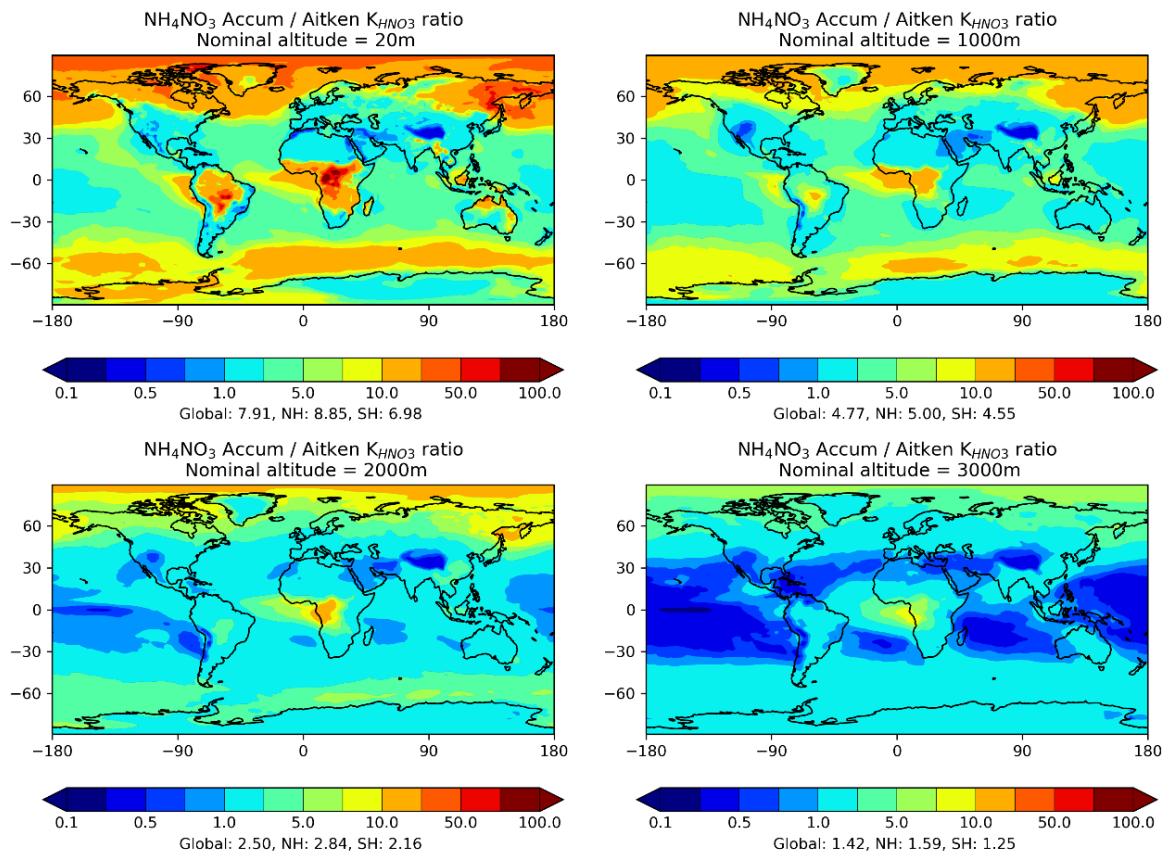


Figure S4: Ratio of Nk_{ACC} to Nk_{AIT} (Eqs S13 and S14) for 4 nominal altitudes determined using output for a single year of a GA7.0 Met-UM11.7 N96 simulation (year 2000 conditions)

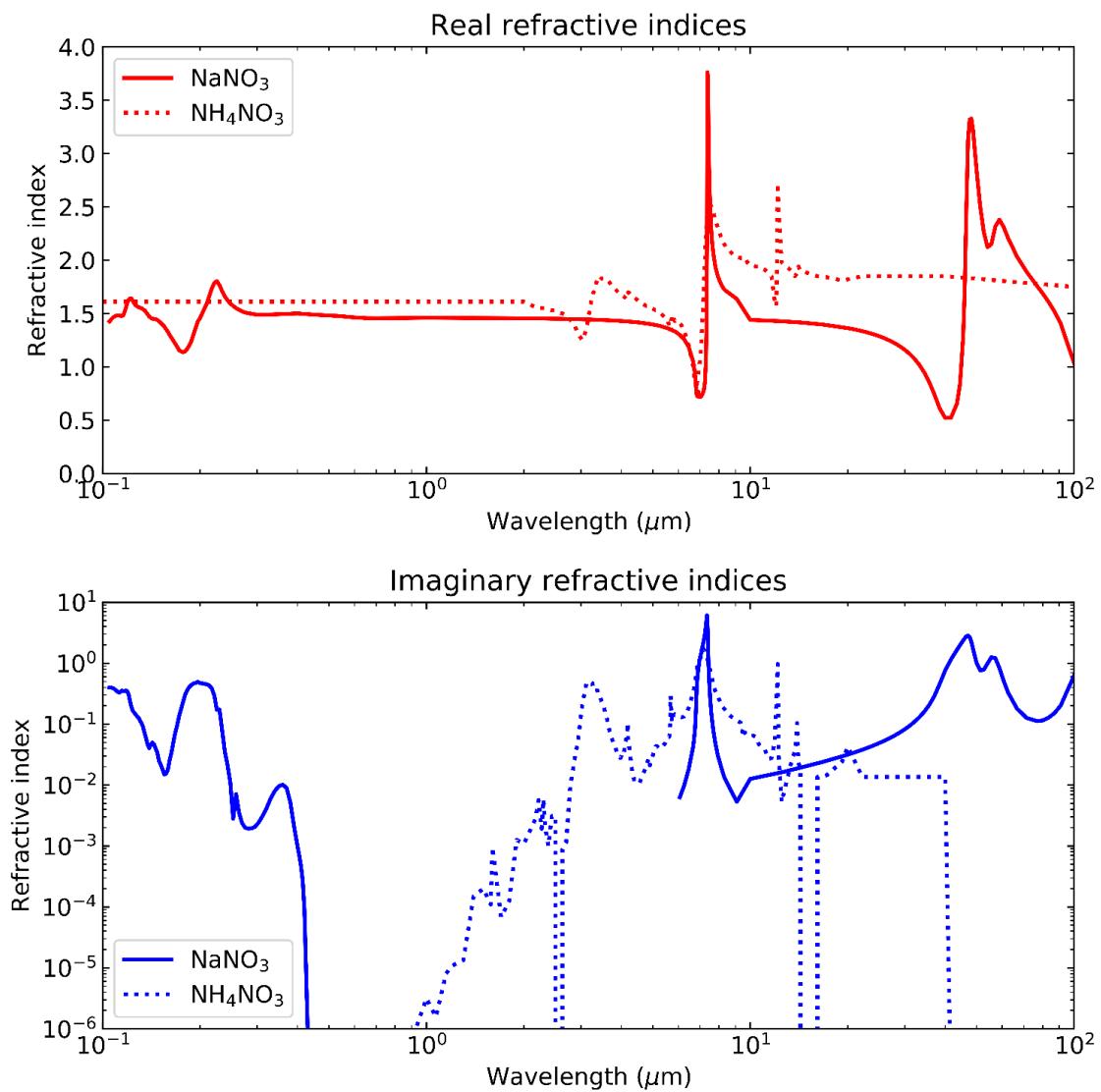


Figure S5: Refractive indices for sodium nitrate (NaNO_3) and ammonium nitrate (NH_4NO_3)

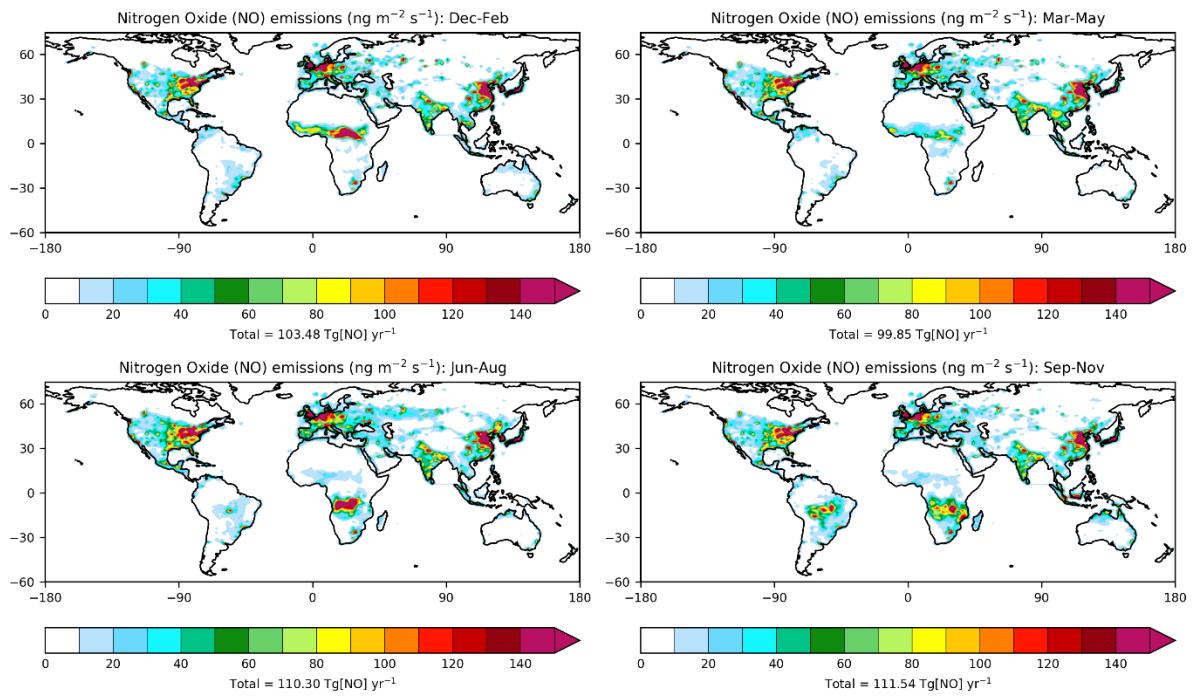


Figure S6a: Seasonal column-integrated nitrogen oxide (NO) emissions in the MetUM simulations

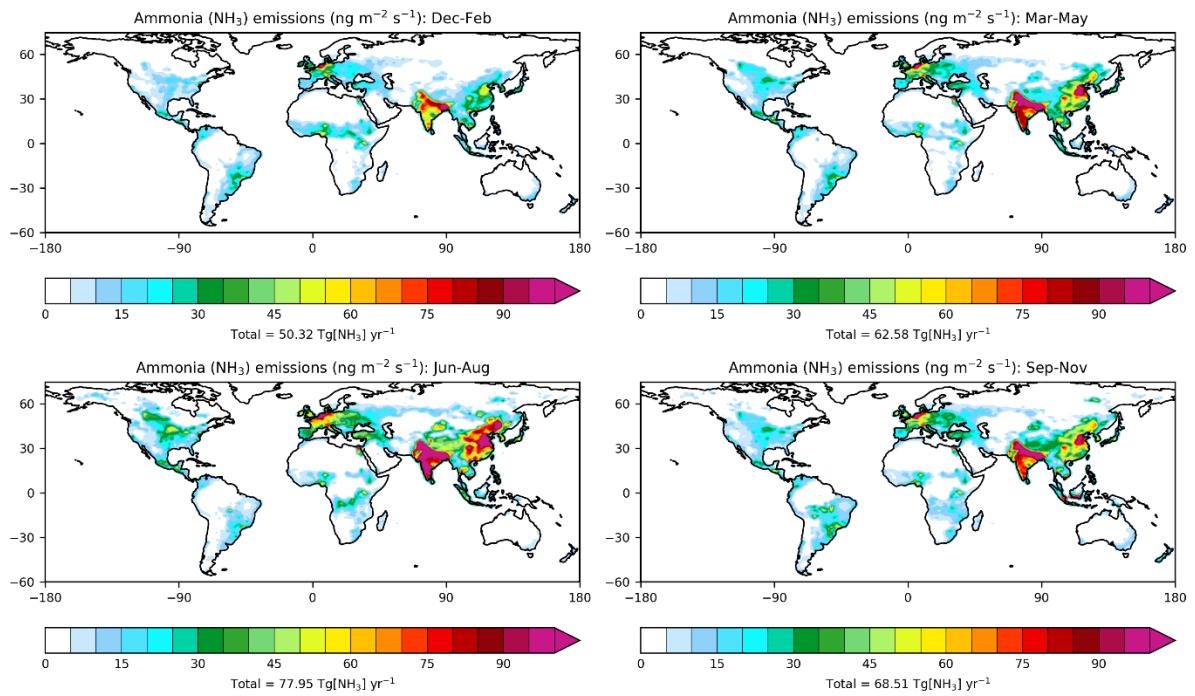


Figure S6b: Seasonal ammonia (NH_3) emissions in the MetUM simulations

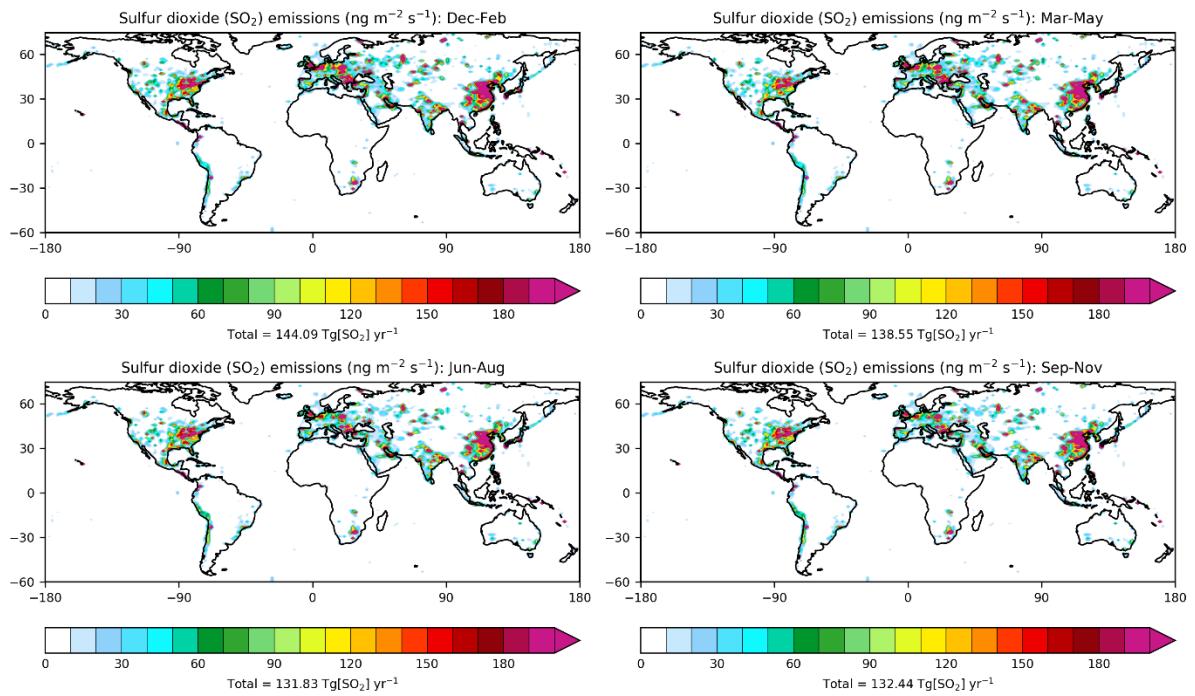


Figure S6c: Seasonal column-integrated sulphur dioxide (SO_2) emissions in the MetUM simulations

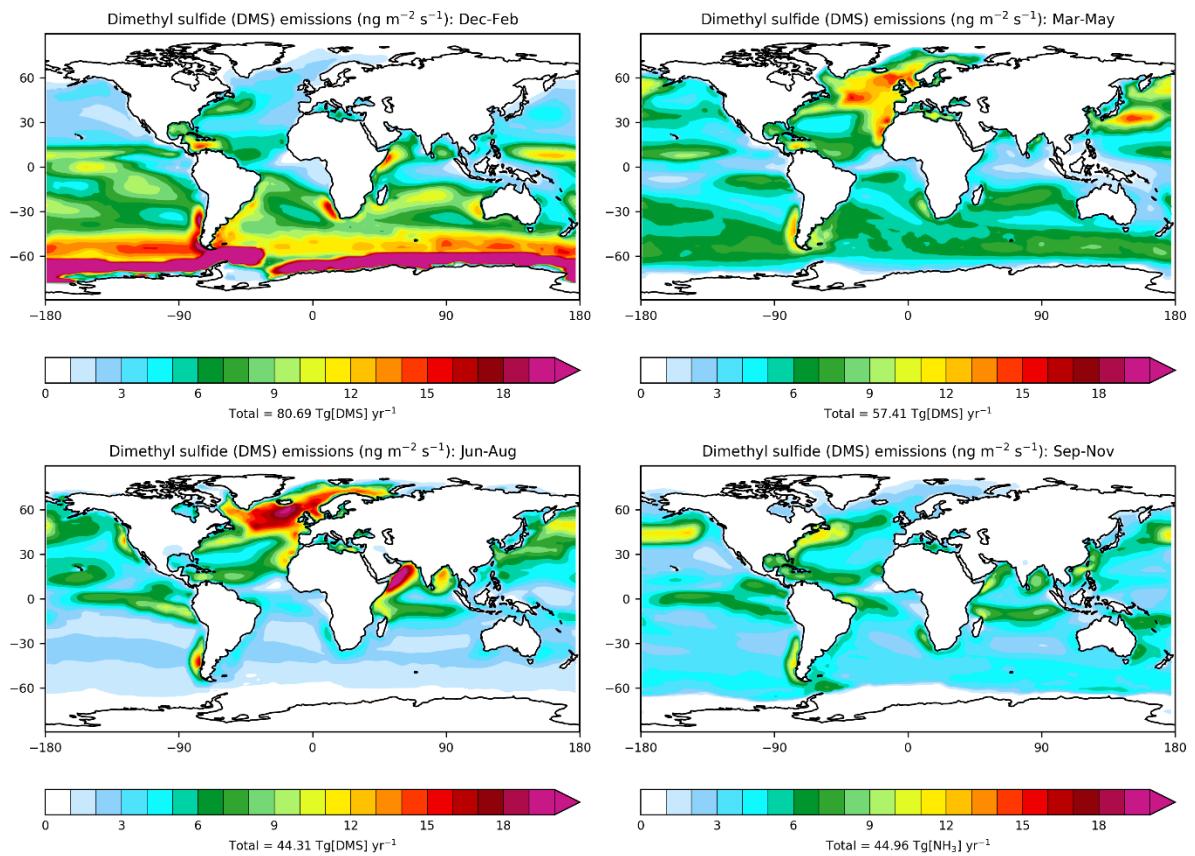


Figure S6d: Seasonal dimethyl sulfide (DMS) emissions in the MetUM simulations

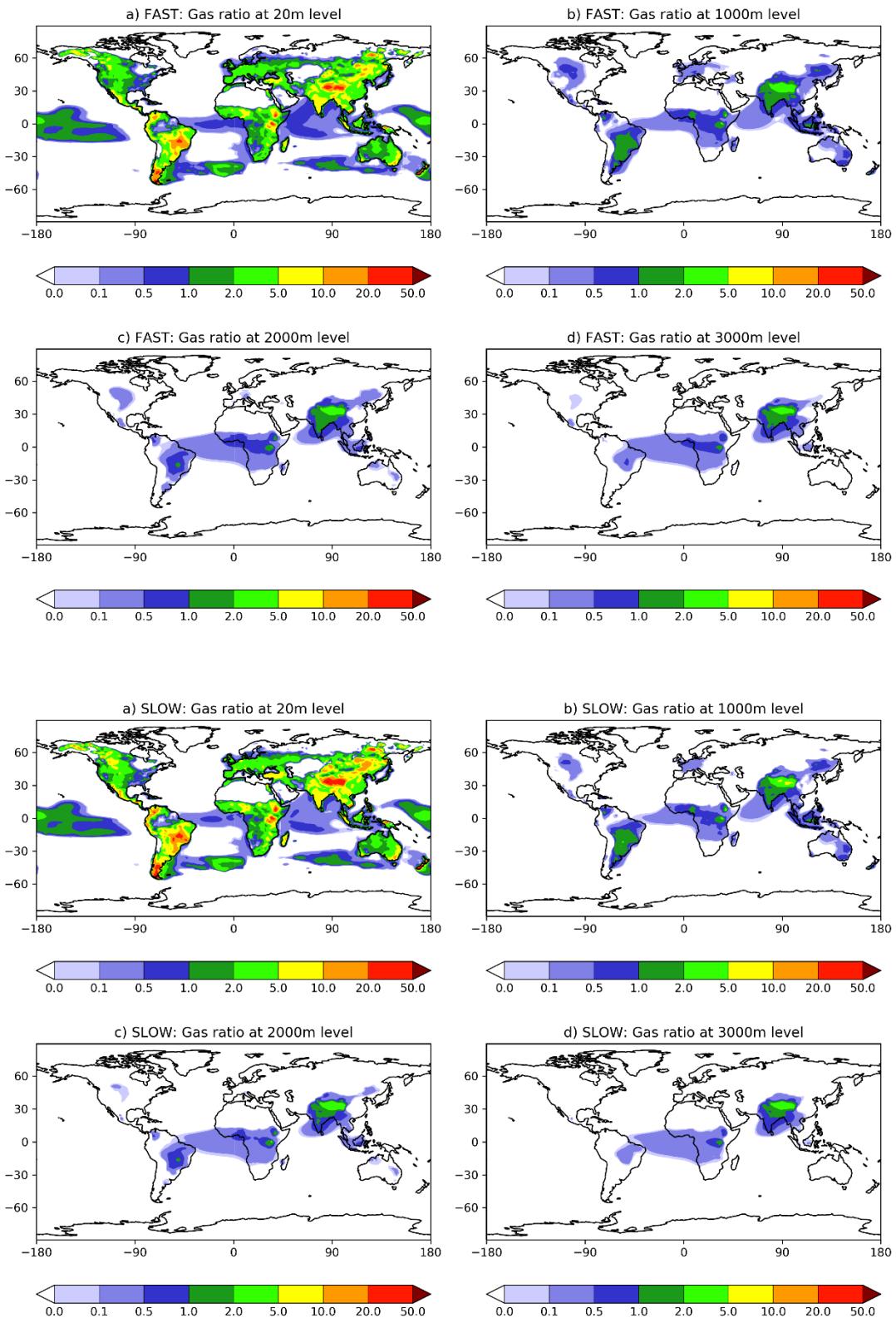


Figure S7: Annual-mean gas ratio in FAST (above) and SLOW (below), defined as $([\text{NH}_3] + [\text{NH}_4] - 2 \times [\text{SO}_4]) / ([\text{HNO}_3] + [\text{NO}_3])$. Values greater than 1 indicate that ammonium nitrate production is HNO₃ limiting, while values below 1 indicate that production is NH₃ limiting

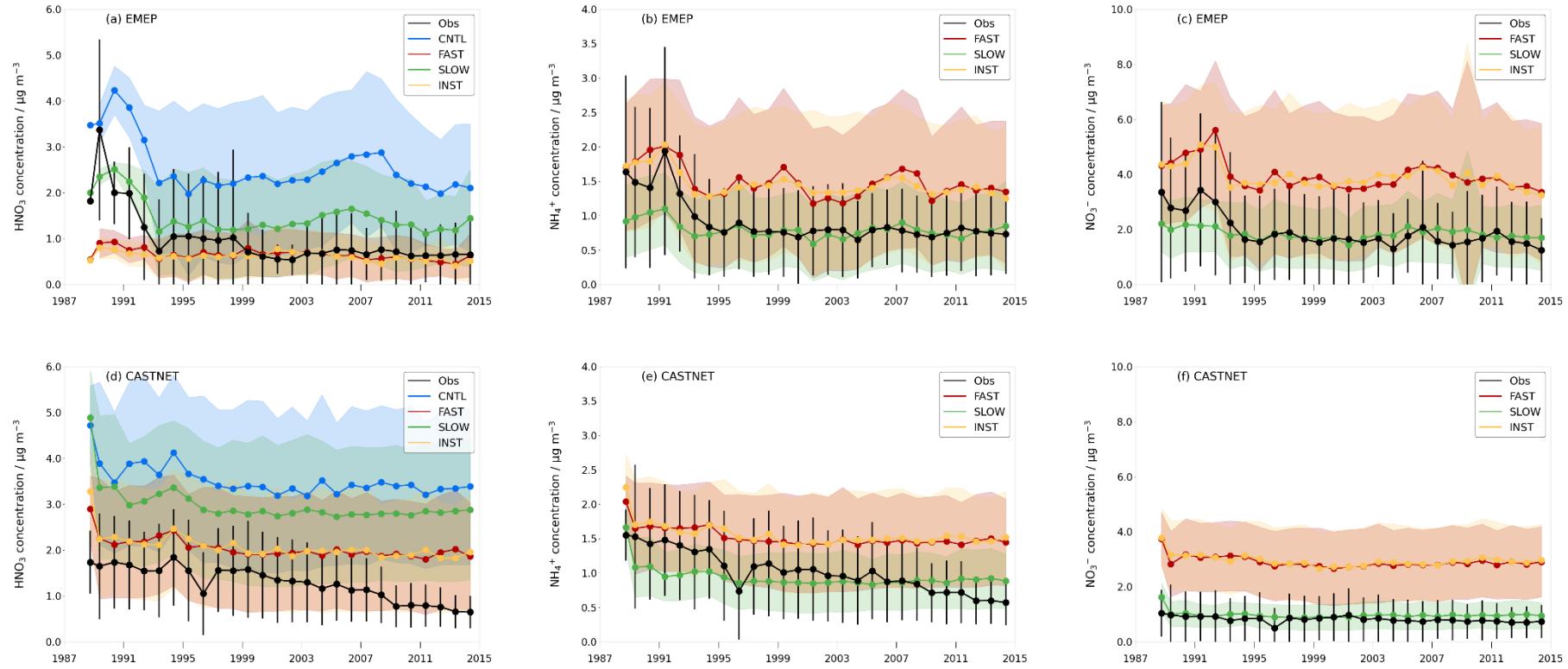


Figure S8: Annual-mean HNO_3 , NH_4 and NO_3 concentrations interpolated to CASTNET (US) and EMEP (Europe) observation sites for the period 1994–2013 and averaged over each network. For CASTNET there are a total of 49 sites. For the EMEP sites there were 59 for HNO_3 , 59 for NH_4 and 80 for NO_3 . Note that the years in the MetUM simulations (CNTL, FAST, SLOW, and INST or INSTANT) are arbitrary – the simulations use perpetual year-2000 conditions – and thus the simulations should not be compared with observations on a year-by-year basis.

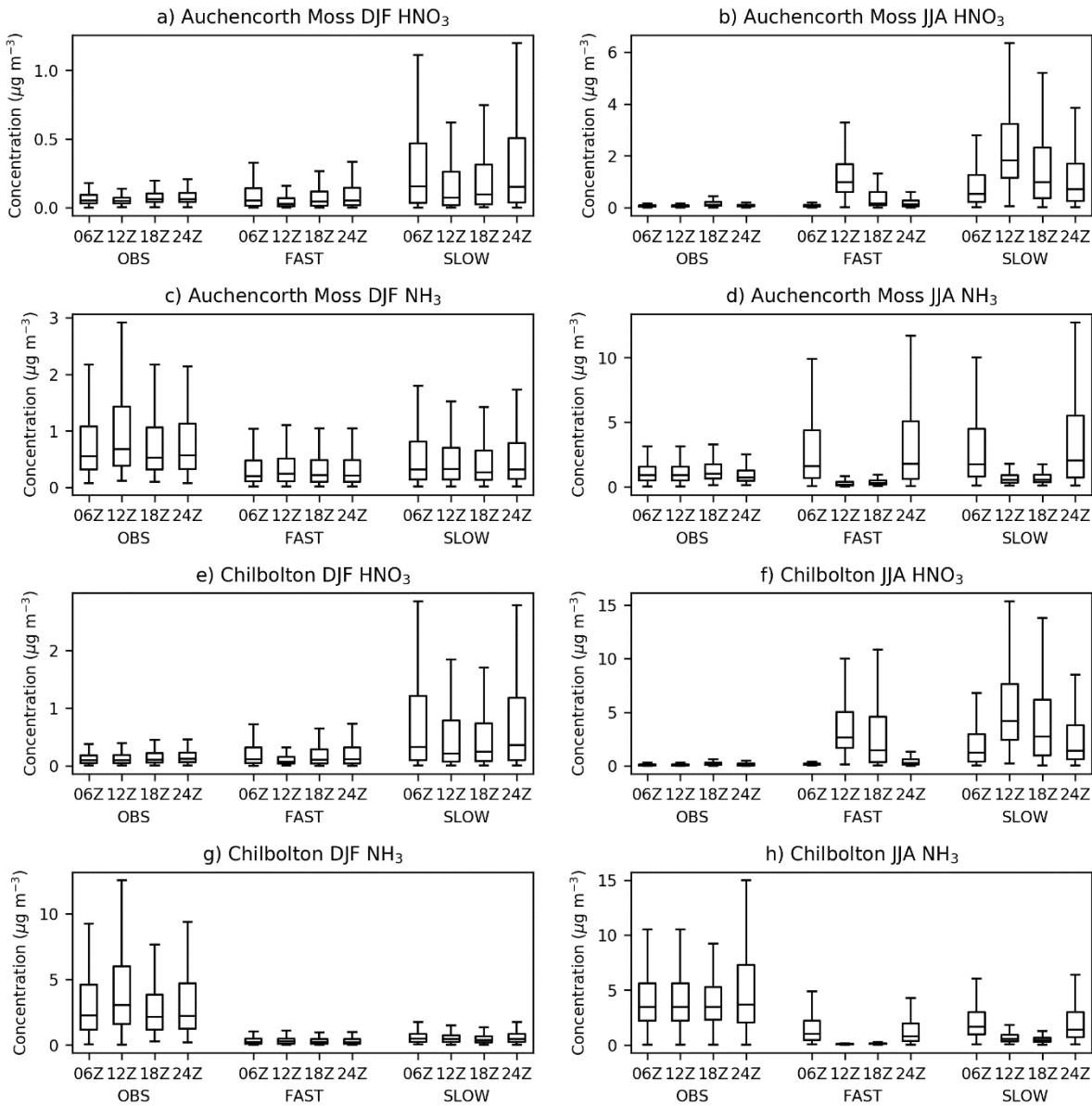


Figure S9: December–February (DJF) and June–August (JJA) diurnal cycles of near-surface HNO₃ and NH₃ concentrations in the FAST and SLOW simulations interpolated to two European Monitoring and Evaluation Programme (EMEP) supersites in the UK – Auchencorth Moss [55.79216°N, -3.2429°E] and Chilbolton [51.149617°N, -1.438228°E], alongside 6 hourly observations from 2014–2018 for Auchencorth Moss and 2016–2020 for Chilbolton. Equivalent to Fig. 7 in the manuscript.

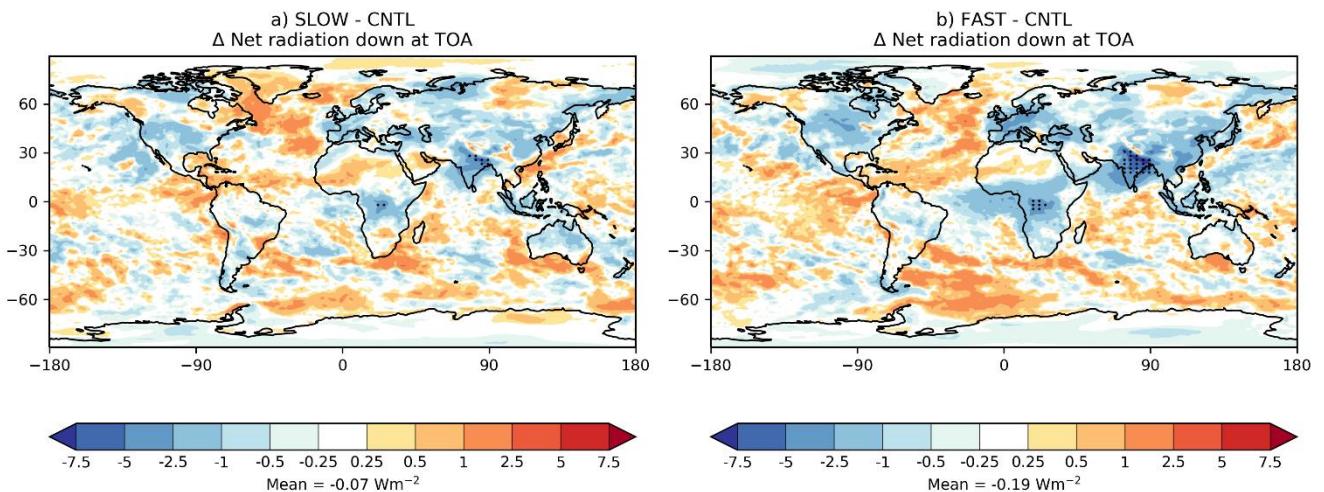


Figure S10: Annual-mean Top-Of-the-Atmosphere (TOA) net downwelling radiative flux (shortwave + longwave) perturbation or effective total radiative forcing in the a) SLOW and b) FAST simulations with respect to CNTL. Stippling indicates where differences are significant at the $\pm 2\sigma$ level