A thermodynamic equilibrium ammonium nitrate scheme in UKCA-mode

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Overview of nitrate scheme

- Semi-volatoile NH₄ and NO₃ emitted into Aitken and accumulation soluble modes using Mozurkewich (1993) parameterisation
- Rate at which NH₄·NO₃ reaches equilibrium is limited by first order uptake theory, i.e.

$$k = \frac{2\pi DD_g}{1 + \frac{4K_n}{3\gamma} \times \left(1 - \frac{0.47\gamma}{1 + K_n}\right)}$$

- The uptake coefficient (γ) is altered between FAST (0.193) and SLOW (0.001) in sensitivity simulations
- HNO₃ uptake on dust and sea-salt also simulated with first order uptake in ACC sol. and COA sol. modes
- This scheme is on the trunk at UM11.8 (see ticket 5262) in mode setup 10

Mode	Species
NUC SOL	SO ₄ , OM
AIT SOL	SO ₄ , BC, OM, NH₄, NO₃
ACC SOL	SO ₄ , BC, OM, SS, NH ₄ , NO ₃ , NaNO ₃
COA SOL	SO ₄ , BC, OM, SS, NH ₄ , NO ₃ , NaNO ₃
AIT INS	BC, OM

$$\frac{\mathrm{NH}_{4} \cdot \mathrm{NO}_{3} \text{ equilibrium theory}}{\mathrm{HNO}_{3} + \mathrm{NH}_{3} \stackrel{K_{p}}{\leftrightarrow} \mathrm{NH}_{4} \mathrm{NO}_{3}}$$
$$[\mathrm{NH}_{4} \mathrm{NO}_{3}]_{eq} = \frac{1}{2} \left[T_{A}^{*} + T_{N} - \sqrt{(T_{A}^{*} + T_{N})^{2} - 4(T_{N}T_{A}^{*} - K_{p})} \right]$$

Simulation	Rose Jobid	Description
CNTL	u-bz552	No nitrate – AMIP, UM11.7, N96, Strattrop
INSTANT	u-ca284	Nitrate – instant equilibrium
FAST	u-bz424	Nitrate – fast uptake ($\gamma = 0.193$)
SLOW	u-bz615	Nitrate – slow uptake ($\gamma = 0.001$)

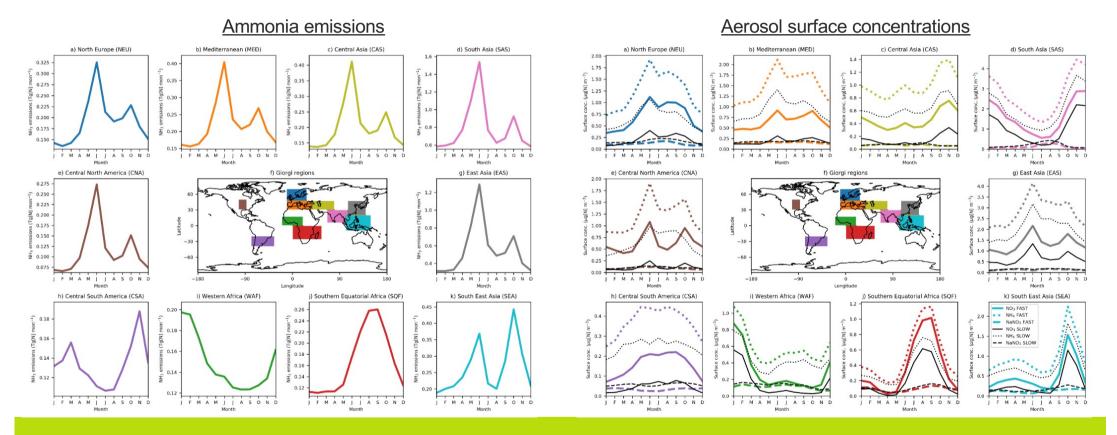
Near surface concentrations

b) FAST: NH₄ surface conc. (μ g[N] m⁻³) c) FAST-SLOW: NH₄ Δ (μ g[N] m⁻³) a) SLOW: NH₄ surface conc. (μ g[N] m⁻³) • Twice as much NO₃ in FAST as in SLOW 60 -60 60 30 30 30 CASTNET observations suggest NO₃ better 0 0 matched with SLOW, too much in FAST -30 -30 -30 -60 -60 -60 -180 -90 180 -180 180 -180 SLOW .01 .03 .05 .07 .03 .05 -0.05 0.01 0.1 0.5 .1 3 5 .7 3 .01 .07 .1 3 .5 .7 1 3 -0.7 -0.2 a) CASTNet West: HNO3 c) CASTNet East: HNO3 1 -2 5.0 5.0 Mean = 0.2368 Mean = 0.3115Mean = 0.0746. . . . b) HNO3 d) SLOW: NO₃ surface conc. (μ g[N] m⁻³) e) FAST: NO₃ surface conc. (μ g[N] m⁻³) f) FAST-SLOW: NO₃ Δ (μ g[N] m⁻³) 4.0 4.0 4.0 -3.0 -3.0 -2.0 -6n 3.0 60 - 19 2.0 30 30 0 1.0 -30 -30 -30 pearson_r=0.87 pearson r=0.50 0.0 ↓ 0.0 0.0*∸* 0.0 -60-60 -60 2.0 3.0 4.0 1.0 2.0 3.0 4.0 5.0 2.0 3.0 4.0 5.0 5.0 0 10 Obs. / µg m⁻³ Obs. / µg m⁻³ $[HNO_3] / \mu g m^{-3}$ -180 -180 -90 180 -180 d) CASTNet West: NH f) CASTNet East: NH 3.0 3.0 e) NH .01 .03 .05 .07 .1 .3 .5 3 .01 .03 .05 .07 .1 .3 .5 1 3 5 -0.2 -0.05 0.01 0.1 0.5 .7 1 .7 -0.7 2.5 -2.5 Mean = 0.0485Mean = 0 1136 Mean = 0.0651 то 2.0-м ди / I.5-1.0-E 2.0 g) SLOW: NaNO₃ surface conc. (μ g[N] m⁻³) h) FAST: NaNO₃ surface conc. (μ g[N] m⁻³) i) FAST-SLOW: NaNO₃ Δ (μ g[N] m⁻³) 6rf / 1.5 / Hodel / 60 30 30 30 0.5 0.5 pearson_r=0.29 pearson r=0.59 0.0 + 0.0 0.0 1.0 1.5 2.0 2.5 1.0 1.5 2.0 2.5 3.0 0.5 3.0 0.5 0.5 1.0 1.5 2.0 2.5 3.0 -30-30 -300 Obs. / μ g m⁻³ Obs. / μ g m⁻³ $[NH_{4}^{+}]/\mu g m^{-3}$ -60 -60 g) CASTNet West: NO3 i) CASTNet East: NO5 6.0 6.0 h) NO -180 180 -180 180 -180 5.0 5.0 $< \square$ E 4.0 .01 .03 .05 .07 .1 .3 .5 .01 .03 .05 .07 1 -0.2 -0.05 0.01 0.1 0.5 Ε 4.0 7 1 3 5 .1 .3 .5 7 3 5 -2 -0.7 Model / hg r 1 6rl / 3.0 Mean = 0.0780Mean = 0.0732Mean = -0.0048Model 50 1.0 -1.0 pearson r=0.83 pearson_r=0.00 0.0 0.0 0.0 2.0 3.0 4.0 5.0 6.0 0 1.0 2.0 3.0 4.0 5.0 6.0 2.0 3.0 4.0 5.0 6. 1.0 0.0 10 Credit: Catherine Hardacre Obs. / µg m-[NO₃] / µg m⁻³ Obs. / µg m⁻³

Near surface concentrations

Met Office

- Seasonal trends in $\text{NH}_4\cdot\text{NO}_3$ concentrations are highly sensitive to NH_3 emissions in most regions



elled PM2.5 (µg m⁻³)

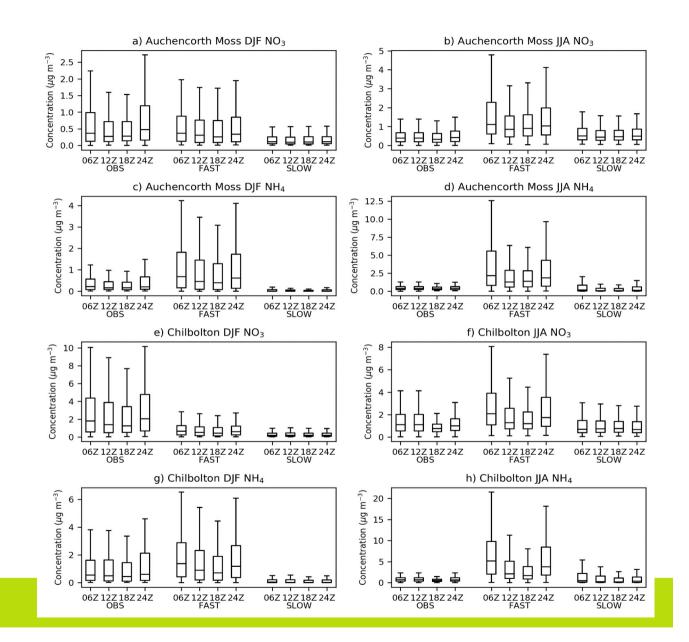
elled PM2.5 (µg m⁻³)

PM_{2.5} concentrations

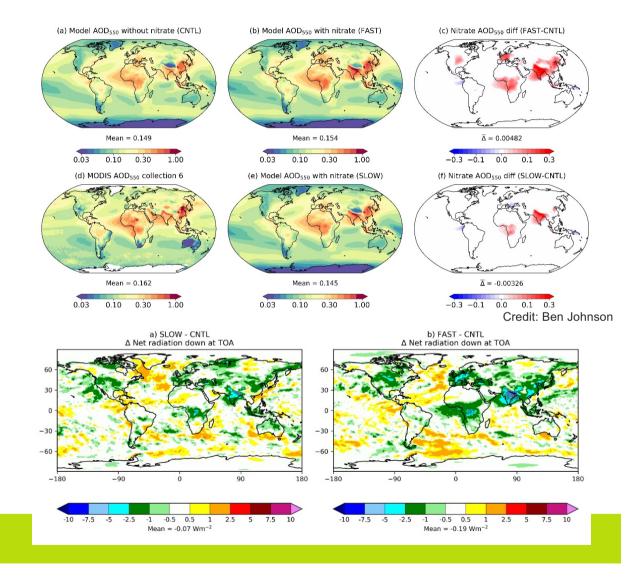
 Anomalous JJA increase in PM_{2.5} North Pole (2) Europe (51) East Asia (6) North America (185) 15.0 25 over Europe linked to spurious 80 12.5 15 20 NH₃ emissions? 10.0 60 15 10 7.5 40 • $PM_{2.5}$ not broken by $NH_4 \cdot NO_3$ 5.0 5 2.5 Concentration ($\mu g m^{-3}$) 0.0 IFMAMIIASOND J F M A M J J A S O N D J F M A M J J A S O N D J F M A M J J A S O N D Ocean (3) a) Control simulation b) Nitrate Fast simulation 12 Number of Points NMB = 0.26 RMSE = 5.84 r² = 0.57 Sigma = 1.10 y = m0.83 + 3.57 Number of Poir NMB = -0.13 RMSE = 5.15 $r^2 = 0.53$ Sigma = 0.82 y = m0.59 + 2 10 Έ Vodelled PM2.5 (µg 103 10 Surface Observations . . MERRA reanalysis control Surface PM_{2.5} nitrate fast 10 nitrate slow 101 101 PM2.5 CMIP6 IFMAMIIASOND Observed PM_{2.5} (µg m⁻³) Observed PM_{2.5} (µg m⁻³) PM2.5 approx CMIP6 c) Nitrate Slow simulation Central America (1) South Asia (5) South East Asia (8) Pacific AUS NZ (5) NMB = 0.02 RMSE = 5.27 $r^2 = 0.55$ Sigma = 1.01 y = m0.74 + 2.32 12.5 40 50 40 10.0 30 40 30 30 20 5.0 20 10 2.5 101 Observed PM_{2.5} (µg m⁻³) Δ 0.0 J F M A M J J A S O N D J F M A M J J A S O N D J F M A M J J A S O N D J F M A M J J A S O N D

Credit: Steve Turnock

 Diurnal cycle of NH₄·NO₃ concentrations peaking at night and lows in the afternoon largely captured by model and seen in UK observations



- △ AOD between -0.003 in SLOW and 0.005 in FAST
- Effective radiative forcing between -0.07 Wm⁻² in SLOW and -0.19 Wm⁻² in FAST
- Significant negative forcing over India, Europe and the DRC



Radiation and AOD

Jones, A. C., A. Hill, S. Remy, N. L. Abraham, M. Dalvi, C. Hardacre, A. J. Hewitt, B. Johnson, J. Mulcahy, and S. Turnock

Exploring the sensitivity of atmospheric nitrate concentrations to nitric acid uptake rate using the MetUM at vn11.7

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