Spatial and temporal CCN variations in convection-permitting aerosol microphysics simulations in an idealised marine tropical domain

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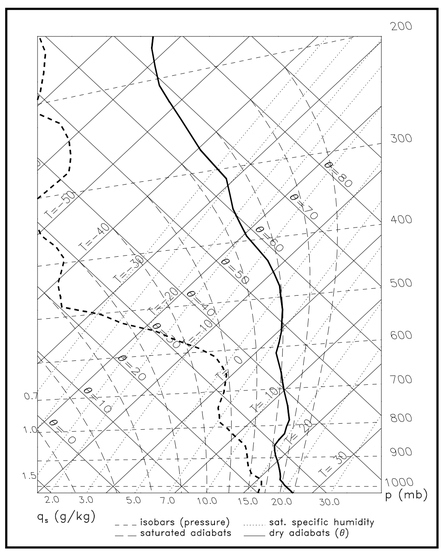
Objectives:

- Assess the temporal and spatial variation of CCN concentrations in the MBL at convection-permitting resolution
- Explore the influence of primary and secondary particles for determining the variability of different sized CCN at these fine spatial scales, considering also the implications for aerosol-cloud interactions simulated by current climate models
- Provide the basis for a ground-breaking new research tool for investigating aerosols-cloud interactions which will combine this interactive aerosol microphysics with also interactive cloud microphysics simulations.

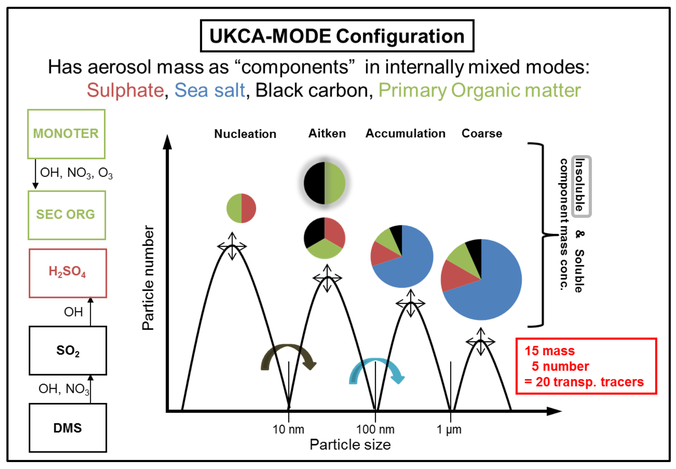
2) Initiation

- ***Domain characteristics:*** 160 x 160 grid points, Δx = Δy = **1.5 km**, Δt = 30 s, time simulation = 24 hours;
- ***Domain position:*** 1.50°-1.14° latitude & 0.0°-2.15° longitude (near the Gulf of Guinea);
- ***Initial atmos. data:*** according to a single model profile from a global aqua-planet start (UM operational run but **all land removed**);
- ***Initial tropo. tracers:*** chemistry tracers = 1 pptv & aerosol precursor gas phase tracers = 0.001 pptv, aerosols initialise to 0;
- ***Emissions:*** no dust; interactive emissions of DMS and sea-salt from the ocean (only fossil fuel BC/OC emissions are non-zero), others re-gridded from existing global emissions ancillaries;
- ***UKCA*:** (with DMS and sea-salt emissions) called every 10 time steps: ≈ 5 min.

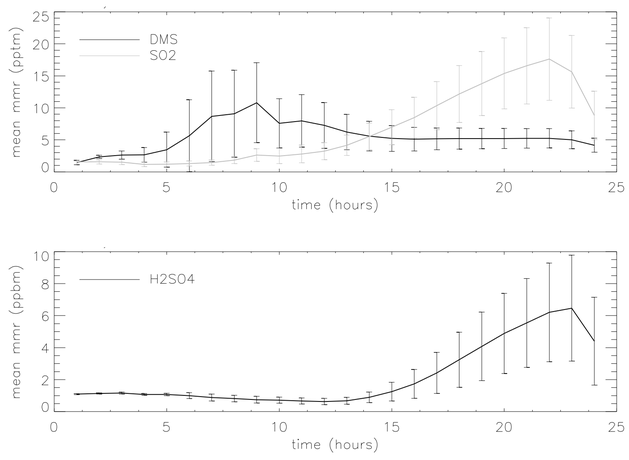


1) UKCA sub-model: UK Chemical and Aerosol

- UKCA = Unified Model + **GLOMAP-mode** aerosol microphysics scheme (*Mann et al., 2010*);
- 4 aerosol components in 5 modes (modal dust not used here [modes 6 and 7 switched off]) with 6-bin MetUM dust (as Woodward, 2001)
- Size-resolved emissions, removal & flux processes

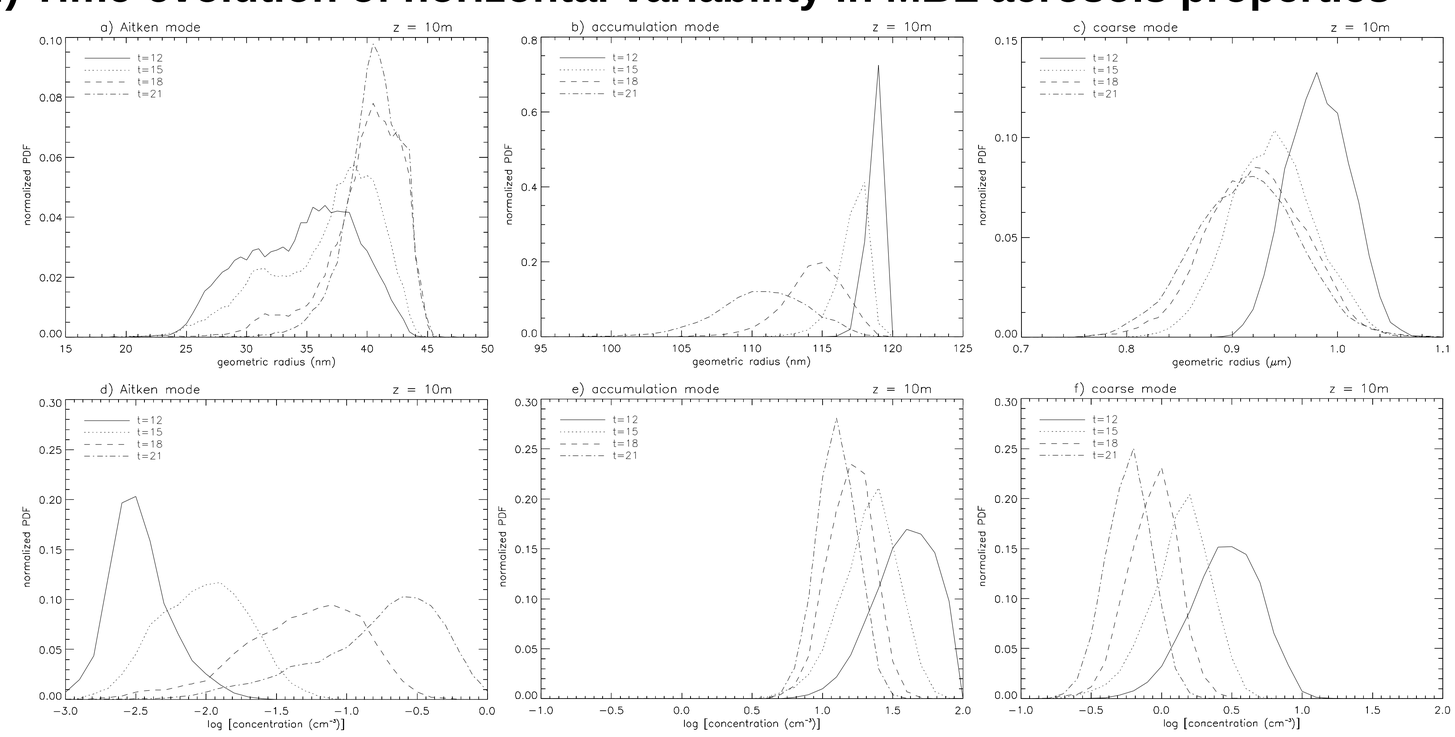


3) Gas-aerosols conversion



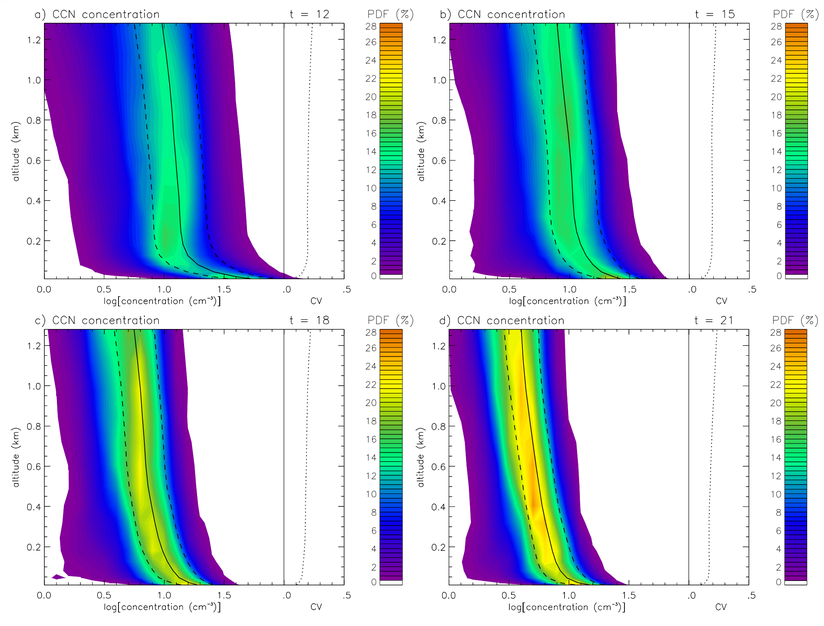
- DMS oxidised by OH and NO_3 producing SO_2 which in the gas phase goes to form H_2SO_4 via reactions with OH.
- H_2SO_4 then nucleate homogeneously or condense on existing particles to form marine CCN-active.

4) Time-evolution of horizontal variability in MBL aerosols properties



- *For the accumulation and coarse modes*, as time progresses, the particle size PDFs shift to smaller sizes. *Coarse mode* size-shift due to sedimentation reaching steady distribution after 18h. *Accumulation mode* size-PDF much wider after 18h due to source of smaller particles from *Aitken mode*.
- By contrast, as *Aitken* mode concentrations increase, the particles are clearly also larger, reflecting that growth processes are acting on the particles with this size-increase ceasing at about 18h when particles grow large enough to be cloud-processed up to accumulation mode.
- Whereas particle concentrations in the *accumulation and coarse modes* steadily decline as the wind speed declines, in the *Aitken mode* particle concentrations increase due to the entrainment of particles formed in the free troposphere.

5) Vertical profile of CCN variability



- The coefficient of variation, defined as the ratio of the standard deviation (dashed lines) to the mean (solid lines), of the CCN concentration increases with altitude from 11% to 22.5%.
- CCN concentration mainly influenced by emissions at the surface and by the transport at higher altitudes.
- Emergence of the secondary nucleated particles visible on the 18-21h period.

Conclusions *(Planche et al, in prep., 2016 – about to be submitted to ACPD)*

- In this marine atmosphere, the two dominant CCN sources are both natural: primary sea-salt particles and DMS-derived secondary sulphate particles formed in the free troposphere following oxidation.
- There is a diverse community of processes: dynamical, chemical, and microphysical, that together combine to determine the number of particles (and its extreme variability) which can activate to cloud droplets.
- The dynamics strongly influences the sea-spray emissions into atmosphere; then, the sea-salt aerosols are transported vertically by turbulent diffusion, with larger particles also being influenced by sedimentation.
- The emissions of DMS strongly vary spatially and temporally according to wind speeds conditions; then, the gas phases produced from the oxidation of DMS are a prequisite for effective new sulphate particle formation in free troposphere and cause growth (e.g. coagulation) of existing particles following vapour condensation.
- A significant proportion of these small secondary particles grow large enough to be cloud processed or mode-merged from the Aitken mode to accumulation mode.

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