Study of the CCN formation using the TM4-ECPL and M7 model

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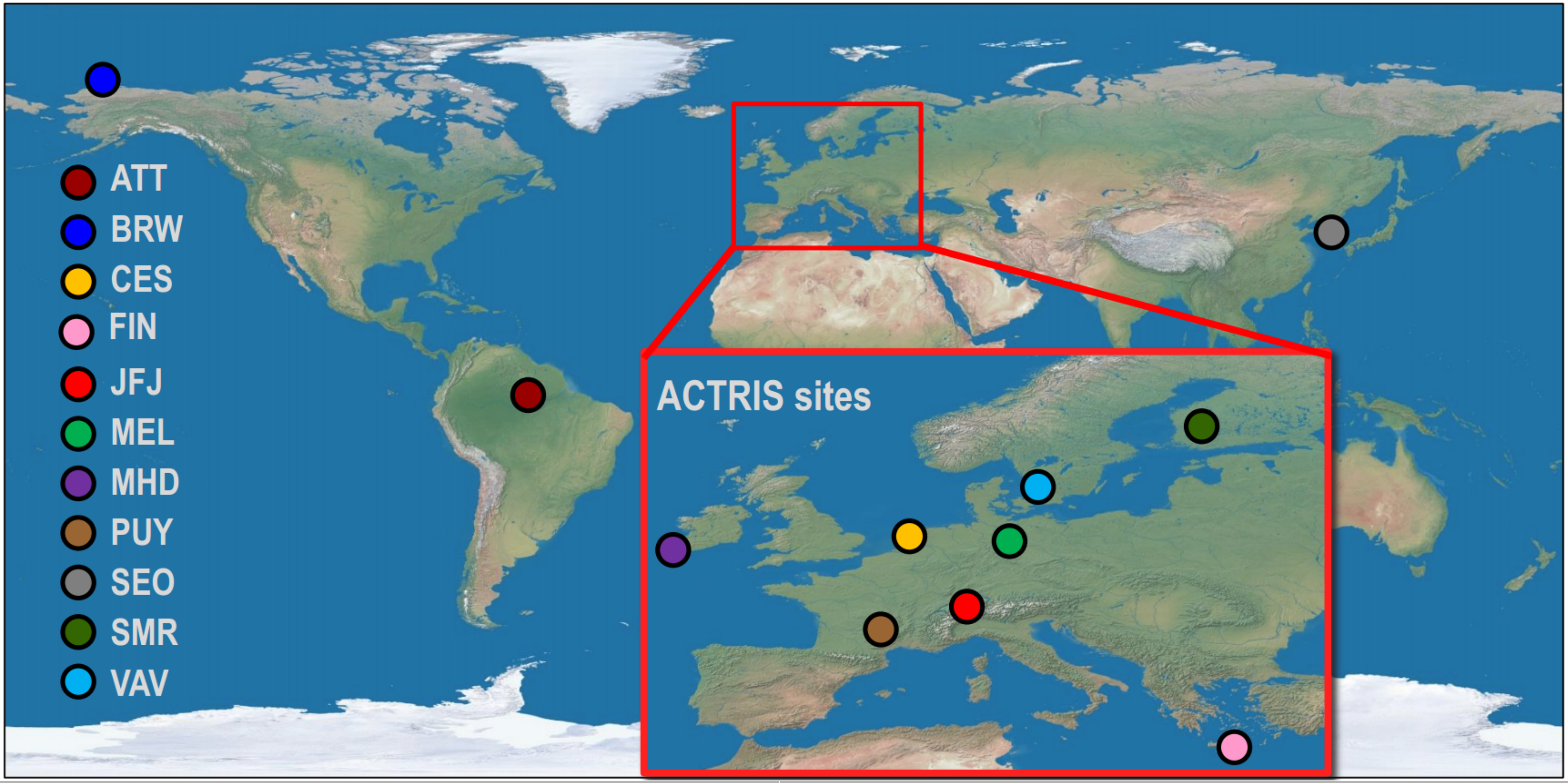
Motivation - Aim

- To examine the relative impact of anthropogenic versus natural aerosols to the CCNs in various environments
- Understand aerosol ageing in the atmosphere and its influence on CCN properties using model simulations
- Improve description of the OA formation and transformation in atmosphere and develop parameterizations of formation and growth of CCN by atmospheric dynamic processing

Observational sites



J. Schmale *et al.* 2015

ATT: ATTO tower, Brazil, rainforest	MHD: Mace Head, Ireland, marine
BRW: Barrow, USA, Arctic remote	PUY: Puy de Dôme, France, mountain
CES: Cabauw, Netherlands, rural/coastal	SEO: Seoul, South-Korea, urban
FIN: Finokalia, Greece, mediterranean	SMR: Hyytiäla, Finland, boreal forest
JFJ: Jungfraujoch, Switzerland, high-altit.	VAV: Vavihill, Sweden, rural-background
MEL: Melpitz, Germany, rural-remote	

Model setup

TM4-ECPL

- multiphase tropospheric chemistry accounting for volatile organic compounds
- major aerosol components, including secondary inorganic and organic aerosols (Kanakidou et al., 2012; Daskalakis et al., 2015)
- low horizontal resolution of 6x4 in longitude and latitude and 34 vertical hybrid layers from the surface up to the 0.1 hPa pressure level
- Anthropogenic and biomass burning emissions from the ACCMIP and biogenic emissions from the MEGAN-MACC databases. Mineral dust daily emissions come from the AEROCOM database and ECMWF (ERA-Interim) meteorology for 2008 is used
- Simulations for the years 2012-2013 after 5-years spinup
- Off line calculation of CCNs

M7 microphysics model (Vignati et al., 2004)

- Particles are divided in the
  - a) water-soluble particles
    - Nucleation (sulphate)
    - Aitken (sulphate, BC, OC, SOA)
    - Accumulation and Coarse (sulphate, BC, OC, dust, SOA)
  - b) insoluble particles
    - Aitken (BC, OC, SOA)
    - Accumulation and Coarse (dust)
- Aerosol dynamics (nucleation, mixing and growth by condensation and coagulation)
- Log-normal distributions describe particles numbers at each mode

Model setup *(cont.)*

CCN calculation

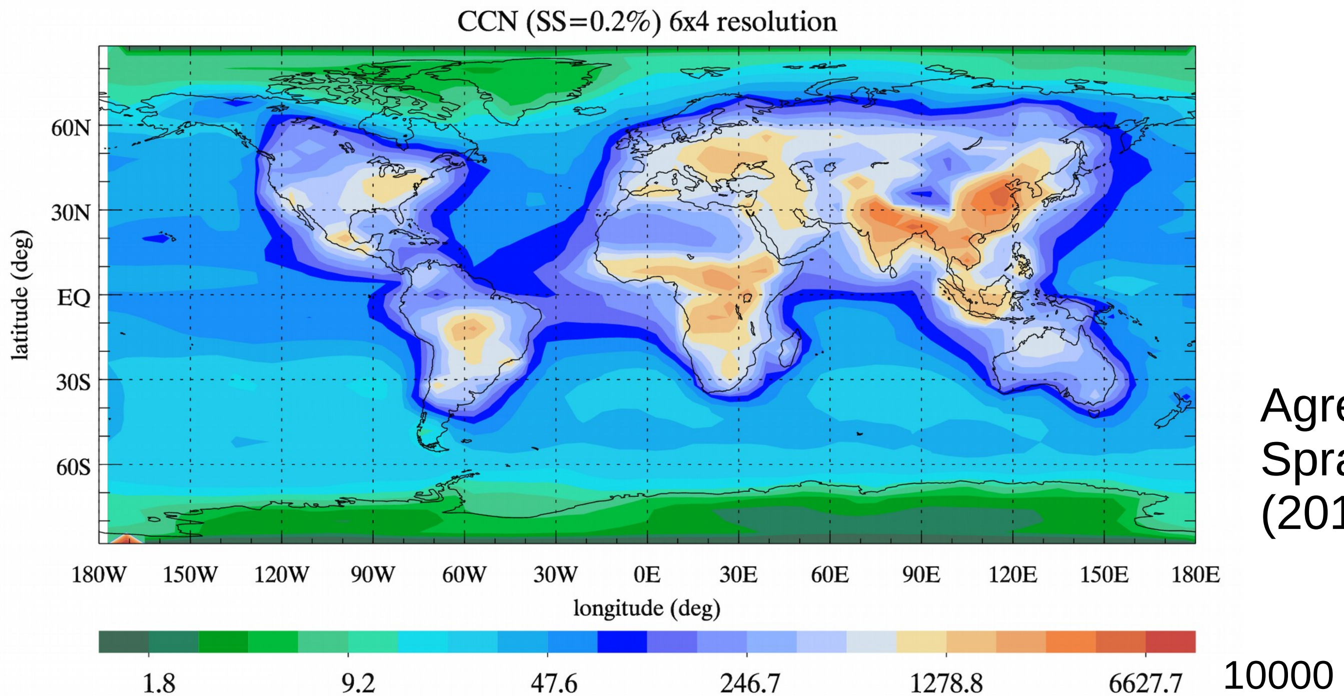
- Hourly output of data necessary for the calculation of CCNs (e.g. temperature, aerosol masses and numbers) during simulations with TM4-ECPL
- CCNs were computed according to the *Petters and Kreidenweis*, ACP, 2007 approach

$$S(D)=\frac{D^3-D_d^3}{D^3-D_d^3(1-\kappa)}\exp\left(\frac{4\sigma_{s/a}M_w}{RT\rho_w D}\right)$$

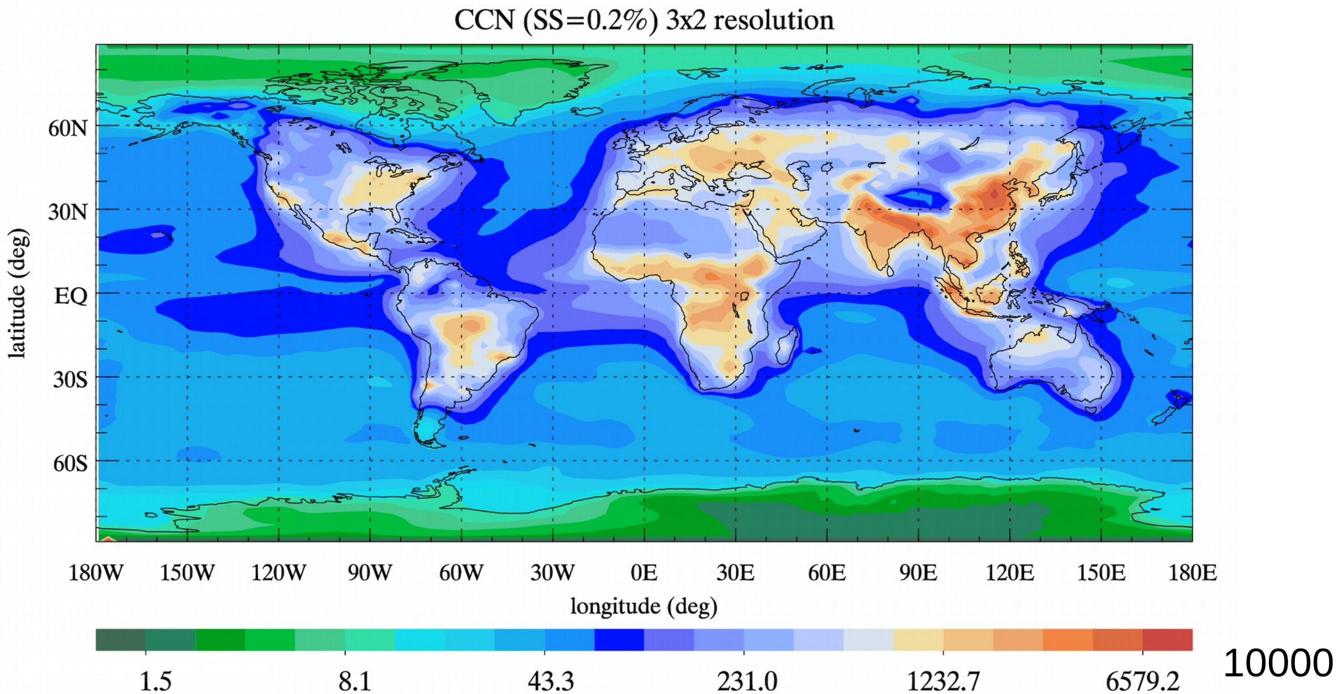
$$\kappa=\sum_i \kappa_i\varepsilon_i, \qquad \varepsilon_i = \text{dry component volume fraction}$$

- Only hydrophilic modes participate to the CCN formation (Spracklen et al., ACP, 2011)
- Various sets of hygroscopicity parameters were examined. For the base case: 0.61 for sulfate particles, 1.28 for sea-salt, 0.227 for primary particulate organic matter (POM) and secondary organic aerosols (SOA), 0.0 for dust and black carbon

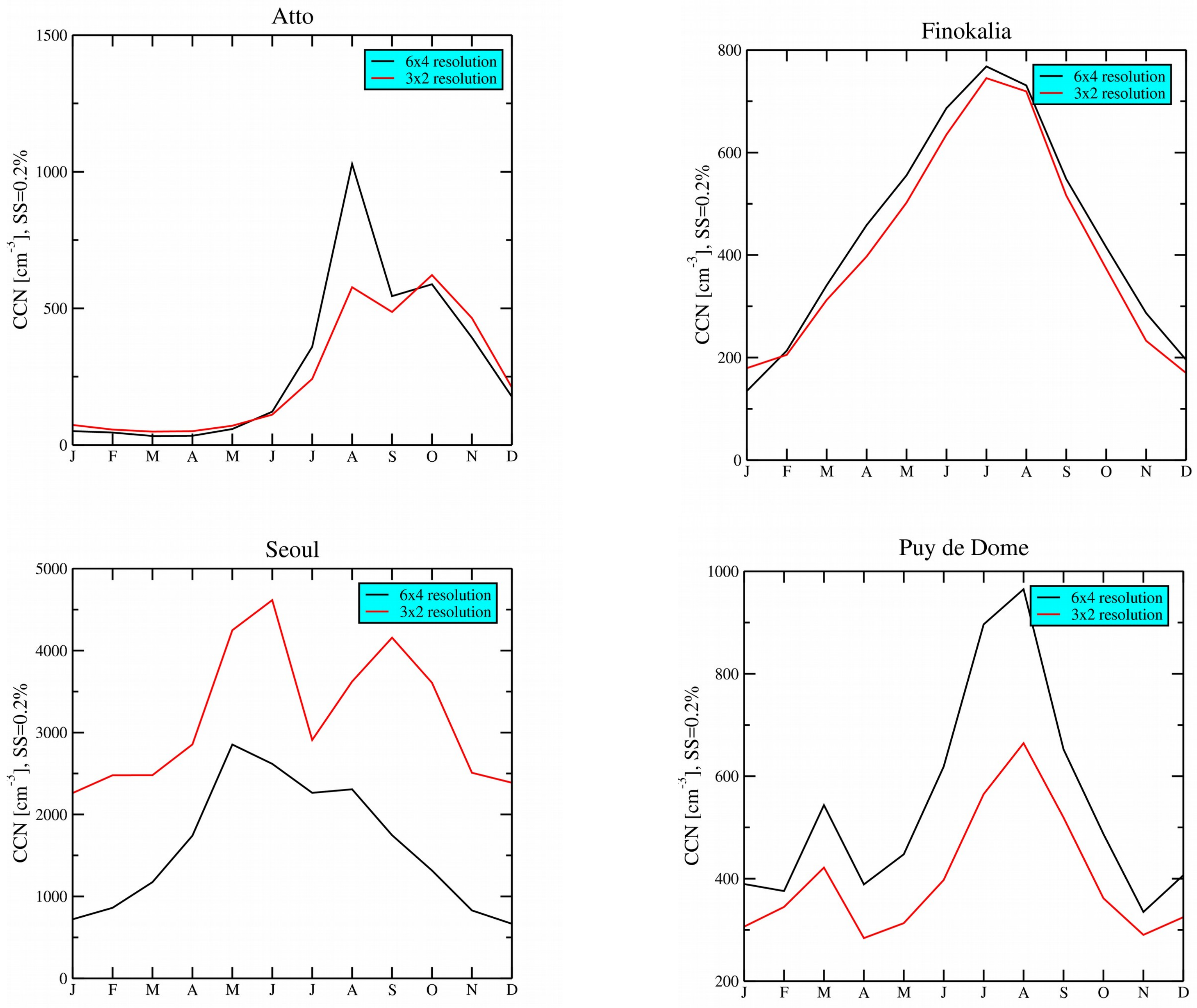
Sensitivity to the model resolution



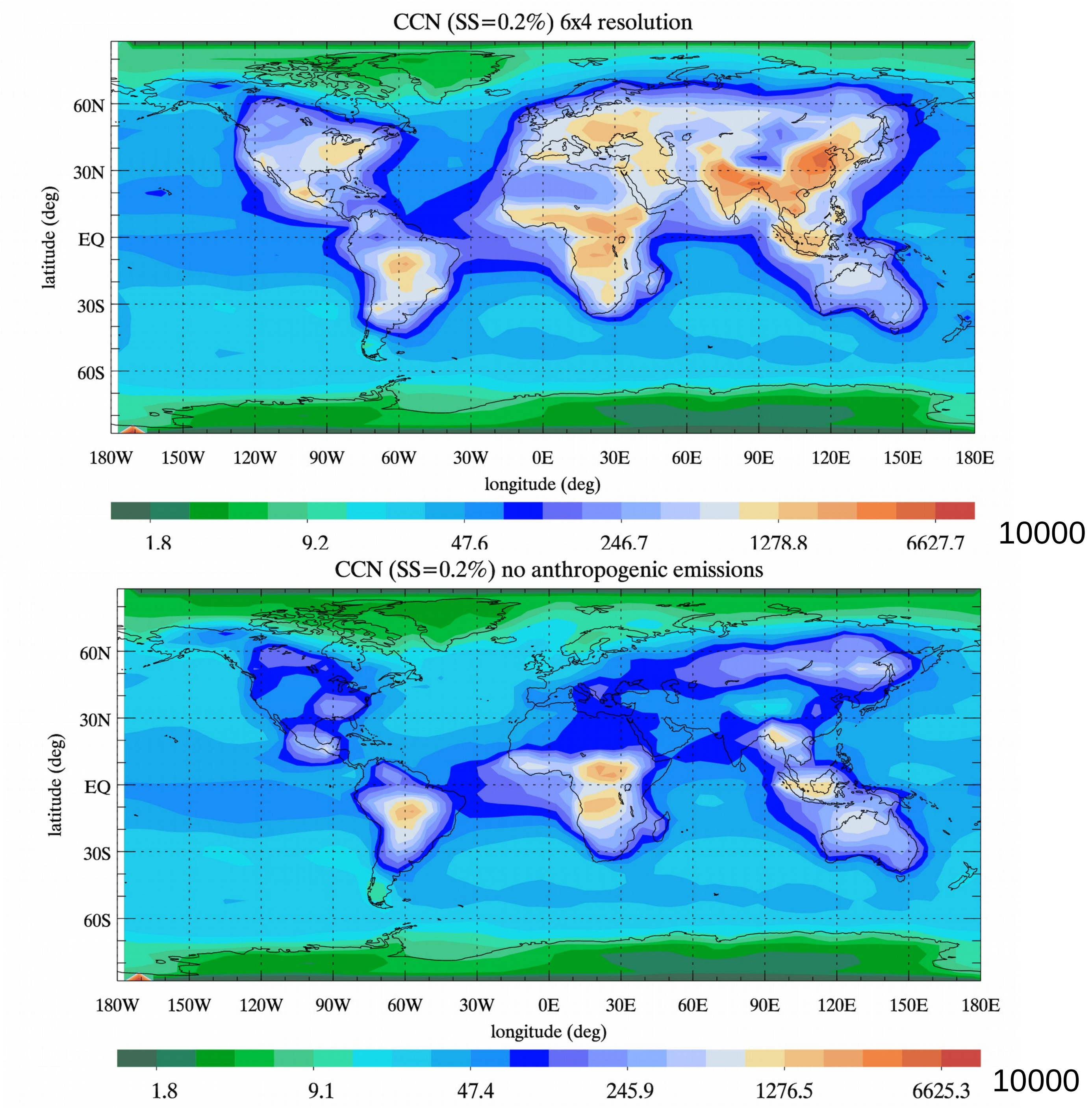
Agreement with Spracklen *et al*., ACP (2011)



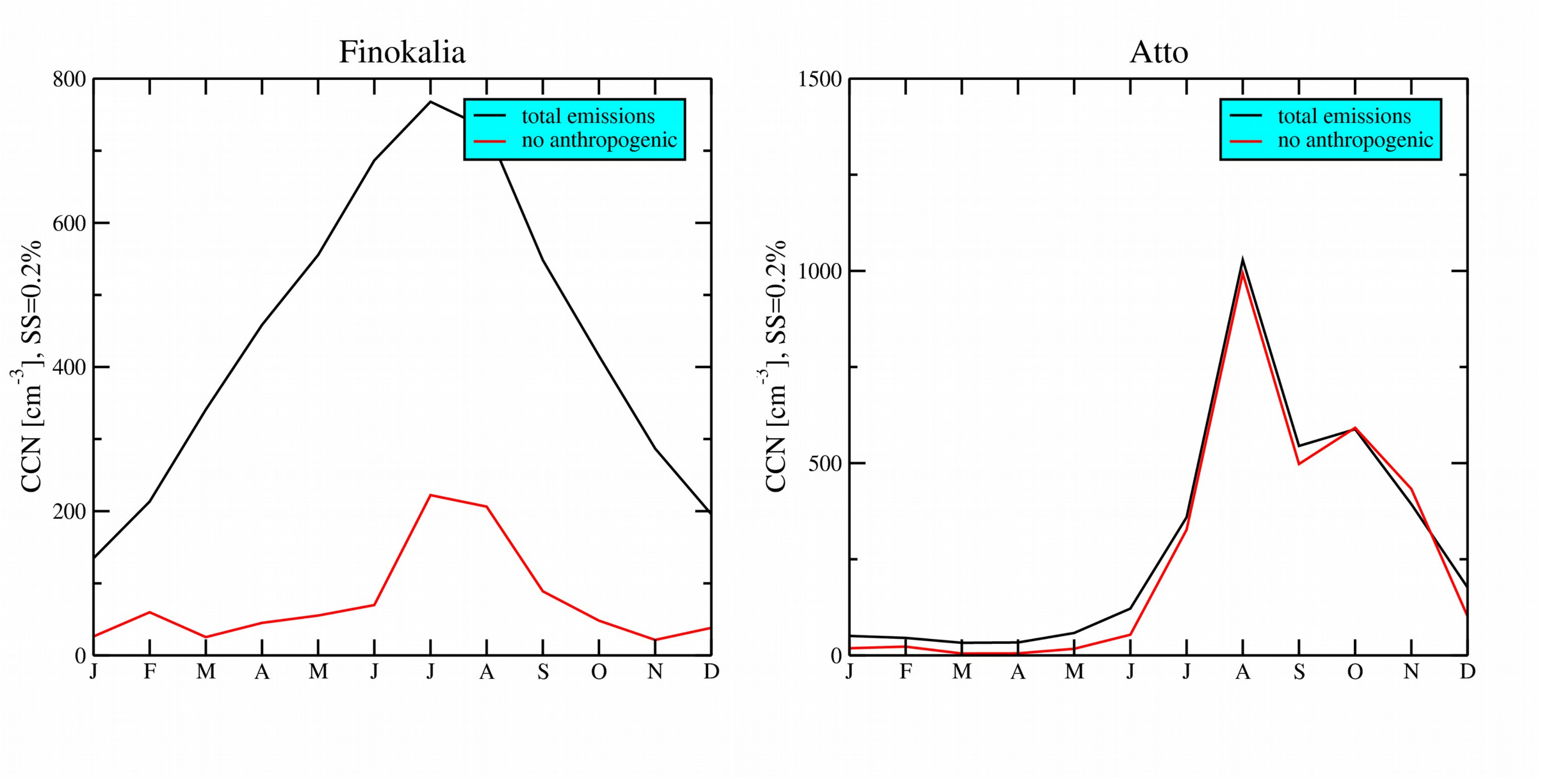
Sensitivity to the model resolution *(cont.)*



Impact of anthropogenic emissions



Impact of anthropogenic emissions (*cont.*)



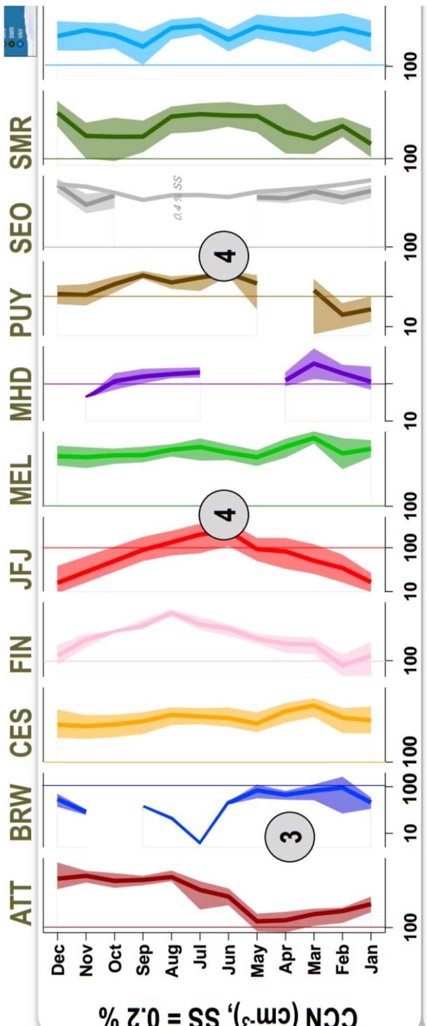
Comparison of CCNs

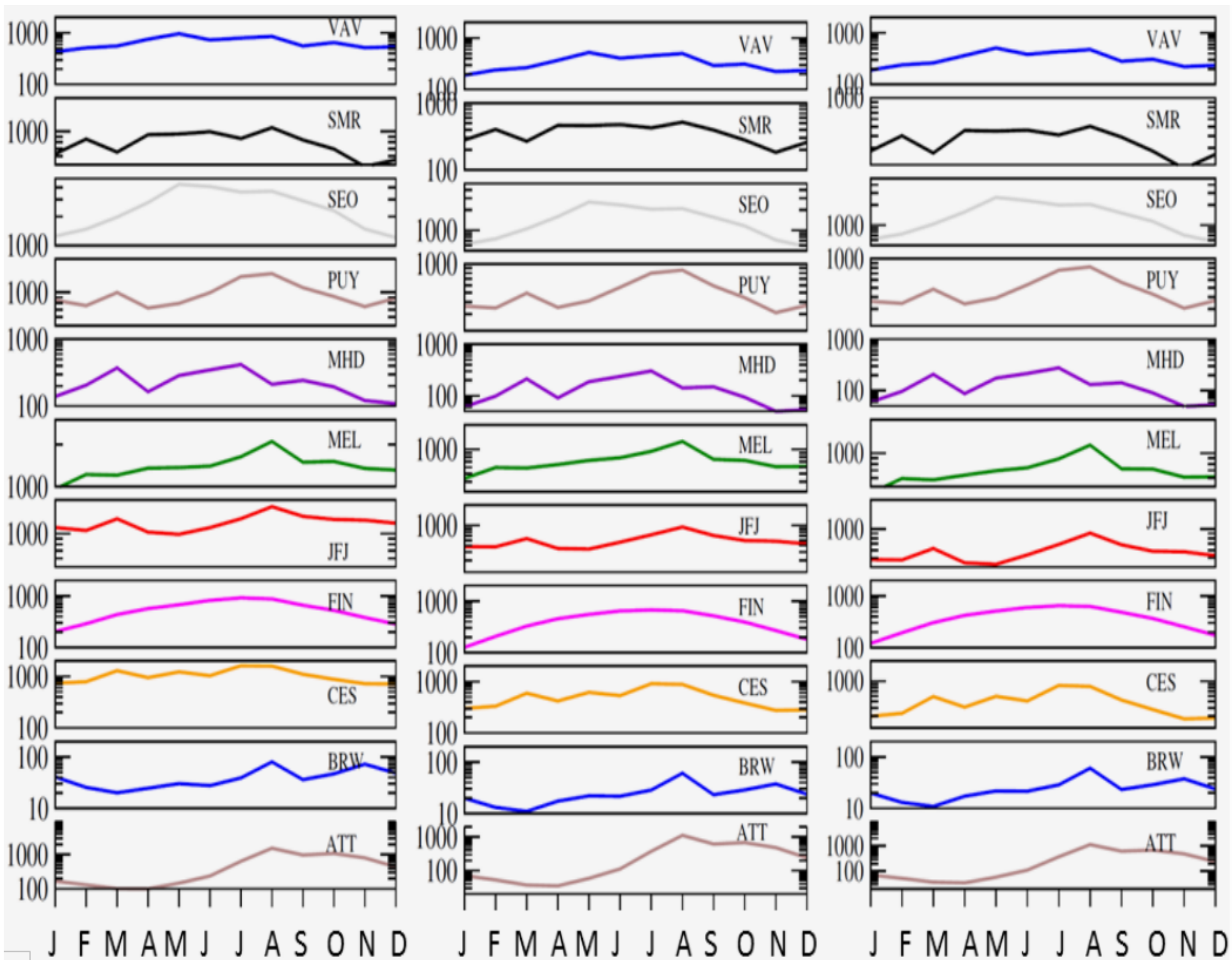
observations

κ_{sulf}=0.61, κ_{SS}=1.28, κ_{POM}=0.227

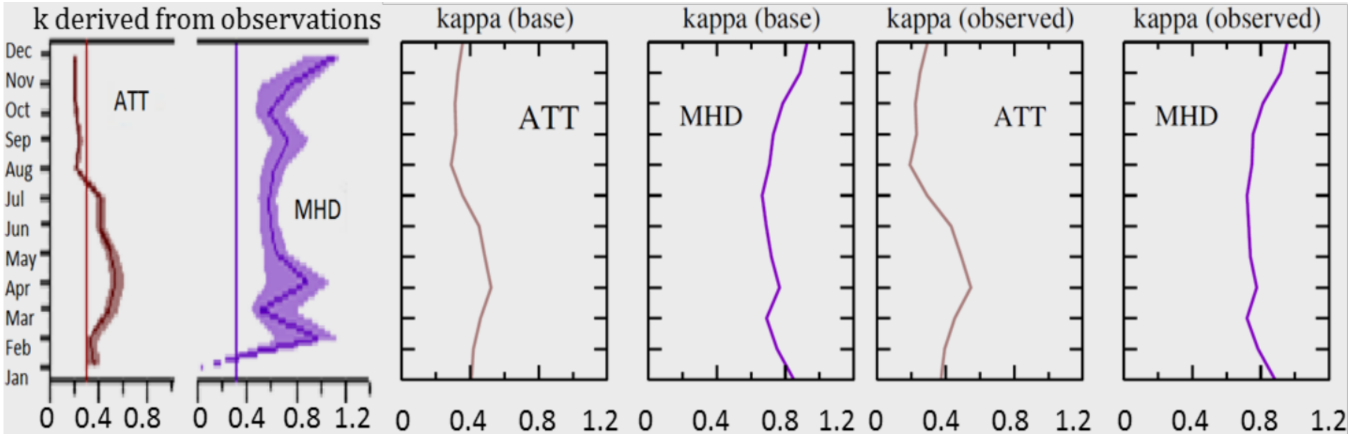
κ_{sulf}=0.72, κ_{SS}=1.31, κ_{POM}=0.1 (Observ.)

κ_{sulf}=0.61, κ_{SS}=1.28, κ_{POM}=0.1

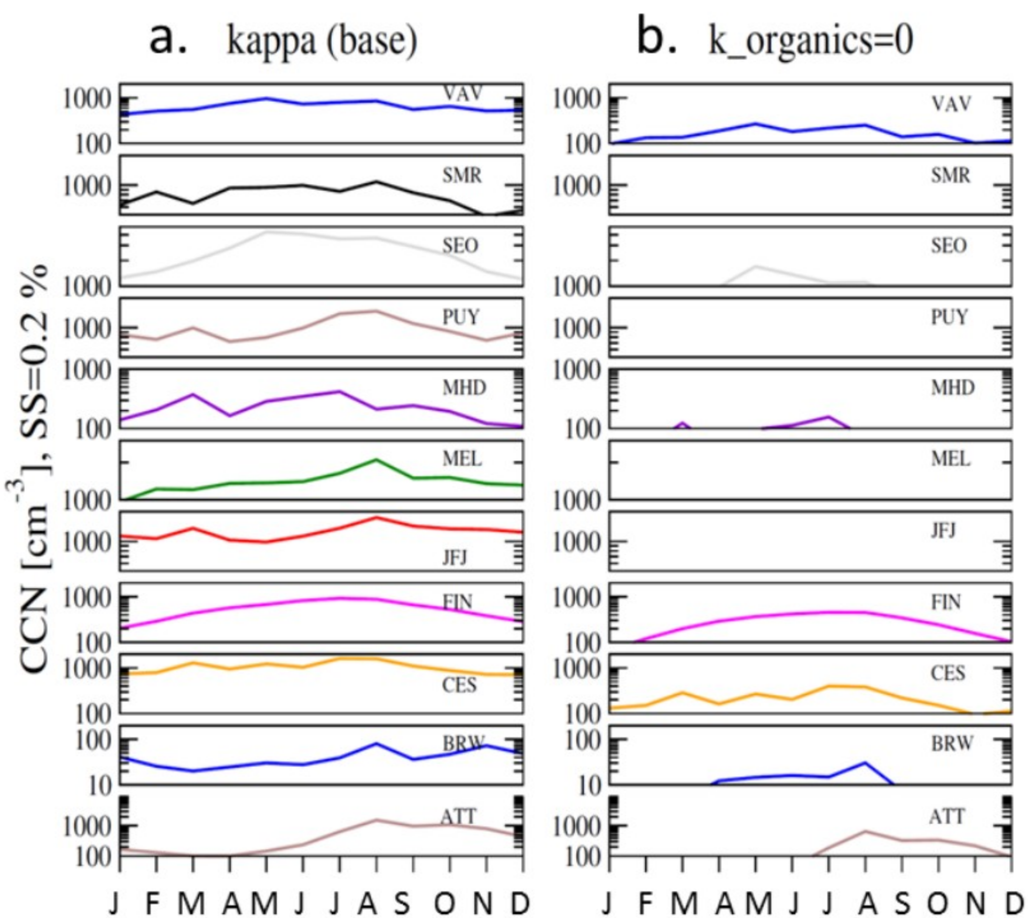




Seasonal variations of hygroscopicity



CCNs (SS=0.2%) as a function of components and hygroscopicity



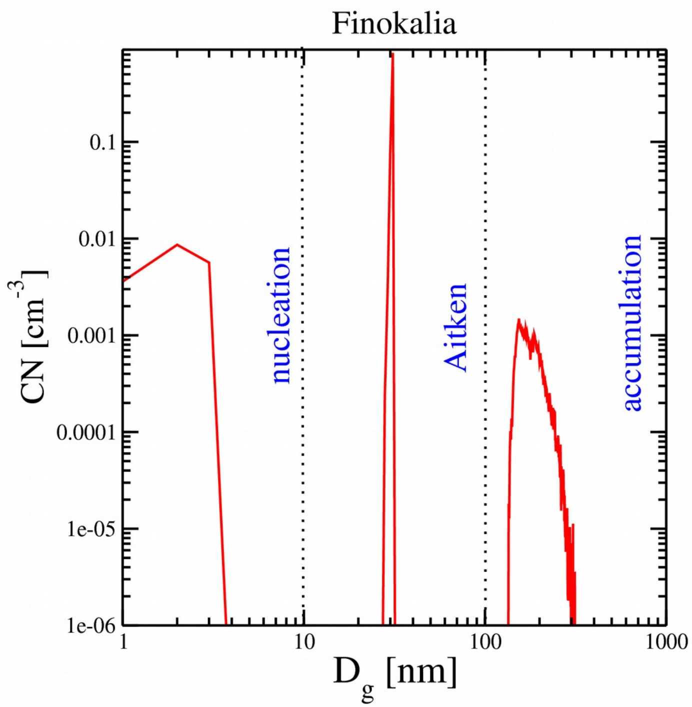
Limitations of the M7 model

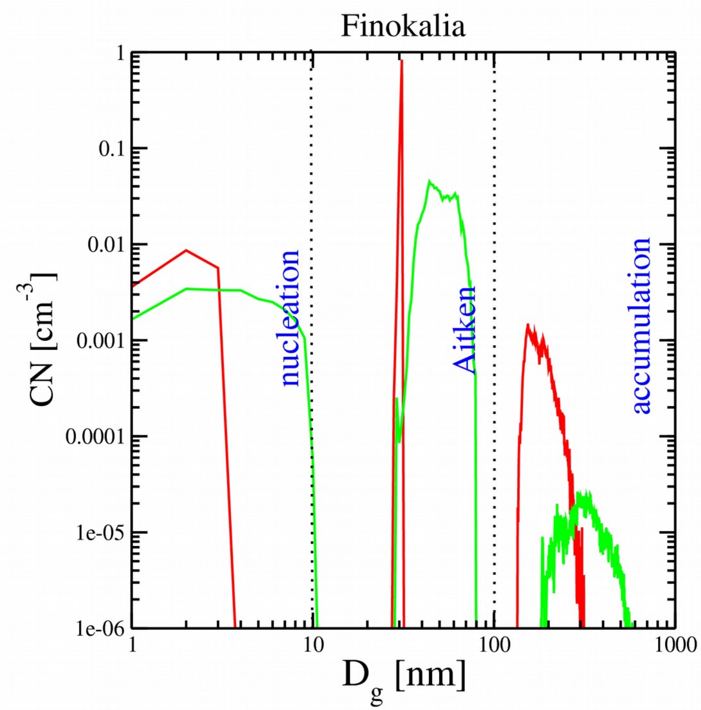
- Despite the agreement with observations in the prediction of CCNs, M7 over-predicts, particle numbers, activation ratios etc.
- Median Diameters of the Lognormal distributions describing aerosol numbers in each mode are allowed to vary within a very narrow range of values. Its maximum value is determined:

$$D_{\text{med}}^{\text{max}} = \sqrt{D_i D_{i+1}}$$

$$(\text{ i.e. } D_{\text{max}}^{\text{nuc}} = 3.16\,nm\,,\, D_{\text{max}}^{\text{ait}} = 31.6\,nm\,,\, D_{\text{max}}^{\text{acc}} = 316\,nm)$$

- Masses and numbers transferred from mode *i* to the *i+1* have diameters $D_{med} < D_{i+1}$





$$D_{\text{med}}^{\text{max}} = D_{i+1}$$

G. W. Mann *et. al.* GMD (2010) – GLOMAP model

Conclusions

- Simulations with high-resolution (3x2 or higher) are needed for accurate estimation of CCNs
- The predictions of TM4-ECPL and M7 model for CCNs are in agreement with the observations
- Hygroscopicity parameters are essential for the accurate representation of the observations
- the presence of organics can be critical for the CCN properties
- Mode-merging procedure of the M7 model significantly affects evolution of particle sizes, restricting their distribution within very narrow size intervals

Future work

- Study of the impact of primary biogenic aerosols in the CCN and IN formation
- Mode-merging description of the M7 model will be revisited

Thank you!