Contribution of primary emissions, secondary organic aerosol and nucleation on global aerosol number concentrations in NorESM

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Atmospheric aerosol number concentrations are sensitive to new particle formation (Makkonen et al., 2009) and primary emissions (Reddington et al., 2011). Some agreement between observations and global simulations of aerosol number can be found (Spracklen et al., 2010), but significant uncertainties still persist in aerosol and precursor emissions, as well as relevant processes.

Here, the Norwegian Earth System Model (NorESM, Kirkevåg et al. 2013) is used to model aerosol number concentrations. We model biogenic secondary organic aerosol (SOA) formation from monoterpene oxidation products. 50 % of the monoterpene ozonolysis product are assumed to contribute to the growth of nucleated particles, rest of the SOA is modeled as particles with an initial diameter of 80 nm. Binary sulfuric acid-water nucleation is active throughout the atmosphere, while activation-type nucleation is included in the boundary layer.

We have run the model with two sets of prescribed black carbon (BC) emission diameter for fossil fuel (24 and 48 nm), as well as with and without nucleation or SOA formation. We analyse the corresponding changes in the total aerosol number concentrations and contributions from the externally mixed aerosol modes (Fig. 1). We also show the sensitivity of cloud droplet number concentrations and aerosol indirect effect to the selected parameters.

In a simulation without nucleation, decreasing the BC emission size from 48 to 24 nm increases the global aerosol number concentration by 360 cm^{-3} (60 %). However, the concentration increase is smaller in magnitude (270 cm^{-3}) when nucleation is included in the model: the increased number of BC particles acts as a sink for both small nuclei and sulfuric acid.

Turning on nucleation has a significant effect on aerosol number. The increase in number concentration is 560 cm^{-3} (470 cm^{-3}) when BC emission size is set to 48 nm (24 nm). SOA formation increases number concentrations by 15-30 % globally, depending on BC size. SOA formation actually decreases the nucleation rates (by increasing the condensation and coagulation sinks), but nuclei survival probabilities are increased due to more rapid particle growth. The particle number increase due to SOA formation is ~75% smaller if nucleation is not accounted for. SOA formation also increases the cloud droplet number concentration (CDNC) by 5 % globally.

Even increased aerosol number concentrations might lead to decreased concentrations of cloud condensation nuclei. When using a smaller size for BC, CNDC is decreased by ~1 %. Despite doubling the total aerosol number concentration, turning on nucleation can decrease CDNC by almost 5 %. Decrease in CDNC can be due to e.g. increased competition of sulfuric acid condensation, leading to slower growth rates.

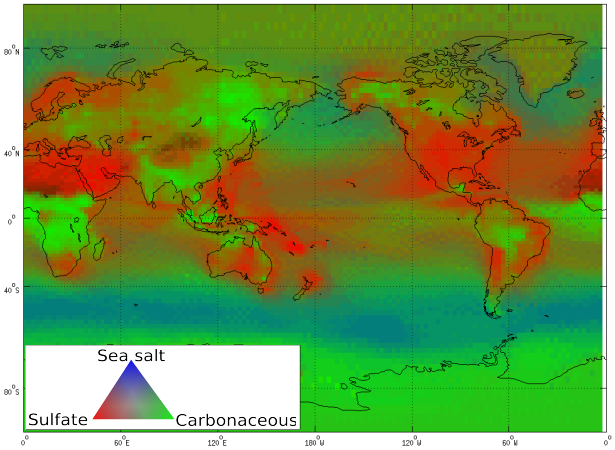

Figure 1. Relative contribution of sulfate (red), black and organic carbon (green) and sea salt (blue) to aerosol number concentration based on 5-year simulation.

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