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Evaluation of a global aerosol microphysics model against size-resolved particle statistics in the marine atmosphere

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Abstract. A statistical synthesis of marine aerosol measurements from experiments in four different oceans is used to evaluate a global aerosol microphysics model (GLOMAP). We compare the model against observed size resolved particle concentrations, probability distributions, and the temporal persistence of different size particles. We attempt to explain the observed sub-micrometre size distributions in terms of sulfate and sea spray and quantify the possible contributions of anthropogenic sulfate and carbonaceous material to the number and mass distribution. The model predicts a bimodal size distribution that agrees well with observations as a grand average over all regions, but there are large regional differences. Notably, observed Aitken mode number concentrations are more than a factor 10 higher than in the model for the N Atlantic but a factor 7 lower than the model in the NW Pacific. We also find that modelled Aitken mode and accumulation mode geometric mean diameters are generally smaller in the model by 10–30%. Comparison with observed free tropospheric Aitken mode distributions suggests that the model underpredicts growth of these particles during descent to the marine boundary layer (MBL). Recent observations of a substantial organic component of free tropospheric aerosol could explain this discrepancy. We find that anthropogenic continental material makes a substantial contribution to N Atlantic MBL aerosol, with typically 60–90% of sulfate across the particle size range coming from anthropogenic sources, even if we analyse air that has spent an average of >120 h away from land. However, anthropogenic primary black carbon and organic carbon particles (at the emission size and quantity assumed here) do not explain the large discrepancies in Aitken mode number. Several explanations for the discrepancy are suggested. The lack of lower atmospheric particle formation in the model may explain low N Atlantic particle concentrations. However, the observed and modelled particle persistence at Cape Grim in the Southern Ocean, does not reveal a diurnal cycle consistent with a photochemically driven local particle source. We also show that a physically based cloud drop activation scheme better explains the observed change in accumulation mode geometric mean diameter with particle number.

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