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Simulation of particle size distribution with a global aerosol model: contribution of nucleation to aerosol and CCN number concentrations

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Abstract. An advanced particle microphysics model with a number of computationally efficient schemes has been incorporated into a global chemistry transport model (GEOS-Chem) to simulate particle number size distributions and cloud condensation nuclei (CCN) concentrations in the atmosphere. Size-resolved microphysics for secondary particles (i.e., those formed from gaseous species) and sea salt has been treated in the present study. The growth of nucleated particles through the condensation of sulfuric acid vapor and equilibrium uptake of nitrate, ammonium, and secondary organic aerosol is explicitly simulated, along with the scavenging of secondary particles by primary particles (dust, black carbon, organic carbon, and sea salt). We calculate secondary particle formation rate based on ion-mediated nucleation (IMN) mechanism and constrain the parameterizations of primary particle emissions with various observations. Our simulations indicate that secondary particles formed via IMN appear to be able to account for the particle number concentrations observed in many parts of the troposphere. A comparison of the simulated annual mean concentrations of condensation nuclei larger than 10 nm (CN10) with those measured values show very good agreement (within a factor of two) in near all 22 sites around the globe that have at least one full year of CN10 measurements. Secondary particles appear to dominate the number abundance in most parts of the troposphere. Calculated CCN concentration at supersaturation of 0.4% (CCN0.4) and the fraction of CCN0.4 that is secondary (f^{sec}_{CCN}) have large spatial variations. Over the middle latitude in the Northern Hemisphere, zonally averaged CCN0.4 decreases from ~400–700 cm^{-3} in the boundary layer (BL) to below 100 cm^{-3} above altitude of ~4 km, the corresponding f^{sec}_{CCN} values change from 50–60% to above ~70%. In the Southern Hemisphere, the zonally averaged CCN0.4 is below 200 cm^{-3} and f^{sec}_{CCN} is generally above 60% except in the BL over the Southern Ocean.

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