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A global off-line model of size-resolved aerosol microphysics: I. Model development and prediction of aerosol properties

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Abstract. A GLObal Model of Aerosol Processes (GLOMAP) has been developed as an extension to the TOMCAT 3-D Eulerian off-line chemical transport model. GLOMAP simulates the evolution of the global aerosol size distribution using a sectional two-moment scheme and includes the processes of aerosol nucleation, condensation, growth, coagulation, wet and dry deposition and cloud processing. We describe the results of a global simulation of sulfuric acid and sea spray aerosol. The model captures features of the aerosol size distribution that are well established from observations in the marine boundary layer and free troposphere. Modelled condensation nuclei (CN>3nm) vary between about 250–500 cm^{-3} in remote marine boundary layer regions and are generally in good agreement with observations. Modelled continental CN concentrations are lower than observed, which may be due to lack of some primary aerosol sources or the neglect of nucleation mechanisms other than binary homogeneous nucleation of sulfuric acid-water particles. Remote marine CN concentrations increase to around 2000–10 000 cm (at standard temperature and pressure) in the upper troposphere, which agrees with typical observed vertical profiles. Cloud condensation nuclei (CCN) at 0.2% supersaturation vary between about 1000 cm^{-3} in polluted regions and between 10 and 500 cm^{-3} in the remote marine boundary layer. New particle formation through sulfuric acid-water binary nucleation occurs predominantly in the upper troposphere, but the model results show that these particles contribute greatly to aerosol concentrations in the marine boundary layer. For this sulfur-sea salt system it is estimated that sea spray emissions account for only ~10% of CCN in the tropical marine boundary layer, but between 20 and 75% in the mid-latitude Southern Ocean. In a run with only natural sulfate and sea salt emissions the global mean surface CN concentration is more than 60% of that from a run with 1985 anthropogenic sulfur emissions, although the natural emissions comprise only 27% of total sulfur emissions. Southern hemisphere marine boundary layer CN are more than 90% natural in origin, while polluted continental CN are more than 90% anthropogenic in origin, although these numbers will change when other anthropogenic CN sources are included in the model.

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