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A global off-line model of size-resolved aerosol microphysics: II. Identification of key uncertainties

D. V. Spracklen, K. J. Pringle, K. S. Carslaw, M. P. Chipperfield, and G. W. Mann
Institute for Atmospheric Science, School of Earth and Environment, University of Leeds, UK

Abstract. We use the new GLOMAP model of global aerosol microphysics to investigate the sensitivity of modelled sulfate and sea salt aerosol properties to uncertainties in the driving microphysical processes and compare these uncertainties with those associated with aerosol and precursor gas emissions. Overall, we conclude that uncertainties in microphysical processes have a larger effect on global sulfate and sea salt derived condensation nuclei (CN) and cloud condensation nuclei (CCN) concentrations than uncertainties in present-day sulfur emissions. Our simulations suggest that uncertainties in predicted sulfate and sea salt CCN abundances due to poorly constrained microphysical processes are likely to be of a similar magnitude to long-term changes in sulfate and sea salt CCN due to changes in anthropogenic emissions. A microphysical treatment of the global sulfate aerosol allows the uncertainty in climate-relevant aerosol properties to be attributed to specific processes in a way that has not been possible with simpler aerosol schemes. In particular we conclude that: (1) changes in the binary H_2SO_4-H_2O nucleation rate and condensation rate of gaseous H_2SO_4 cause a shift in the vertical location of the upper tropospheric CN layer by as much as 3 km, while the shape of the CN profile is essentially pre-served (2) uncertainties in the binary H_2SO_4-H_2O nucleation rate have a relatively insignificant effect on marine boundary layer (MBL) aerosol properties; (3) emitting a fraction of anthropogenic SO_2 as particulates (to represent production of sulfate particles in power plant plumes below the scale of the model grid (which is of the order of 300 km)) has the potential to change the global mean MBL sulfate-derived CN concentrations by up to 72%, and changes of up to a factor 20 can occur in polluted continental regions; (4) predicted global mean MBL sulfate and sea salt CCN concentrations change by 10 to 60% when several microphysical processes are changed within reasonable uncertainty ranges; (5) sulfate and sea salt derived CCN concentrations are particularly sensitive to primary particle emissions, with global mean MBL sulfate and sea salt CCN changing by up to 27% and local concentrations over continental regions changing by more than 100% when the percentage of anthropogenic SO_2 emitted as particulates is changed from 0 to 5%; (6) large changes in sea spray flux have insignificant effects on global sulfate aerosol except when the mass accommodation coefficient of sulfuric acid on the salt particles is set unrealistically low.

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