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SENSITIVITY OF CONCENTRATION OF ACCUMULATION-MODE AEROSOL PARTICLES TO THE REPRESENTATION OF NEW PARTICLE FORMATION AND PARTICLE EMISSIONS IN CHEMICAL TRANSPORT MODELS

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ABSTRACT

The two sources of particle number concentration, primary particle emissions and secondary new particle formation (NPF) from gaseous precursors, are both subject to large uncertainties. The derivation of number emission rates from inventory mass emission rates requires assumption of an average emitted particle size. Theoretical nucleation rates necessary to calculate ultrafine particle formation rates vary widely among mechanisms and parameterizations. With modal aerosol size representations, calculation of cloud condensation nucleus number concentration is typically based on accumulation mode number concentration (N_{acc}) and is thus sensitive to these uncertainties. In this study sensitivity studies were performed with a variety of approaches to NPF and several number emission rates, using the Community Multiscale Air Quality (CMAQ) regional-scale model over the continental U.S. with 60-km resolution for July 2004. As intermodal transfer from the Aitken to the accumulation mode (IMTR) is necessary in modal models to maintain distinct modes, and as transfer is not governed by a physical process, large NPF rates introduces a substantial uncertainty in accumulation mode number. Reducing the mean diameter of emitted particles by a factor of 2 (for fixed mass emission rate) for both the Aitken and accumulation modes gave a modest decrease in both NPF rate and IMTR but a substantial increase in N_{acc}. This results from increased surface area and reduced NPF when emitted mass is apportioned into smaller particles. When the NPF rate is large, the subsequent reduction of NPF rate due to limited H_2SO_4 was also large when using a 15-min time step. Nucleation and condensation are partially operator-split in CMAQ and when its binary nucleation rate (Vehkamaki *et al.*, 2002) is replaced by the ternary rate (Napari *et al.*, 2002), H_2SO_4 consumption is biased in favor of NPF. In the Aitken mode, average NPF and coagulation rates are nearly balanced. In the accumulation mode, number emissions and intermodal transfer are balanced by wet deposition.