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A hybrid formalism of aerosol gas phase interaction for 3-D global models

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Aerosol chemical composition is a relevant factor to the global climate system with respect to both atmospheric chemistry and the aerosol direct and indirect effects. Aerosol chemical composition determines the capacity of aerosol particles to act as cloud condensation nuclei both explicitly via particle size and implicitly via the aerosol hygroscopic property. Due to the primary role of clouds in the climate system and the sensitivity of cloud formation and radiative properties to the cloud droplet number it is necessary to determine with accuracy the chemical composition of the aerosol. Dissolution, although a formally fairly well known process, may be subject to numerically prohibitive properties that result from the chemical interaction of the species engaged. So-far approaches to model the dissolution of inorganics into the aerosol liquid phase in the framework of a 3-D global model were based on an equilibrium, transient or hybrid equilibrium-transient approach. All of these methods present the disadvantage of a priori assumptions with respect to the mechanism and/or are numerically not manageable in the context of a global climate system model.

In this paper a new hybrid formalism to aerosol gas phase interaction is presented within the framework of the H2SO4/HNO3/HCl/NH3 system and a modal approach of aerosol size discretisation. The formalism is distinct from prior hybrid approaches in as much as no a priori assumption on the nature of the regime a particular aerosol mode is in is made. Whether a particular mode is set to be in the equilibrium or the transitory regime is continuously determined during each time increment against relevant criteria considering the estimated equilibration time interval and the interdependence of the aerosol modes relative to the partitioning of the dissolving species. Doing this the aerosol composition range of numerical stiffness due to species interaction during transient dissolution is effectively eluded, and the numerical expense of dissolution in the transient regime is reduced through the minimisation of the number of modes in this regime and a larger time step. Containment of the numerical expense of the modes in the equilibrium regime is ensured through the usage of either an analytical equilibrium solver that requires iteration among the equilibrium modes, or a simple numerical solver based on a differential approach that requires iteration among the chemical species. Both equilibrium solvers require iteration over the water content and the activity coefficients. Decision for using either one or the other solver is made upon the consideration of the actual equilibrating mechanism, either chemical interaction or gas phase partial pressure variation, respectively. The formalism should thus ally appropriate process simplification resulting in reasonable computation time to a high degree of real process conformity as it is ensured by a transitory representation of dissolution. The resulting effectiveness and limits of the formalism are illustrated with numerical examples.