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Kinetic model framework for aerosol and cloud surface chemistry and gas-particle interactions – Part 2: Exemplary practical applications and numerical simulations

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Abstract. A kinetic model framework with consistent and unambiguous terminology and universally applicable rate equations and parameters for aerosol and cloud surface chemistry and gas-particle interactions has been presented in the preceding companion paper by Pöschl, Rudich and Ammann (Pöschl et al., 2007), abbreviated PRA. It allows to describe mass transport and chemical reaction at the gas-particle interface and to link aerosol and cloud surface processes with gas phase and particle bulk processes. Here we present multiple exemplary model systems and calculations illustrating how the general mass balance and rate equations of the PRA framework can be easily reduced to compact sets of equations which enable a mechanistic description of time and concentration dependencies of trace gas uptake and particle composition in systems with one or more chemical components and physicochemical processes.

Time-dependent model scenarios show the effects of reversible adsorption, surface-bulk transport, and chemical aging on the temporal evolution of trace gas uptake by solid particles and solubility saturation of liquid particles. They demonstrate how the transformation of particles and the variation of trace gas accommodation and uptake coefficients by orders of magnitude over time scales of microseconds to days can be explained and predicted from the initial composition and basic kinetic parameters of model systems by iterative calculations using standard spreadsheet programs. Moreover, they show how apparently inconsistent experimental data sets obtained with different techniques and on different time scales can be efficiently linked and mechanistically explained by application of consistent model formalisms and terminologies within the PRA framework.

Steady-state model scenarios illustrate characteristic effects of gas phase composition and basic kinetic parameters on the rates of mass transport and chemical reactions. They demonstrate how adsorption and surface saturation effects can explain non-linear gas phase concentration dependencies of surface and bulk accommodation coefficients, uptake coefficients, and bulk solubilities (deviations from Henry's law). Such effects are expected to play an important role in many real atmospheric aerosol

and cloud systems involving a wide range of organic and inorganic components of concentrated aqueous and organic solution droplets, ice crystals, and other crystalline or amorphous solid particles.

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