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Kinetic Model Framework for Aerosol and Cloud Surface Chemistry and Gas-Particle Interactions

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Aerosols and clouds play central roles in atmospheric chemistry and physics, climate, air pollution, and public health. The mechanistic understanding and predictability of aerosol and cloud properties, interactions, transformations, and effects are, however, still very limited. This is due not only to the limited availability of measurement data, but also to the limited applicability and compatibility of model formalisms used for the analysis, interpretation, and description of heterogeneous and multiphase processes (Pöschl, 2005; and references therein).

To support the investigation and elucidation of atmospheric aerosol and cloud surface chemistry and gas-particle interactions, we present a comprehensive kinetic model framework with consistent and unambiguous terminology and universally applicable rate equations and parameters. It allows to describe mass transport and chemical reactions at the gas-particle interface and to link aerosol and cloud surface processes with gas phase and particle bulk processes in systems with multiple chemical components and competing physicochemical processes.

The key elements and essential aspects of the presented framework are: a simple and descriptive double-layer surface model (sorption layer and quasi-static layer); straightforward flux-based mass balance and rate equations; clear separation of mass transport and chemical reactions; well-defined rate parameters (uptake and accommodation coefficients, reaction and transport rate coefficients); clear distinction between gas phase, gas-surface, and surface-bulk transport (gas phase diffusion correction, surface and bulk accommodation); clear distinction between gas-surface, surface layer, and surface-bulk reactions (Langmuir-Hinshelwood and Eley-Rideal mecha-

nisms); mechanistic description of concentration and time dependencies; flexible inclusion/omission of chemical species and physicochemical processes; flexible convolution/deconvolution of species and processes; and full compatibility with traditional resistor model formulations.

We expect that the presented model framework will serve as a useful tool and basis for experimental and theoretical studies investigating and describing atmospheric aerosol and cloud surface chemistry and gas-particle interactions. In particular, it is meant to support the planning and design of laboratory experiments for the elucidation and determination of kinetic parameters; the establishment, evaluation, and quality assurance of comprehensive and self-consistent collections of rate parameters; and the development of detailed master mechanisms for process models and the derivation of simplified but yet realistic parameterizations for atmospheric and climate models.

The full formalism and exemplary practical applications and model calculations illustrating the relevance of the above aspects have been presented in several recent publications (Ammann et al., 2003; Pöschl et al., 2005; Ammann and Pöschl, 2005). The key aspects and implications for atmospheric research will be highlighted in the conference presentation.

References:

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