Atmospheric Chemistry and Physics
An Interactive Open Access Journal of the European Geosciences Union

Atmos. Chem. Phys., 7, 5989-6023, 2007
www.atmos-chem-phys.net/7/5989/2007/

Kinetic model framework for aerosol and cloud surface chemistry and gas-particle interactions – Part 1: General equations, parameters, and terminology

U. Pöschl[1,*], Y. Rudich[2], and M. Ammann[3]
[1]Technical University of Munich, Institute of Hydrochemistry, 81377 Munich, Germany
[2]Department of Environmental Sciences, Weizmann Institute of Science, Rehovot 76100, Israel
[3]Paul Scherrer Institute, 5232 Villigen PSI, Switzerland
[*]now at: Max Planck Institute for Chemistry, Biogeochemistry Department, 55128 Mainz, Germany

Abstract. 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. 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 enables a detailed description of mass transport and chemical reactions at the gas-particle interface, and it allows linking 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 and consistent 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, surface and bulk accommodation); clear distinction between gas-surface, surface layer, and surface-bulk reactions (Langmuir-Hinshelwood and Eley-Rideal mechanisms); mechanistic description of concentration and time dependences (transient and steady-state conditions); flexible addition of unlimited numbers of chemical species and physicochemical processes; optional aggregation or resolution of intermediate species, sequential processes, and surface layers; and full compatibility with traditional resistor model formulations. The outlined double-layer surface concept and formalisms represent a minimum of model complexity required for a consistent description of the non-linear concentration and time

dependences observed in experimental studies of atmospheric multiphase processes (competitive co-adsorption and surface saturation effects, etc.). Exemplary practical applications and model calculations illustrating the relevance of the above aspects are presented in a companion paper (Ammann and Pöschl, 2007). 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. It shall help to end the "Babylonian confusion" that seems to inhibit scientific progress in the understanding of heterogeneous chemical reactions and other multiphase processes in aerosols and clouds. In particular, it shall support the planning and design of laboratory experiments for the elucidation and determination of fundamental 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 derivation of simplified but yet realistic parameterizations for atmospheric and climate models.

Final Revised Paper (PDF, 853 KB) Discussion Paper (ACPD)

Citation: Pöschl, U., Rudich, Y., and Ammann, M.: Kinetic model framework for aerosol and cloud surface chemistry and gas-particle interactions – Part 1: General equations, parameters, and terminology, Atmos. Chem. Phys., 7, 5989-6023, 2007. Bibtex EndNote Reference Manager