



Gas–particle interactions of tropospheric aerosols: Kinetic and thermodynamic perspectives of multiphase chemical reactions, amorphous organic substances, and the activation of cloud condensation nuclei

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ABSTRACT

Aerosols are of central importance in the Earth system, influencing the atmosphere, biosphere, climate, and public health. This article outlines recent advances and perspectives in the investigation and effects of gas–particle interactions in tropospheric aerosols, including physical, chemical and biological aspects. In particular, it addresses how multiphase processes and heterogeneous chemical reactions can be efficiently described by kinetic models, how amorphous organic substances can influence the kinetics of water uptake and phase transitions, and how the abundance and activation of cloud condensation nuclei (CCN) depend on thermodynamic and kinetic parameters.

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1. Introduction

The effects of aerosols on the atmosphere, biosphere, climate, and public health are among the central topics in current environmental research. Aerosol particles scatter and absorb radiation, they are involved in the formation of clouds and precipitation as cloud condensation and ice nuclei, and they affect the abundance and distribution of atmospheric trace gases by heterogeneous chemical reactions and other multiphase processes. Moreover, airborne particles play an important role in the spreading of biological organisms and ecosystems, and they can cause or enhance human, animal and plant diseases (Fig. 1, Pöschl, 2005).

Atmospheric aerosols originate from a wide variety of natural and anthropogenic sources. Primary particles are directly emitted as liquids or solids from sources such as biomass burning, incomplete combustion of fossil fuels, volcanic eruptions, and wind-driven or traffic-related suspen-

sion of dust, sea salt, and biological materials (bacterial, fungal spores, pollen, plant fragments, etc.). Secondary particles, on the other hand, are formed by gas-to-particle conversion in the atmosphere (new particle formation by nucleation and condensation of gaseous precursors). Moreover, airborne particles and particulate matter (PM) continuously undergo physical and chemical interactions and transformations (Finlayson-Pitts and Pitts, 2000; Pöschl, 2005; Fuzzi et al., 2006; Seinfeld and Pandis, 2006; Solomon et al., 2007; Finlayson-Pitts, 2009; Hallquist et al., 2009; Jimenez et al., 2009; Monks et al., 2009; George and Abbatt, 2010; Kolb et al., 2010; and references therein).

As illustrated in Figs. 2 and 3, the formation and transformation (aging) of atmospheric aerosol particles and components generally proceed via multiple steps of mass transport and chemical reaction in and between the gas phase, molecular clusters, aerosol, cloud, and precipitation particles (multiphase processes and heterogeneous reactions). The combination of gas and liquid diffusion, reversible and competitive adsorption, and surface and bulk reactions usually leads to non-linear concentration and time dependencies and limits the applicability of linear extrapolations and steady-state assumptions (Ammann and Pöschl, 2007; Pöschl et al., 2007; Kolb et al.,

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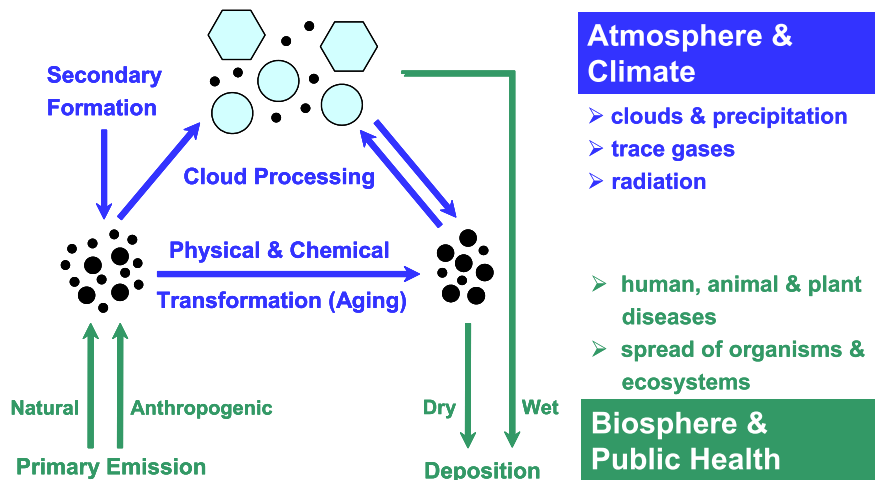


Fig. 1. Atmospheric aerosol cycling and effects (Pöschl, 2005; Fuzzi et al., 2006).

2010). This article outlines recent advances in the modeling of multiphase chemical kinetics (Section 2); in the understanding of water uptake and phase transitions of amorphous organic substances (Section 3); and in predicting the abundance and activation of cloud condensation nuclei from thermodynamic and kinetic parameters (CCN, Section 4).

2. Multiphase chemical kinetic models

The mechanistic understanding of atmospheric aerosol transformation and aging is limited not only by the availability of measurement data but also by the limited applicability and compatibility of model formalisms used for the analysis, interpretation, and description of heterogeneous and multiphase processes (“Babylonian confusion” of terms and parameters; Pöschl et al., 2007; Kolb et al., 2010). To support efficient investigation and elucidation of aerosol surface chemistry and gas–particle interactions, Pöschl et al. (2007), abbreviated PRA, presented a comprehensive kinetic model framework with consistent and unambiguous terminology and universally applicable rate equations and parameters. The PRA framework 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 PRA 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 double-layer surface concept and formalisms of the PRA framework 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.).

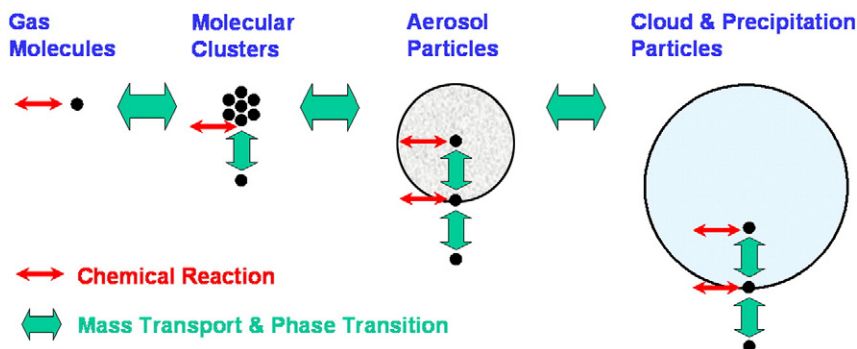


Fig. 2. Atmospheric aerosol transformation: chemical reaction, mass transport and phase transition (Pöschl, 2005; Fuzzi et al., 2006).

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