

Barbara Ervens^{1,*}. (1) Cooperative Institute for Research in Environmental Sciences, University of Colorado, Boulder, CO, USA, (2) NOAA/ESRL, Chemical Sciences Division, Boulder, CO, USA; *barbara.ervens@noaa.gov.

Water plays an important role in the atmosphere in form of cloud, fog or rain droplets and associated with aerosol particles. In this aqueous phase, unique chemical reactions occur that often lead to different products and take place on different time scales than in the surrounding air [1]. Examples include the oxidation of sulfur dioxide (SO₂) to sulfate (SO₄²⁻), the major contributor to acid rain or the formation of secondary organic aerosol in water (aqSOA). Implementing chemical aqueous phase processes in models is a great challenge as often the resolution of models is much coarser than the size of individual clouds and/or the small spatial and temporal scales, on which particle composition and water content change, cannot be resolved. Thus, computationally efficient parameterizations are needed to allow the description of such processes in large scale models.

There is general agreement that in-cloud formation is globally the major sulfate source. However, the implementation of sulfate formation processes among models greatly differs in complexity and accuracy. I will briefly discuss different approaches as included in global and regional models.

Organic mass is another significant contributor to atmospheric particle loading. Most organic aerosol is secondary, i.e. formed from gases in the atmosphere. Traditionally, secondary organic aerosol (SOA) has been described as condensation of low volatility gases on pre-existing particles. Over the past 15 years, evidence from model, laboratory and field studies has shown that secondary organic aerosol can be also formed in the aqueous phase of clouds and particles (aqSOA) [2]. I will highlight process model studies that demonstrate the ability of models to explain the presence of aqSOA compounds in clouds. Oxalate can often be considered a tracer compounds for such processes; I will contrast scenarios where this assumption is valid to those where aqueous processing is occurring, but oxalate cannot be detected.

The representation of aqSOA formation in deliquesced aerosol particles is much more

complex and difficult as the aqueous phase is not dilute and, thus, approaches for ideal solutions, such as Henry's law, do not apply. I will give an overview of current assumptions and approaches that are used in models that describe aqSOA formation in aerosol water. In addition, I will discuss chemical mechanisms in aerosol particles that lead to products that have a higher molecular weight than the initial reactants. Such accretion or oligomerisation reactions are unique to aerosol water where high organic concentrations are present.

Many of the organic reactions in the atmospheric aqueous phase are initiated by the OH radical. Thus, an understanding of OH concentrations, sources and sinks in the aqueous phase is important to accurately model aqSOA formation. The direct uptake of OH from the gas phase is a major OH source in the aqueous phase. Since OH is only moderately soluble but highly reactive in the aqueous phase, its reaction rates show an apparent dependence on the drop surface [3]. As the implementation of cloud droplet distributions in models is often not feasible due to model resolution and/or lack of observational input data, a simplified approach is needed that both represent the size-resolved OH concentrations, and consequently, aqSOA formation rates, but is sufficiently simple for models. I will show how a limited set of microphysical parameters can be used to parameterize OH concentrations in clouds and thus to improve the representation of aqSOA formation [4].

References

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