CCN activity of multi-component organic particles: The role of the water solubility distribution

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Introduction

Interactions of atmospheric aerosol particles with the ambient water vapour determine to a large extent the influence that aerosols have on climate. To pin down the climate effects of aerosol particles on clouds and climate it is thus necessary to know how much they absorb water at sub-saturated conditions and at which conditions they can activate as CCN and form cloud droplets. The solubility in water is one of the key properties governing the water-absorption and CCN activation behaviour of aerosol particles.

Organic constituents contribute a large fraction (20-90%, depending on the environment) of atmospheric submicron particulate mass which is the part of the aerosol size distribution that typically dominates the CCN numbers. Atmospheric organic compounds have a wide range of solubilities, spanning from practically insoluble material to highly water soluble compounds (e.g. Raymond and Pandis 2003). To accurately predict the water content and CCN activation of atmospheric OA information on the dissolution behaviour and aqueous phase interactions of these complex mixtures is needed.

We investigate the dissolution behaviour of complex organic mixtures and their CCN activity using a theoretical framework (Solubility Basis Set, SBS) representing the mixture components with a continuous distribution of solubilities, similar to the VBS (Donahue et al., 2006).

Method

In this study we consider a monodisperse population of spherical aerosol particles consisting of an internal mixture of organic compounds. When exposed to water vapour, these particles grow reaching a thermodynamic equilibrium between the water vapour and the particle phase. The wet particle is allowed to consist of maximum two phases: the insoluble organic phase and the aqueous phase. The compositions of the organic and aqueous phases are determined on one hand by the equilibrium between the aqueous phase and the water vapour, and on the other hand by the equilibrium of the aqueous phase with the organic insoluble phase.

We varied the range of pure component water solubilities present in the mixture, the shape of the solubility distribution, and the number of components n in the distribution, which gave us 72 different organic mixtures with varying solubility distributions. By assuming two different interactions between the organic compounds in the insoluble phase, 1) Interacting: organics limit each other's dissolution 2) Non-interacting: organics behave as pure compounds, we ended up with (72×2) organic mixtures. Critical supersaturations and the dissolution behaviour at the point of CCN activation were calculated utilizing the Köhler theory for all organic mixtures as a full model. The full model predictions were compared with a number of simple models (complete dissolution, soluble fraction (epsilon) and hygroscopicity parameter (kappa)).

Conclusions

Describing the mixture with single soluble fraction (epsilon) or hygroscopicity parameter (kappa) perform reasonably well on average in predicting the CCN activation. The complete dissolution model shows the poorest agreement with the full model.

Mass weighted average solubility of the mixture besides the solubility of the least soluble component determine the CCN behaviour of complex organic aerosols.