



Connecting the solubility and CCN activation of complex organic aerosols: A theoretical study using the Solubility Basis Set (SBS)

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INTRODUCTION

Organic aerosol particles often consist of thousands of compounds with different properties. One of these properties is solubility, which affects the hygroscopic growth and CCN activation of the organic particles. Here we investigate the CCN activation behavior of complex organic aerosols accounting for the distribution of solubilities present in these mixtures.

METHODS

We considered a monodisperse population of spherical aerosol particles consisting of an internal mixture of organic compounds. When exposed to water vapor, these particles were assumed to grow reaching a thermodynamic equilibrium between the water vapor and the particle phase. The composition of the organic and aqueous phases was determined on one hand by the equilibrium between the aqueous phase and water vapor, and on the other hand by the equilibrium of the aqueous phase with the organic insoluble phase. We modelled the mixtures with the help of a solubility basis set (SBS, analogous to the volatility basis set VBS, Donahue et al. 2006, 2011, 2012), describing the mixture with n surrogate compounds with varying solubilities. We varied the range and shape of the solubility distribution, and the number of components n in the distribution, we also assumed two different kinds of interactions between the organic compounds in the insoluble phase 1) ideal mixture, where organics limit each other's dissolution; 2) unity activity, where organics behave as pure compounds and do not influence each other's dissolution. Critical supersaturations and the dissolution behavior at the point of CCN activation were calculated utilizing the Köhler theory for all organic mixtures (denoted here as the "full model"). The full model predictions were compared with the three simplified models: 1) assuming complete dissolution of all compounds; 2) treating the organic mixture solubility with the hygroscopicity parameter κ and 3) assuming a fixed soluble fraction for each mixture.

RESULTS AND CONCLUSIONS

The comparison between the full model and the complete dissolution assumption demonstrated a systematic under-prediction (up to 40%) of the activation diameter, while the κ and ε -based solubility models were generally within 10% (in most cases within 5%) of the activation diameter predicted using the full solubility distribution representation. The ε and κ values were found to correspond to the fraction of material with solubilities larger than a given threshold solubility c_t . The median threshold assuming the organics to form an ideal mixture was 10 g L⁻¹, with most of the c_t values falling between 1 and 100 g L⁻¹. For the unity organic activity assumption this median threshold was 1 g L⁻¹, with most of the c_t values falling between 0.1 and 10 g L⁻¹. Our results suggest that the solubility range causing limited dissolution in CCN activation is between 0.1-100 g L⁻¹ but in most cases material below 1 g L⁻¹ is practically insoluble and material above 10 g L⁻¹ completely soluble upon CCN activation at atmospheric supersaturations.

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