Abstract

Atmospheric aerosols have an important impact on the radiation balance, and thus, on the climate of the Earth. Aerosol particles scatter and absorb incoming solar and terrestrial radiation. Apart from this direct effect, aerosol particles act as cloud condensation nuclei (CCN), thereby greatly influencing the microphysics of clouds. Secondary organic aerosols (SOA) are an important fraction of the total aerosol mass. In many environments these organic compounds are mainly products of the oxidation of biogenic volatile organic compounds (VOC).

In this study the hygroscopic growth and CCN activation of biogenic SOA were investigated which was formed by the oxidation of VOC with O_3 and photochemically formed OH radicals under low NO_x conditions. For this purpose, a complex mixture of VOC emitted by boreal tree species as gas-phase precursors was used in the Jülich Plant Atmosphere Chamber (JPAC). In long-term studies in the atmosphere simulation chamber SAPHIR α -pinene or a defined mixture of α -pinene, β -pinene, limonene, ocimene, Δ -3-carene served as precursors. Initial precursor concentrations between 40 and 1000 ppbC were investigated.

The observed SOA particles were slightly hygroscopic with an average hygroscopicity parameter $\kappa(\text{CCN}) = 0.10 \pm 0.02$ and $\kappa(90\% RH) = 0.05 \pm 0.01$. Closure between hygroscopic growth and CCN activation data could be achieved allowing either surface tension reduction, limited solubility, or non-ideality of the solution in the droplet. The SOA solutions in equilibrium with RH < 95% are possible highly non-ideal. Therefore the organic-water interaction were investigated by applying the UNIFAC model. Calculations for surrogate compounds exhibited the same strong concentration (i.e. RH) dependence of κ at sub-saturation. The growth curves could be fitted and CCN activation predicted by assuming a binary mixture of water and one hypothetical organic compound. The occurrence of functional groups in this compound was adjusted to reproduce the observed growth curves. However, further information on surface tension and the ratio of the molecular mass and density of the solute is needed to predict activation behavior from hygroscopic growth measurements.

A dependence of κ on the ratio of primarily produced OH to initial VOC level was observed. The higher κ values for low precursor concentrations could be attributed to a higher OH/VOC level. The detailed chemical composition of the gas-phase precursors had only little effect on κ . In long term experiments there was no significant effect of the observed chemical aging of the particles on κ . The observed low variability of κ for biogenic SOA particles simplifies their treatment in global models as an average value of $\kappa = 0.1$ can be used.