

Closure between Hygroscopic Growth and CCN Activation of Photochemical Aged Secondary Organic Aerosols

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The effects of organics on the hygroscopic properties of particles has not yet been fully understood. There is still a discrepancy between the observed cloud condensation nucleus (CCN) activation and hygroscopic growth measurements when predicting CCN activity with κ -Köhler theory following Petters & Kreidenweis (2007). In this study the ability of secondary organic aerosol (SOA) particles, produced from photooxidation and ozonolysis of different monoterpene mixtures (MT), to absorb water and activate to cloud droplets during aging was investigated. Hygroscopic growth measurements were performed with a HTDMA (RH < 97%, FZ Jülich) and LACIS-mobile (RH > 97%, IfT) while the CCN activity was measured with a CCNC (RH > 100%, DMT). All experiments took place at the SAPHIR-chamber at the research centre Jülich in summer 2008 under near atmospheric conditions such as natural sunlight, low precursor and O₃ concentrations (4-100 ppb) and long reaction times (2 days). As precursor we used a monoterpene mixture (cis-ocimene, limonene, α -pinene, β -pinene, Δ 3-carene). Alternatively, we added the sesquiterpenes trans-caryophyllene and farnesene. All mixtures had been identified as major constituents of plant emissions in previous experiments.

Differences in the hygroscopic growth and activation behaviour were found for different radiation intensities. At sunny days the SOA particles were more hygroscopic than on cloudy days. The weather effect was even stronger than the observed aging which takes mainly place during the first hours of an experiment. We also found that different precursor concentrations do not significantly effect the hygroscopic growth of the formed particles.

Tab. 1: κ -values were calculated from hygroscopic growth and activation measurements. The κ values derived from hygroscopic growth measurements were used to calculate the critical supersaturation (S_c) for activation, 08/06/10, 100 ppb MT, $D_{dry} = 150$ nm.

D_{dry} [nm]	κ	calculated S_c [%]	measured S_c [%]
150	minimum ¹	0.05	0.29
150	maximum ¹	0.11	0.20
150	average ¹	0.07	0.24
150	variable σ ²	0.05	0.23
154	activation	0.11	-
			0.2

¹calculated from hygroscopic growth measurements, $\sigma = 72$ mN m⁻¹

²calculated from hygroscopic growth and activation measurements, σ =variable (Ziese et al., 2008)

The hygroscopicity parameter κ following Petters & Kreidenweis (2007) was determined from all hygroscopic growth measurements. Tab. 1 shows the averaged, minimum and maximum κ -values. Afterwards the theoretical Köhler-curves were calculated with these κ (Fig. 1) using a constant surface tension of water. From this curves, values for the critical supersaturation (S_c) were derived, too. Also a method for calculating Köhler curves with a concentration dependent surface tension following Ziese et al. (2008) was utilized. The insert in figure 1 shows κ as a function of relative humidity (RH).

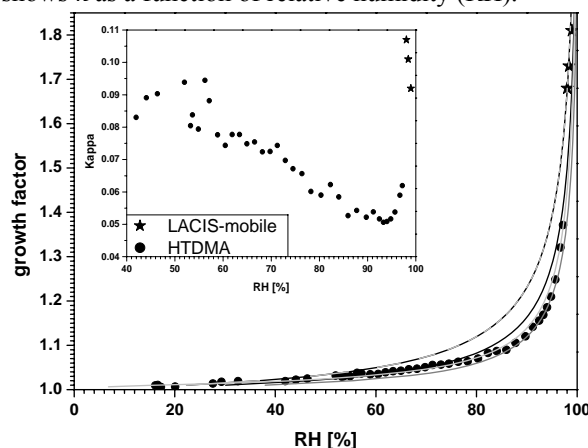


Fig. 1: Closure of CCNC and hygroscopic growth for a sunny day at the SAPHIR chamber, 08/06/10, 100 ppb MT, $D_{dry} = 150$ nm. Köhler curves for κ max and activation (grey-black dashed line), κ average (black line), κ min (dark grey line) and variable σ (light grey line).

At a fixed RH, the calculated growth factor increases with increasing κ . The calculated Köhler curves for the highest κ derived from hygroscopic growth measurements and from the activation measurements are identical. In contrast to this there is a gap between the averaged κ and the CCN activation. This shows that the assumption of a constant κ (Petters & Kreidenweis, 2007) is not valid if we are using a constant surface tension. Using the concentration dependent surface tension method following Ziese et al. (2008) for calculating the Köhler curve a closure between hygroscopic growth and CCNC activation can be achieved.

Ziese, M., et al. (2008). *Atmos. Chem. Phys.*, 8, 1855-1866.

Petters, M.D. & Kreidenweis, S.M. (2007). *Atmos. Chem. Phys.*, 7, 1961-1971.