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Closure between hygroscopic growth and CCN activity of secondary organic aerosol: using UNIFAC for water activity parametrization

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Knowledge of the microphysical properties (i.e. the hygroscopic growth and cloud condensation nuclei (CCN) activity) is important to understand the impact of aerosols on climate.

Generally, the microphysical properties of aerosols have to be measured separately for the sub- and super-saturated regime due to instrumental limitations. Below 100% relative humidity (RH) the hygroscopic growth factor (GF) is determined depending on particle size and RH e.g. with a Hygroscopicity Tandem Differential Analyzer. Above 100% RH the activation point is measured depending on the super-saturation (SS=RH-100%) and the size of the dry particle e.g. by combination of a CCN-Counter and a SMPS/CPC system. The Koehler theory describes the hygroscopic behavior of aerosol particles in both the sub- and super-saturated regime. A parametrization of this equation is necessary since several of the parameters (e.g. the water activity in the droplets) are unknown for atmospheric aerosols.

The κ parametrization of the water activity (Petters and Kreidenweis, 2007) can be used to compare the data for sub- and super-saturated conditions. In the presented study with secondary organic aerosol (SOA) from monoterpene photo-oxidation products, κ derived from CCN data was up to a factor of 3 higher than κ derived from GF data. There was a RH dependence of κ with a minimum between 90 and 95% RH. To explain the observed discrepancies, an additional parameter is necessary.

The Universal Quasi chemical Functional Group Activity Coefficients model (UNIFAC) allows predicting the water activity in an arbitrary mixture of water and organic compounds. A hypothetical organic molecule was determined whose functionality and corresponding κ(RH) reproduced the observed hygroscopic growth. Additionally, real molecules, which can be thought of as surrogates for organics in SOA, were calculated with UNIFAC. The exhibited the same general functionality in κ(RH) as the measured data with a distinct minimum between 90 and 95% RH.

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