Geophysical Research Abstracts
Vol. 15, EGU2013-12684, 2013
EGU General Assembly 2013

Mass-based hygroscopicity parameter interaction model and measurement of atmospheric aerosol water uptake

Eugene Mikhailov (1,2), Sergey Vlasenko (2), Diana Rose (1), and Ulrich Pöschl (1)

(1) Max Planck Institute for Chemistry, Multiphase Chemistry Department, Mainz, Germany (u.poschl@mpic.de, +49-(0)6131-305-6019), (2) 2Atmospheric Physics Department, Institute of Physics, St. Petersburg State University, St. Petersburg, Russia

In this study we derive and apply a mass-based hygroscopicity parameter interaction model for efficient description of concentration-dependent water uptake by atmospheric aerosol particles with complex chemical composition. The model approach builds on the single hygroscopicity parameter model of Petters and Kreidenweis (2007). We introduce an observable mass-based hygroscopicity parameter κ_{m} which can be deconvoluted into a dilute hygroscopicity parameter (κ_{m}^{0}) and additional self- and cross-interaction parameters describing non-ideal solution behavior and concentration dependencies of single- and multi-component systems.

For reference aerosol samples of sodium chloride and ammonium sulfate, the κ_{m}-interaction model (KIM) captures the experimentally observed concentration and humidity dependence of the hygroscopicity parameter and is in good agreement with an accurate reference model based on the Pitzer ion-interaction approach (Aerosol Inorganic Model, AIM). Experimental results for pure organic particles (malonic acid, levoglucosan) and for mixed organic-inorganic particles (malonic acid – ammonium sulfate) are also well reproduced by KIM, taking into accounting apparent or equilibrium solubilities for stepwise or gradual deliquescence and efflorescence transitions.

The mixed organic-inorganic particles as well as atmospheric aerosol samples exhibit three distinctly different regimes of hygroscopicity: (I) a quasi-eutonic deliquescence & efflorescence regime at low-humidity where substances are just partly dissolved and exist also in a non-dissolved phase, (II) a gradual deliquescence & efflorescence regime at intermediate humidity where different solutes undergo gradual dissolution or solidification in the aqueous phase; and (III) a dilute regime at high humidity where the solutes are fully dissolved approaching their dilute hygroscopicity.

For atmospheric aerosol samples collected from boreal rural air and from pristine tropical rainforest air (secondary organic aerosol) we present first mass-based measurements of water uptake over a wide range of relative humidity (1–99.4 %) obtained with a new filter-based differential hygroscopicity analyzer (FDHA) technique. For these samples the concentration dependence of κ_{m} can be described by a simple KIM model equation based on observable mass growth factors and a total of only six fit parameters summarizing the combined effects of the dilute hygroscopicity parameters, self- and cross-interaction parameters, and solubilities of all involved chemical components. One of the fit parameters represents κ_{m}^{0} and can be used to predict critical dry diameters for the activation of cloud condensation nuclei (CCN) as a function of water vapor supersaturation according to Köhler theory. For sodium chloride and ammonium sulfate reference particles as well as for pristine rainforest aerosols consisting mostly of secondary organic matter, we obtained good agreement between the KIM predictions and measurement data of CCN activation.

The application of KIM and mass-based measurement techniques shall help to bridge gaps in the current understanding of water uptake by atmospheric aerosols: (1) the gap between hygroscopicity parameters determined by hygroscopic growth measurements under sub-saturated conditions and by CCN activation measurements at water vapor supersaturation, and (2) the gap between the results of simplified single parameter models widely used in atmospheric or climate science and the results of complex multi-parameter ion- and molecule-interaction models frequently used in physical chemistry and solution thermodynamics (e.g., AIM, E-AIM, ADDEM, UNIFAC, AIOMFAC).

Reference: Mikhailov, E., Merkulov, V., Vlasenko, S., Rose, D., and Pöschl, U.: Mass-based hygroscopicity parameter interaction model and measurement of atmospheric aerosol water uptake, Atmos. Chem. Phys. Discuss., 11, 30877-30918, doi:10.5194/acpd-11-30877-2011, 2011; revised version accepted for publication in Atmos. Chem. Phys., 2013.