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Aerosol hygroscopicity distribution in size-resolved CCN measurements: the concept, validation and applications

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This work proposed a concept of aerosol hygroscopicity distribution for the analysis and modeling of cloud condensation nucleus (CCN) activity. The cumulative distribution function of particle hygroscopicity is defined as the number fraction of particles with a given dry diameter and with an effective hygroscopicity parameter smaller than a hygroscopicity parameter. The hygroscopicity distribution concept clarified the meaning of CCN spectrum and reflected the heterogeneous properties of aerosols. A range of model scenarios were used to explain and illustrate the concept and exemplary practical applications were shown with CCN measurement data from polluted megacity. The hygroscopicity distribution for polluted megacity air in Beijing exhibits a lognormally distributed mode with hygroscopicity kappa values around 0.2-0.4, and a significant particle fraction (ca. 10-20%) with hygroscopicity kappa values <0.1, which could be attributed to externally mixed soot particles. Lognormal distribution functions were found to be suitable for approximately describing the hygroscopicity distribution of aerosols in polluted megacity air as well as in pristine rainforest air. The width of hygroscopicity distributions can be used as an aerosol mixing state indicator and was found to be in agreement with aerosol mixing state indicators derived by volatility tandem differential mobility analyzer (VTDMA).
Previous studies showed that multiple charges on aerosols and DMA transfer function could significantly affect the size-resolved CCN measurement results and hence need to be corrected. However, the commonly used correction approach is based on the assumption that particles are of the same hygroscopicity parameter. For atmospheric aerosols, this assumption and corresponding correction approaches might not be valid. So we developed a CCN simulator model to quantify the effects of multiple charges and DMA transfer function on the CCN spectra. Several data correction approaches were evaluated and a more rebust correction approach was recommended.