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Cloud condensation nuclei closure study on long-term observation data

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Aerosol-cloud interactions (ACI) are currently the least understood influence on climate change (IPCC, 2013). ACI are largely controlled by the relative change in cloud condensation nuclei (CCN) and ice nuclei (IN) number concentrations. As direct CCN and IN measurements are not always at hand, being able to predict their concentrations is important.

Focusing on CCN, we use monitoring type data from 5 stations within the ACTRIS network in Europe (http://www.actris.net/) and the ATTO site in Brazil to compare measured CCN concentrations at various supersaturations with predicted concentrations based on kappa-Köhler theory. The locations represent a variety of different environments including the rain and boreal forests, and continental-remote, marine and high-alpine conditions. At all sites, at least one full year of CCN concentrations, size distribution and chemical composition data were available for the period between 2012 and 2014. Submicron particle chemical composition data were provided by either Aerodyne aerosol mass spectrometers (AMS) or aerosol chemical speciation monitors (ACSM) and used to derive the hygroscopicity parameter kappa.

We explore how well standard kappa-Köhler theory can be applied in the different environments. We find kappa ranging between 0.2 (median) for forest environments, 0.35 for continental-remote and high-alpine conditions, and 0.75 for the marine site. Generally, theory can predict actual CCN concentration within $\pm$ 25 % with relatively high correlation coefficients > 0.8 for all supersaturations and throughout all seasons. Applying a fixed kappa of 0.3 instead of hourly derived values yields similarly good results in most cases, while it leads to a discrepancy mismatch for the marine site and a slight difference for the rain forest aerosol.

In addition, we find a number of mismatches that can be explained by data quality issues rather than deficiencies in the theory. A sensitivity study shows that only unrealistic assumptions for the surface tension, a parameter to derive kappa, and large errors in the kappa value itself, e.g., caused by uncertainties in the chemical composition, can lead to predictions outside $\pm$ 30 %. A more likely source of uncertainty are errors in particle counting and sizing. This shows that there are certain limitations and data have to be used with caution estimating whether the potential uncertainty in the data is lower than the uncertainty of the expected results, e.g., model-measurement intercomparisons.