



Cloud condensation nuclei in polluted air and biomass burning smoke: Size-resolved measurements and implications for the modeling of aerosol particle hygroscopicity and CCN activity

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Atmospheric aerosol particles serving as cloud condensation nuclei (CCN) are key elements of the hydrological cycle and climate, but their abundance, properties and sources are highly variable and not well known. We have measured and characterized CCN in polluted air and biomass burning smoke during the PRIDE-PRD2006 campaign on 1–30 July 2006 at a rural site ~60 km northwest of the mega-city Guangzhou in southeastern China.

CCN efficiency spectra (activated fraction vs. dry particle diameter; 20–300 nm) were recorded at water vapor supersaturations (S) in the range of 0.07% to 1.27%. Depending on S , the dry CCN activation diameters were in the range of 30–200 nm, corresponding to effective hygroscopicity parameters κ in the range of 0.1–0.5. The hygroscopicity of particles in the accumulation size range was generally higher than that of particles in the nucleation and Aitken size range. The campaign average value of κ for all aerosol particles across the investigated size range was 0.3, which equals the average value of κ for other continental locations. During a strong local biomass burning event, the activation diameters increased by ~10% and the average value of κ dropped to 0.2, which can be considered as characteristic for freshly emitted smoke from the burning of agricultural waste. At low S ($\leq 0.27\%$), the maximum activated fraction remained generally well below one, which indicates substantial proportions of externally mixed CCN-inactive particles with much lower hygroscopicity – most likely soot particles (up to ~60% at ~250 nm).

The mean CCN number concentrations ($N_{\text{CCN},S}$) ranged from 1100 cm^{-3} at $S=0.07\%$ to 16 000 cm^{-3} at $S=1.27\%$, representing ~7% to ~85% of the total aerosol particle number concentration. Based on the measurement data, we have tested different model approaches (power laws and kappa-Köhler model) for the approximation/prediction of $N_{\text{CCN},S}$ as a function of water vapor supersaturation, aerosol particle number concentration, size distribution and hygroscopicity. Depending on S and on the model approach, the relative deviations between measured and predicted $N_{\text{CCN},S}$ ranged from a few percent to several hundred percent. The largest deviations occurred at low S and with power laws based on particle number concentration. With the kappa-Köhler model and a constant hygroscopicity parameter of 0.3, the deviations were on average less than ~20%, which confirms that $\kappa=0.3$ may be suitable for approximating the hygroscopicity and CCN activity of continental aerosols in large scale models of the atmosphere and climate. On the other hand, the temporal variations of $N_{\text{CCN},S}$ observed during the biomass burning event and in diurnal cycles could not be captured with constant κ (deviations up to ~80%). With variable κ values obtained from individual CCN efficiency spectra, the relative deviations were on average less than ~10% and hardly exceeded 20%, confirming the applicability of the kappa-Köhler model approach for efficient description of the CCN activity of atmospheric aerosols. Note, however, that different types of κ -parameters have to be distinguished for external mixtures of CCN-active and -inactive aerosol particles.

References:

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