



## Cloud condensation nuclei from fresh and aged air pollution in the megacity region of Beijing: size-resolved measurements and parameterization of aerosol chemical composition, 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. CCN properties were measured and characterized during the CAREBeijing-2006 campaign at a suburban site south of the megacity Beijing, China. Size-resolved CCN efficiency spectra recorded for a supersaturation range of  $S = 0.07\%$  to  $0.86\%$  yielded average activation diameters in the range of 190 nm to 45 nm. The corresponding effective hygroscopicity parameters ( $\kappa$ ) exhibited a strong size dependence ranging from  $\sim 0.25$  in the Aitken size range to  $\sim 0.45$  in the accumulation size range. The campaign average value ( $\kappa = 0.3 \pm 0.1$ ) was similar to the values observed and modeled for other populated continental regions.

The hygroscopicity parameters derived from the CCN measurements were consistent with chemical composition data recorded by an aerosol mass spectrometer (AMS) and thermo-optical measurements of elemental and organic carbon (EC/OC). The CCN hygroscopicity and its size dependence could be parameterized as a function of AMS-based organic and inorganic mass fractions using the simple mixing rule  $\kappa_p \approx 0.1*f_{org} + 0.7*f_{inorg}$ .

When the measured air masses originated in the north and passed rapidly over the center of Beijing (fresh city pollution), the average particle hygroscopicity was reduced ( $\kappa = 0.2 \pm 0.1$ ), which is consistent with enhanced mass fractions of organic compounds ( $\sim 50\%$ ) and EC ( $\sim 30\%$ ) in fine particulate matter (PM1). Moreover, large fractions of CCN-inactive particles with much lower hygroscopicity were observed at low supersaturation ( $S = 0.07\%$ ), which can be explained by freshly emitted, externally mixed soot particles. Particles in stagnant air from the industrialized megacity region south of Beijing (aged regional pollution) were on average larger and more hygroscopic, which is consistent with enhanced mass fractions ( $\sim 60\%$ ) of soluble inorganic ions (mostly sulfate, ammonium, and nitrate). Accordingly, the number concentration of CCN in aged air from the megacity region was higher ( $(2.5\text{--}9.9) \times 10^3 \text{ cm}^{-3}$  vs.  $(0.4\text{--}8.3) \times 10^3 \text{ cm}^{-3}$ ) although the total aerosol particle number concentration was lower than in fresh city outflow ( $1.2 \times 10^4 \text{ cm}^{-3}$  vs.  $2.3 \times 10^4 \text{ cm}^{-3}$ ). A comparison with related studies suggests that the fresh outflow from Chinese megacity centers may generally contain more but smaller and less hygroscopic aerosol particles and thus fewer CCN than the aged outflow from megacity regions.