Supplement

S1. CCN activation model

The Activation model describes the CCNC response of transferred polydisperse charge-equilibrated particles that passed through an ideal DMA. The underlying method is similar to Petters et al. (2007), but the calculation part of CCN activation fraction is modified for the BS2-CCN system. Here, we summarize procedure and relevant equations.

DMA classifies particles with a narrow range of electrical mobilities. The electrical mobility, Z_p, is defined in Eq. (S1) to determine the particle size associated with the range of mobilities.

$$Z_p = \frac{neC}{3\pi\mu D_p} \qquad \text{(S1)}$$

where e is the elementary charge, C is the Cunningham slip correction, μ is the dynamic viscosity of air and n is the number of elementary charges on the particle. The relationship between particle electrical mobility (Z_p^*) and DMA parameters are defined by Knutson and Whitby (1975). The relationship is given in Eq. (S2).

$$Z_p^* = \frac{Q_{sh}}{2\pi LV} ln\left[\frac{R_2}{R_1}\right] \qquad \text{(S2)}$$

The mobility bandwidth, Z_p, is:

$$\Delta Z_p = \frac{Q_a}{Q_{sh}} Z_p^* \qquad \text{(S3)}$$

where Q_{sh}, and Q_a are the volumetric sheath flow and aerosol flow, respectively. L is the length of the DMA column, V is the negative potential applied to the inner cylinder. R_1 and R_2 are the inner and outer radius of the annular space of DMA, respectively. The transfer function for a cylindrical DMA column, $\Lambda(Z)$, is a piecewise linear probability function of triangular shape which is $\Lambda(Z_p^*) = 1$, $\Lambda(Z_p^* - 0.5\Delta Z_p) = \Lambda(Z_p^* + 0.5\Delta Z_p)$, and $\Lambda(Z_p^* - \Delta Z_p) = \Lambda(Z_p^* + \Delta Z_p) = 0$.

The fraction of particles carrying n charges (+1, +2) at charge equilibrium, $f(D,n)$, is calculated based on Wiedensohler (1988).

$$log_{10}[f(D, n = +1, +2)] = \sum_{i=0}^{5} a_i(n)(log_{10}D)^i \qquad \text{(S4)}$$

where $a_i(n)$ are coefficients obtained from DMA (TSI 3081) manual. As the size range of the particles covered in this study is less than 300nm, the fraction of particles carrying three or more charges is excluded.

With an assumed particle size distribution, an idealized CCN instrument response is determined by the following procedures. N_{CN} at D_{Z*} can be calculated using Eq. (S5). Equation (8) is for transforming the particle size distribution, dN/dD into the mobility domain.

$$N_{CN}(D_{Z*}) = \sum_{n=1}^{2}\left[\int_{Z=Z*+\Delta Z}^{Z=Z*-\Delta Z} f(D_{Z,n}, n)\Lambda(Z)\frac{dN_n}{dZ} dZ\right] \qquad \text{(S5)}$$

$$\frac{dN_n}{dZ} = \frac{dD}{dZ}\frac{dN}{dD} \qquad \text{(S6)}$$

where, dN_n/dZ is the differential size distribution of +n-charged particles in the mobility domain. It is noted that electrical mobility Z^* is based on a particle with +1 charge $\left(D_{Z*,1}\right)$, which should be assumed to define $\Lambda(Z)$. Similar to the N_{CN}, the number of particles that activate as CCN is calculated as follows:

$$N_{CCN}(D_{Z*}) = \sum_{n=1}^{2}\left[\int_{Z = Z* + \Delta Z}^{Z = Z* - \Delta Z} h\left(D_{Z,n}\right) f\left(D_{Z,n}, n\right) \Lambda(Z) \frac{dN_n}{dZ} dZ\right] \quad \text{(S7)}$$

where $h\left(D_{Z,n}\right)$ is a function for a fraction of particles that activate as cloud droplets. For the BS2-CCN system, the activation fraction, $h\left(D_{Z,n}\right)$, is calculated by Eq. (S8) and (S9). The activation fraction of aerosol particles with the same $S_{aerosol}\left(D_{Z,n}\right)$ is calculated by integrating the activation fraction function $g(x)$ and flow velocity $v(r)$ over the cross-section of the aerosol flow (Su et al., 2016). This is calculated as follows:

$$h\left(D_{Z,n}\right) = \frac{2\pi \int_0^r v g\left(S_{aerosol}(D_{Z,n}) - S_{tube}(r)\right) r dr}{2\pi \int_0^r v r dr} \quad \text{(S8)}$$

$$g(x) = \begin{cases} 1 & if\ x \leq 0 \\ 0 & if\ x > 0 \end{cases} \quad \text{(S9)}$$

Where r is the radial distance to the centerline of the activation unit (i.e., r = 0 for the center). $S_{tube}(r)$ is a typical distribution of supersaturation in the activation tube of CCNC and can be calculated as $S_{tube}(r) = S_{max} \times cos(0.14 \times r)$. The S_{tube} profile is adopted from http://nenes.eas.gatech. edu/Experiments/CFSTGC. html. It is noted that the unit of S is %, and the unit of r is millimeters. Although $S_{tube}(r)$ in the activation tube is also dependent on an axial (Robert and Nenes, 2005), $S_{tube}(r)$ in this study simply represent the maximum S in the axial direction at a given r. Flow velocity is prescribed as $v(r) = v_m \times (1 - r^2/R^2)$, where v_m is the maximum velocity at the centerline of the activation unit. It is noted that specific $S_{aerosol}\left(D_{Z,n}\right)$ corresponding to each D_{Z*} of a known particle can be calculated by Eq. (2) In this study, we used ammonium sulfate as calibration aerosols and set 0.63% for S_{max}.

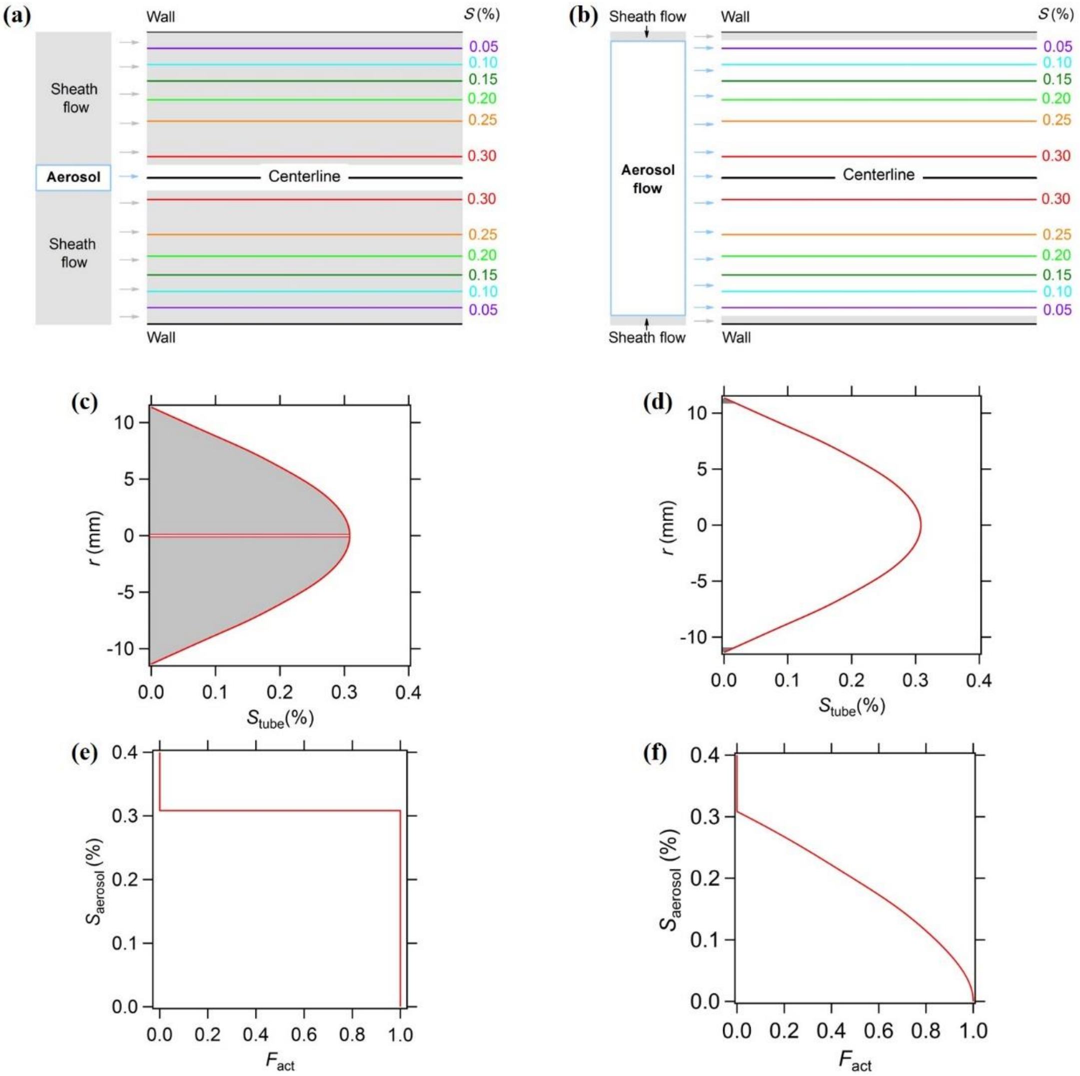

Figure S1: Schematics of typical CCN ((a), (c) and (e)) and BS2-CCN measurement ((b), (d) and (f)). (a) and (b) Contour of supersaturation in the CCN activation unit and configuration of aerosol and sheath flow; (c) and (d) Distribution of supersaturation in the activation unit (S_{tube}). r is the radial distance to the centerline. The shaded areas represent the sheath flow part, and non-shaded areas represent the aerosol flow part. (e) and (f) Plot of the activation supersaturation of aerosol particles S_{tube} against the activation fraction F_{act}. Reprinted from Su et al., (2016) under the Creative Commons Attribution 4.0 License.

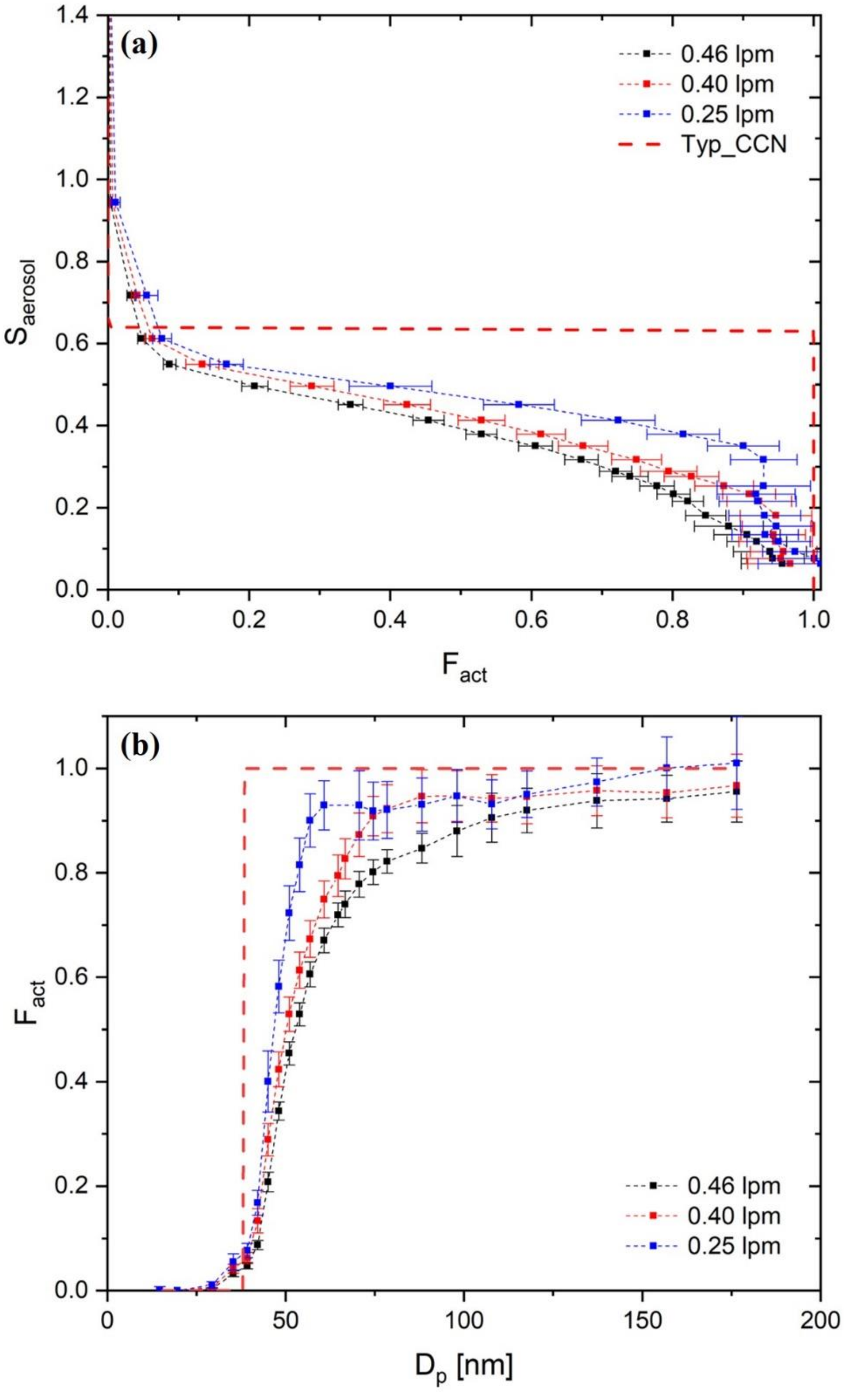

Figure S2: (a) Calibration curves ($F_{act} - S_{aerosol}$) and (b) activation curve ($F_{act} - D_p$) for three different sample flows (0.46 lpm (black), 0.40 lpm (red) and 0.25 lpm (blue)). Red dashed line indicates an ideal result of typical CCNC for comparison. Total flow (sample + sheath) is set to be 0.50 lpm. The experiment is conducted under dT=8 K setting with ammonium sulfate particles.

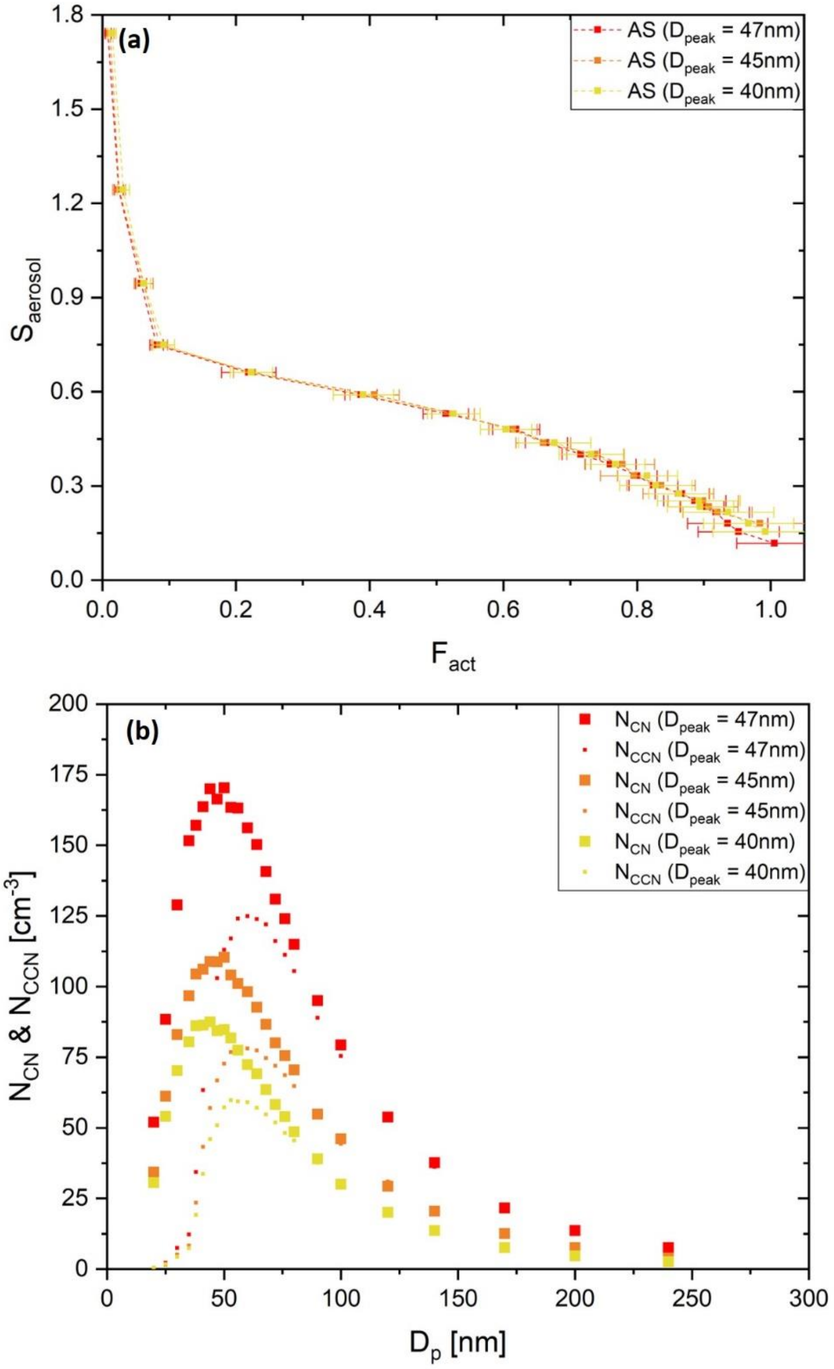

Figure S3: (a) Calibration curves and (b) number size distributions of N_{CN} and N_{CCN} of ammonium sulfate particle for dT=10 K.

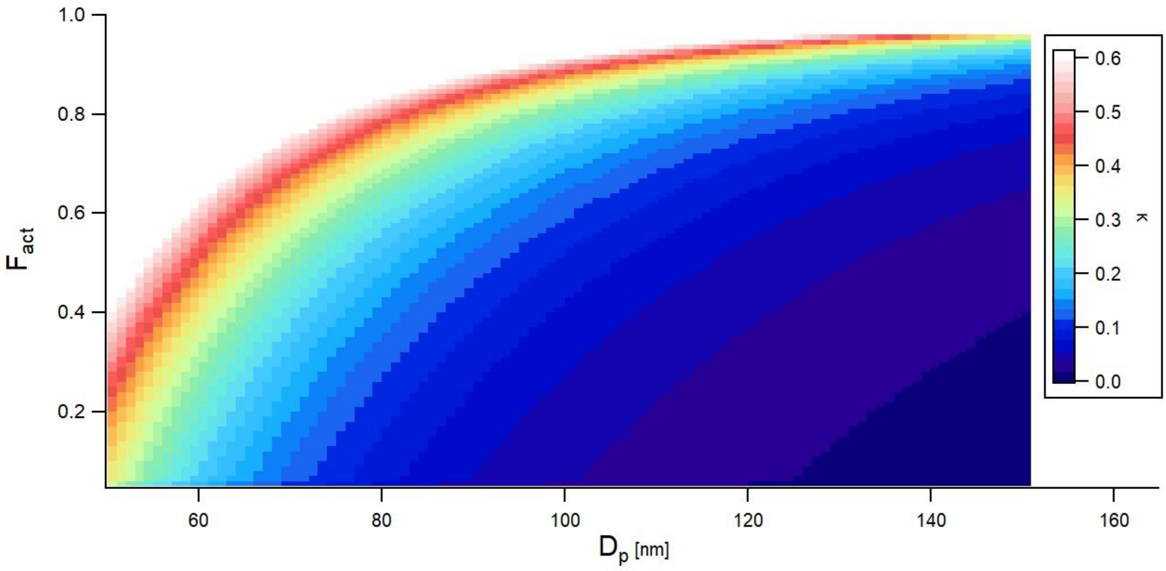

Figure S4: κ distribution which corresponds to the F_{act} values of a particle ranged from 50 nm to 150 nm. It is calculated from the fitting curve (Eq.3) of $F_{act} - S_{aerosol}$ relation for dT=8 K condition. Details of the fitting curve equation are described in Section 2.4, and the coefficients of the equation are presented in Table 1.

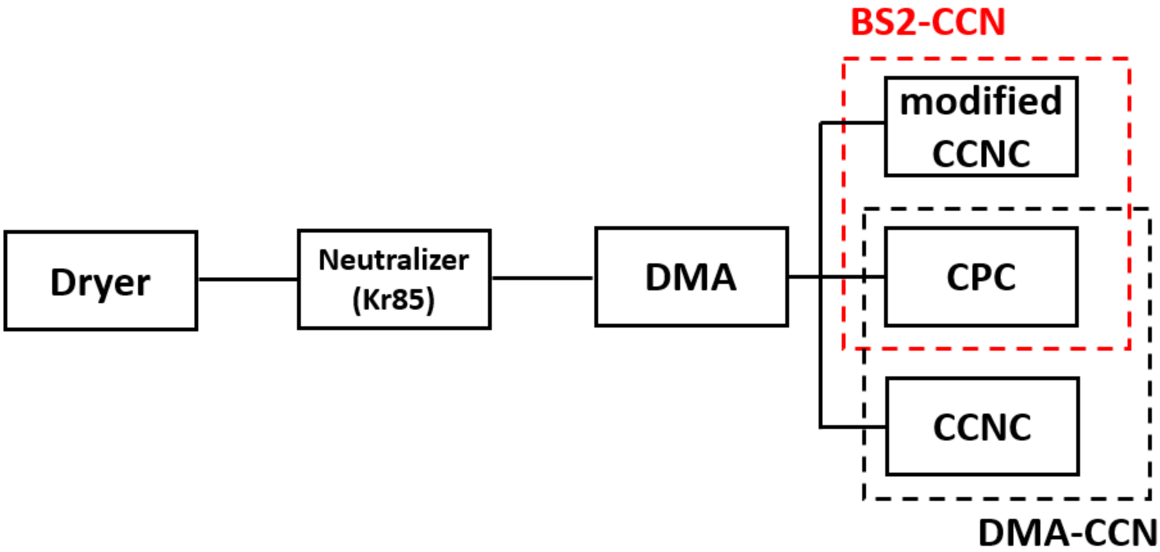

Figure S5: Schematic plot of instrumental setup for inter-comparison between DMA-CCN and BS2-CCN measurement

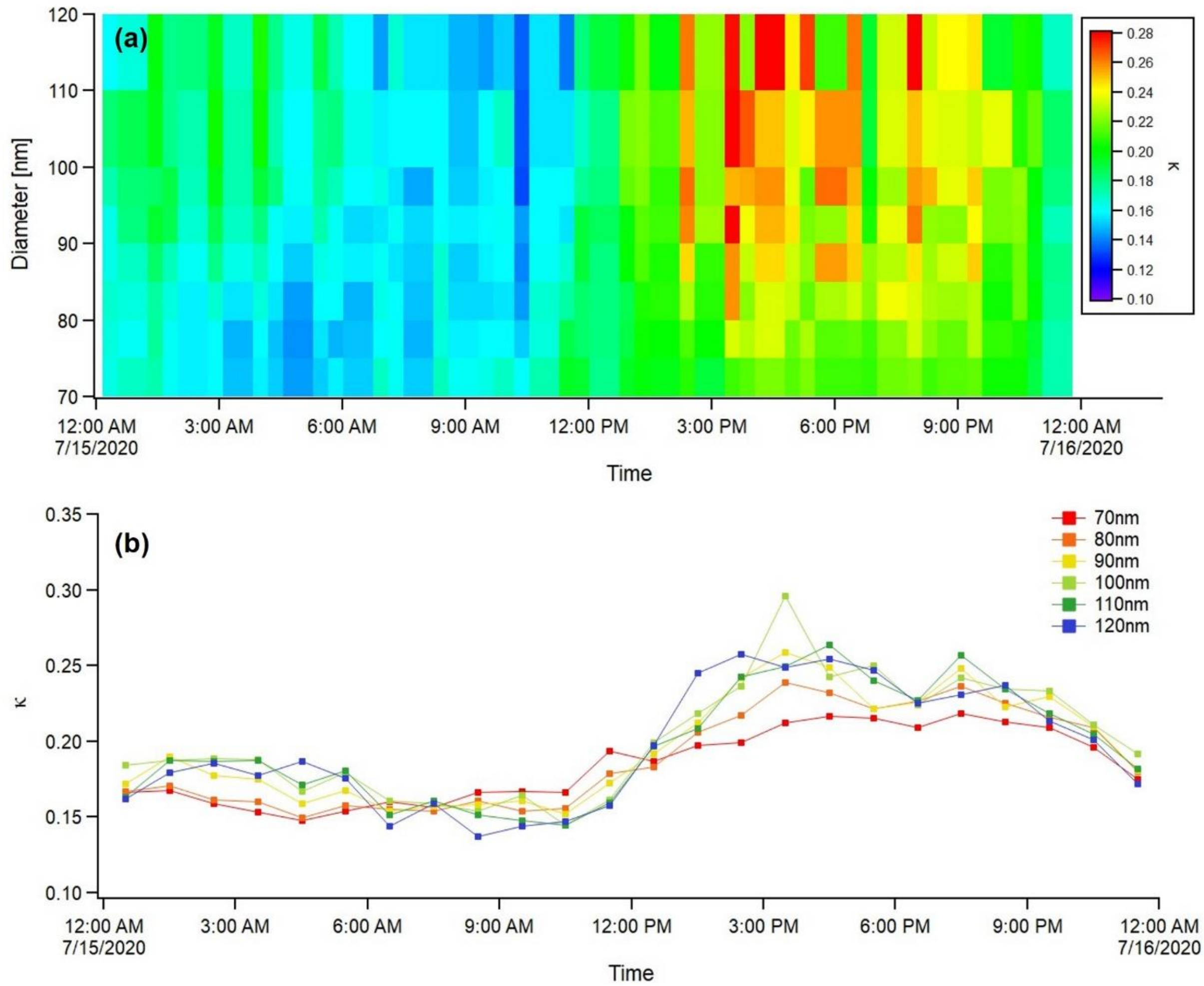

Figure S6: Diurnal variation of (a) κ distribution and (b) κ value of each D_p (between 70 nm and 120 nm) during the intercomparison experiment for ambient aerosols.

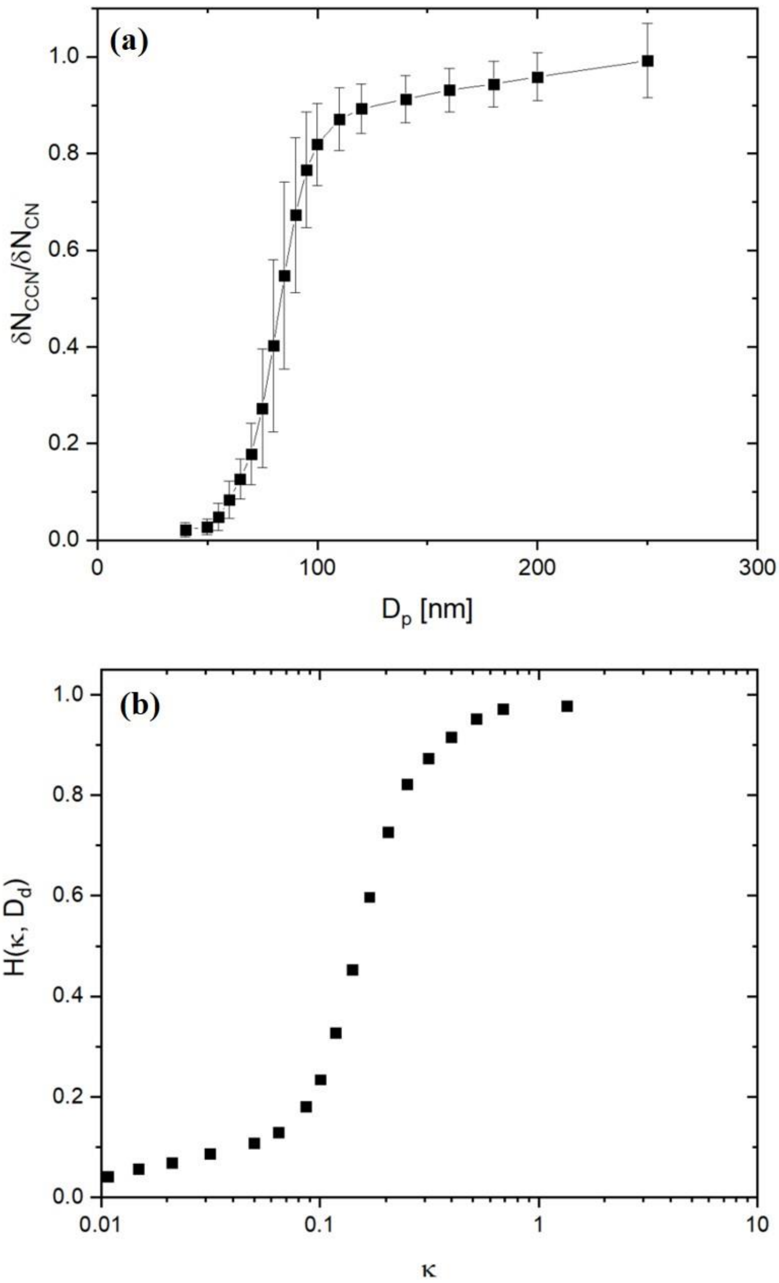

Figure S7: (a) Average CCN efficiency spectra and (b) average cumulative particle hygroscopicity distribution, $H(\kappa, D_d)$, of DMA-CCN measurement for 0.4% S. The $H(\kappa, D_d)$ value in panel (b) refers to different sizes. $H(\kappa, D_d)$ is defined as the number fraction of particles having a hygroscopicity parameter smaller than κ at a given dry diameter, D_d (Su et al., 2010).