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Size-resolved cloud condensation nuclei (CCN) activity and closure analysis at the HKUST Supersite in Hong Kong

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1 Uncertainty of N_{CN}, N_{CCN}, κ_{CCN} and κ_{AMS}

The relative uncertainty of N_{CN} ($\varepsilon_{\mathrm{CN}}$) is mainly determined by the uncertainty in the flow rate (10%) and number counting (10%) of TSI 3785 WCPC, and the overall $\varepsilon_{\mathrm{CN}}$ is 14%. The relative N_{CCN} ($\varepsilon_{\mathrm{CCN}}$) uncertainty depends on the concentration-dependent Poisson statistical uncertainty and the CCNc flow rate uncertainty (Moore et al., 2012; Roberts and Nenes, 2005):

$$\varepsilon_{\mathrm{CCN}}^2 = \varepsilon_{Q_{\mathrm{CCN}}}^2 + \frac{\tau_{\mathrm{CCN}}}{N_{\mathrm{CCN}} Q_{\mathrm{CCN}}} \qquad (1)$$

where τ_{CCN} is the integration time (1 second) of CCNc optical particle counter (OPC) and Q_{CCN} is the sample flow rate of CCNc (45 $\mathrm{cm^3\ min^{-1}}$), the $\varepsilon_{Q_{\mathrm{CCN}}}$ is about 5%. Overall, $\varepsilon_{\mathrm{CCN}}$ increases with the decease of *SS*, since the N_{CCN} decreases as *SS* decreases. The maximum uncertainty is ~38% at *SS* = 0.15% in this study.

The uncertainty of κ_{CCN} comes from the accuracy of the dry particles classified by the DMA and the uncertainty of the activation efficiency ($\varepsilon_{\mathrm{CCN/CN}}$) used for D_{50} determination. The $\varepsilon_{\mathrm{CCN/CN}}$ can be obtained from

$$\varepsilon_{\mathrm{CCN/CN}}^2 = \varepsilon_{\mathrm{CN}}^2 + \varepsilon_{\mathrm{CCN}}^2 \qquad (2)$$

The sizing accuracy of DMA was determined by the accuracy of DMA sheath flow rate and classifying voltage. The typical value is less than 3% (Wang et al. 2003). The overall uncertainly in derived κ_{CCN} ranges from 23% to 39%.

The uncertainty of κ_{AMS} comes from the uncertainty in κ_{CCN} (as κ_{org} and κ_{inorg} are derived from κ_{CCN} shown in main text), as well as the species densities and the volume fractions of organics and inorganics that are derived from the AMS mass concentrations. The uncertainty of inorganic compositions densities could be considered negligible. For organics, a mean value of 1.36 ± 0.11 for H:C and 0.40 ± 0.11 for O:C were found (Lee et al., 2013), and the organic density estimated from the ratio of O:C ranging from 0.29 to 0.46 and H:C ranging from 1.49 to 1.28 were from 1.15 $\mathrm{g\ cm^{-3}}$ to 1.35 $\mathrm{g\ cm^{-3}}$ (Kuwata et al., 2011). The uncertainty of an assumed organic density of 1.3 $\mathrm{g\ cm^{-3}}$ is less than 8%, which is smaller than the uncertainty in mass concentrations (~30%) measured by AMS because of the uncertainty in collection efficiency

(*CE*) (Middlebrook et al., 2012). The large fractions of semi-volatile oxygenated organics aerosols (SV-OOA) (23.5%) and low-volatile oxygenated organic aerosols (LV-OOA) (53.9%) suggested that particles were largely aged and likely internally mixed. An internal mixing state implies that the influences of *CE* on both NR inorganic and organic species are in the same degree, thus have little impact on the derived volume fractions. In all, an uncertainty of 16% is estimated for determination of inorganics and organics volume fractions, which are mainly due to the uncertainties in relative ionization efficiency (*RIE*) (Bahreini et al., 2009; Mei et al., 2013). The signal-to-noise ratio of the AMS data concerned, i.e. the ratio of mass concentrations for the measurement period to that for the filter period, was higher than 6 for this particle size range. It is worth noting that the low signal-to-noise ratios of AMS measurements for small particles (D_m < 50 nm) will cause high uncertainty in κ_{AMS} derivation.

2 Supporting Figures

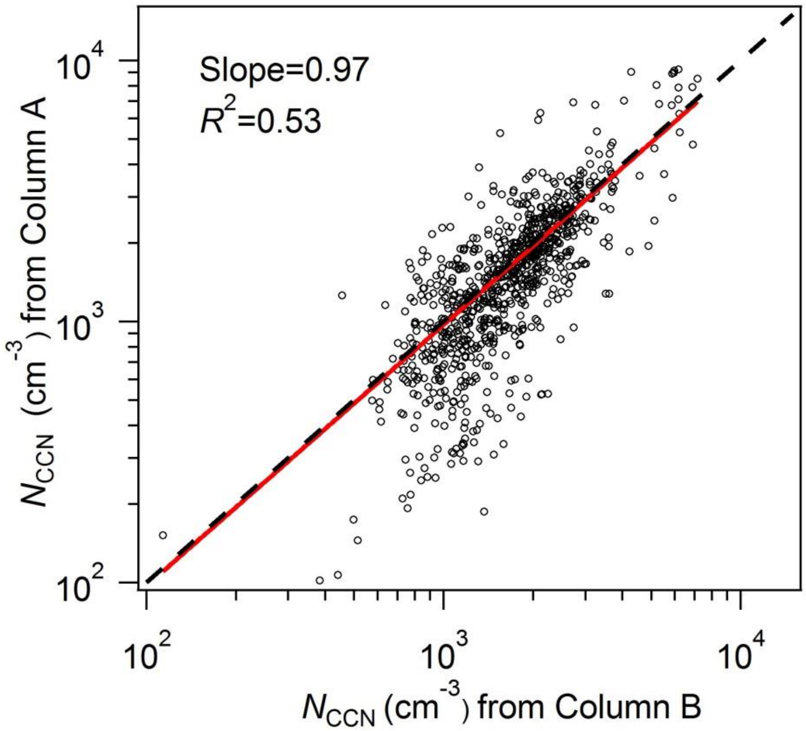

Fig.S1 Correlation of N_{CCN} from size-resolved CCN measurement (Column A) and bulk measurement (Column B).

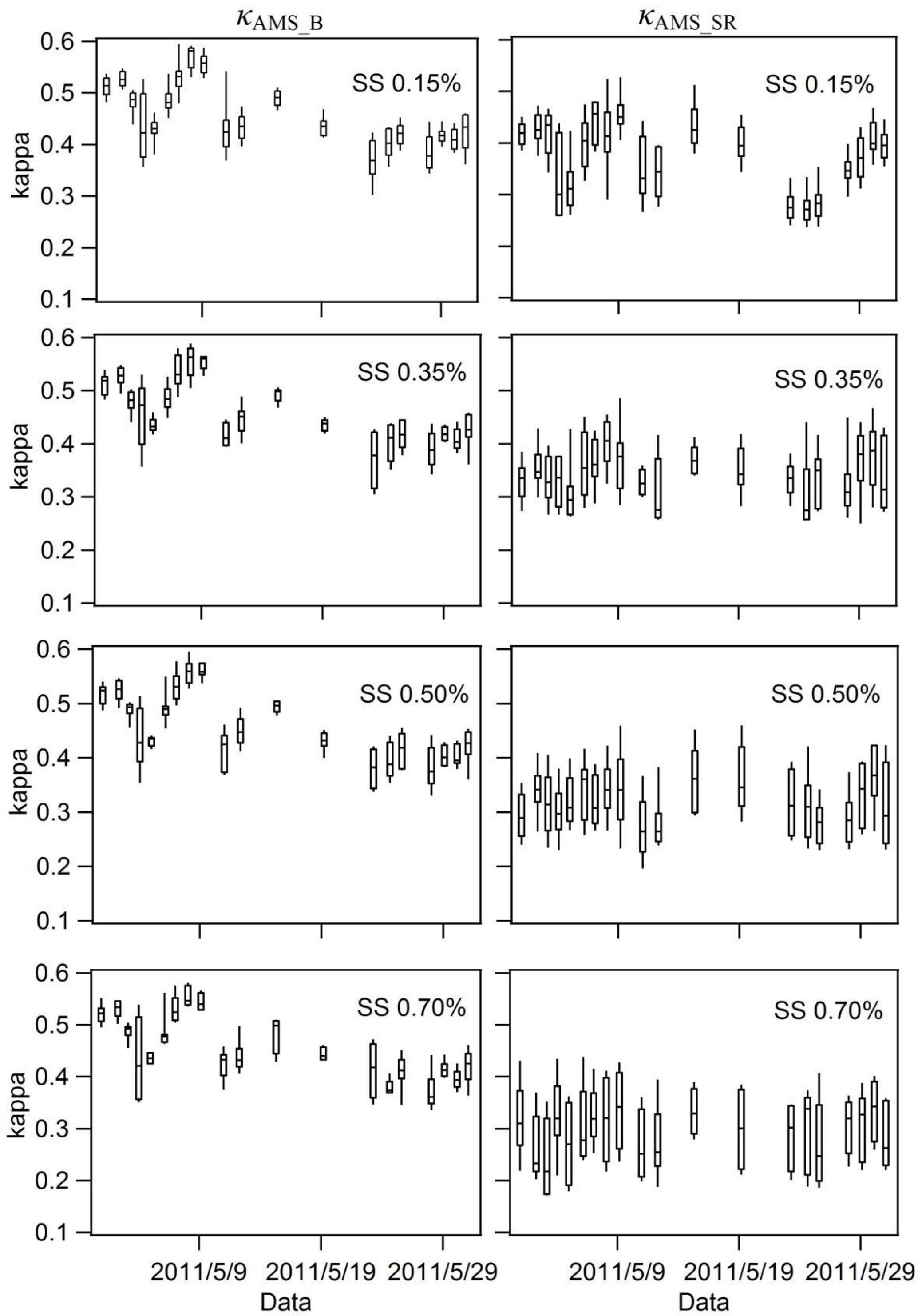

Fig.S2 The bulk κ_{AMS} (κ_{AMS_B}) and the size-resolved κ_{AMS} (κ_{AMS_SR}) derived from AMS measurement.

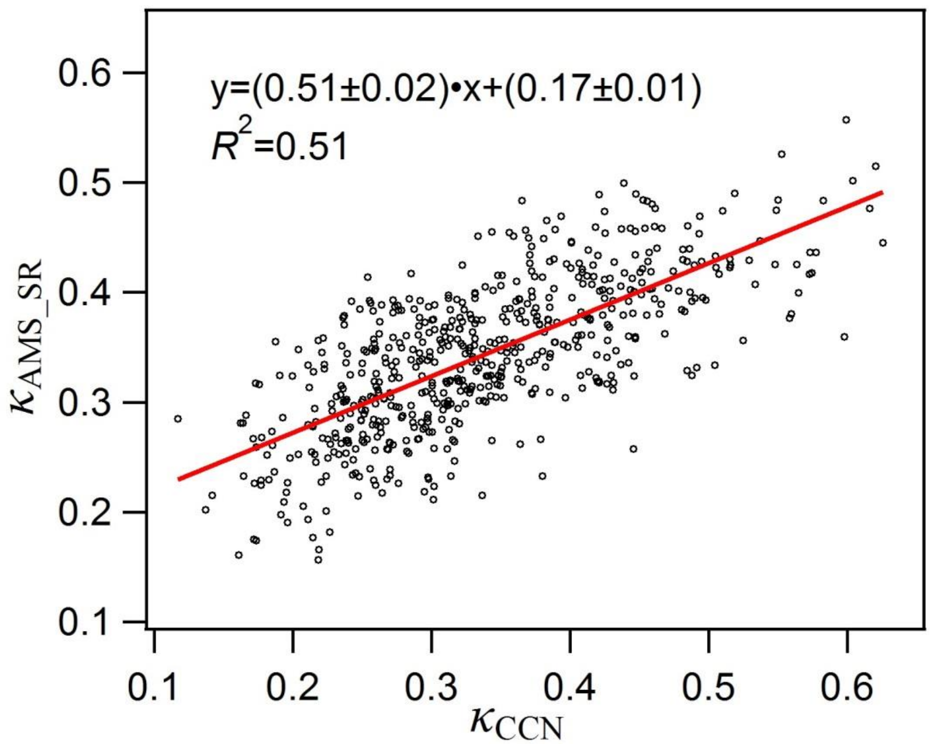

Fig.S3 Correlation of κ_{CCN} derived from CCN measurement and κ_{AMS_SR} from AMS measurement.

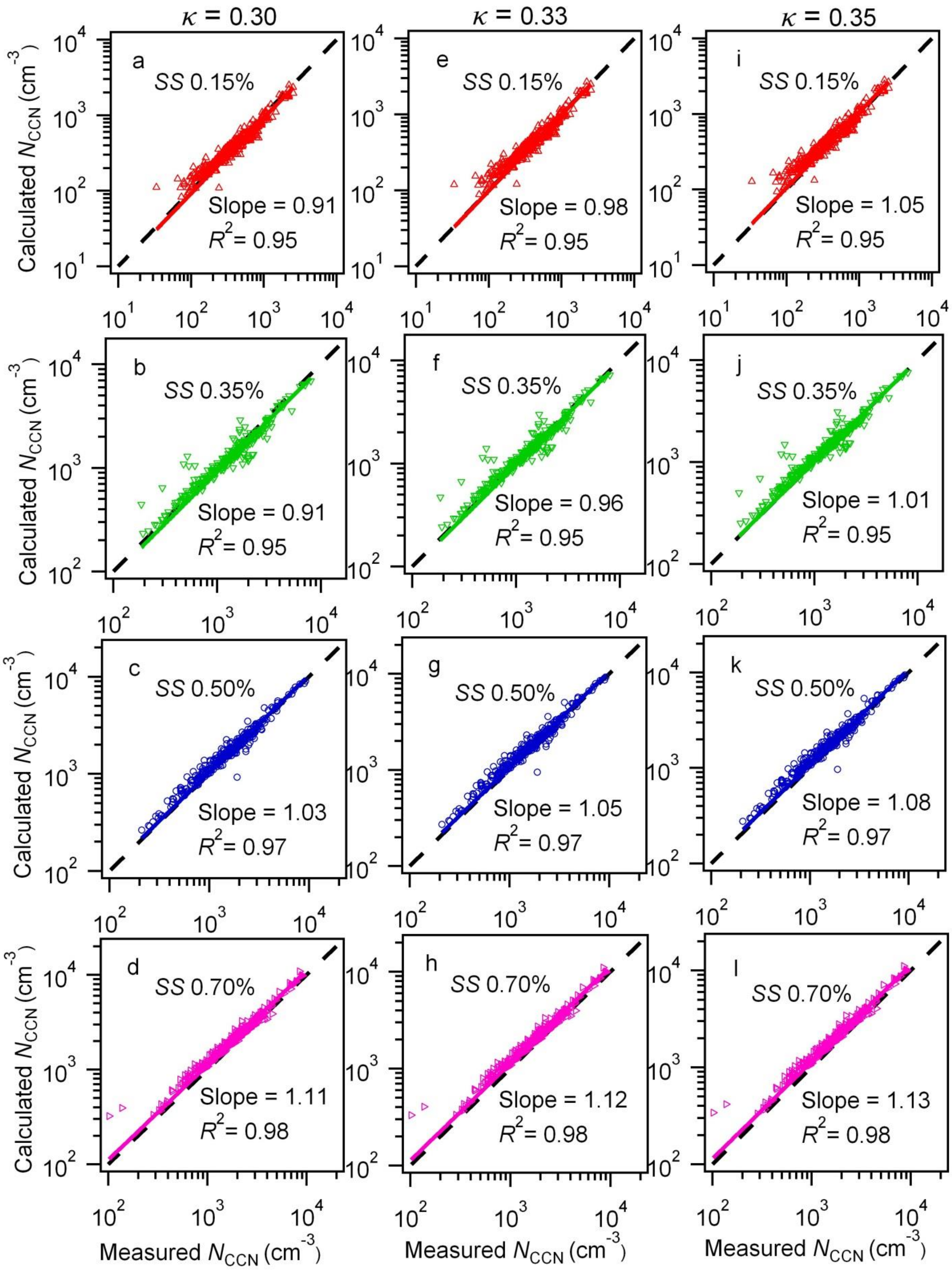

Fig.S4. Predictions of N_{CCN} based on D_{50} derived from constant κ of (a-d) 0.30, (e-h) 0.33 and (i-l) 0.35 during whole period, respectively.

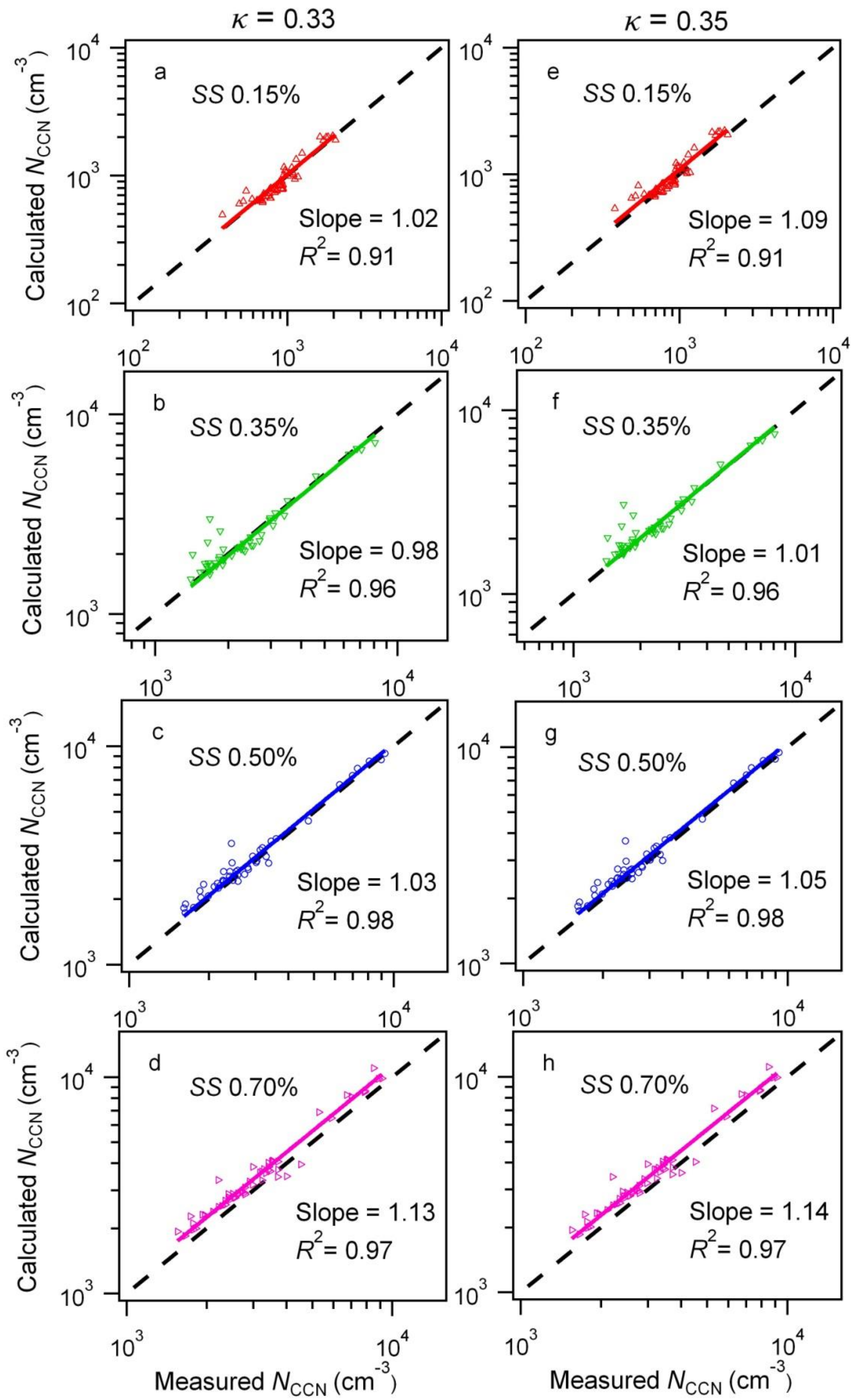

Fig.S5. Predictions of N_{CCN} based on D_{50} derived from constant κ of (a-d) 0.33 and (e-h) 0.35 during hazy period.

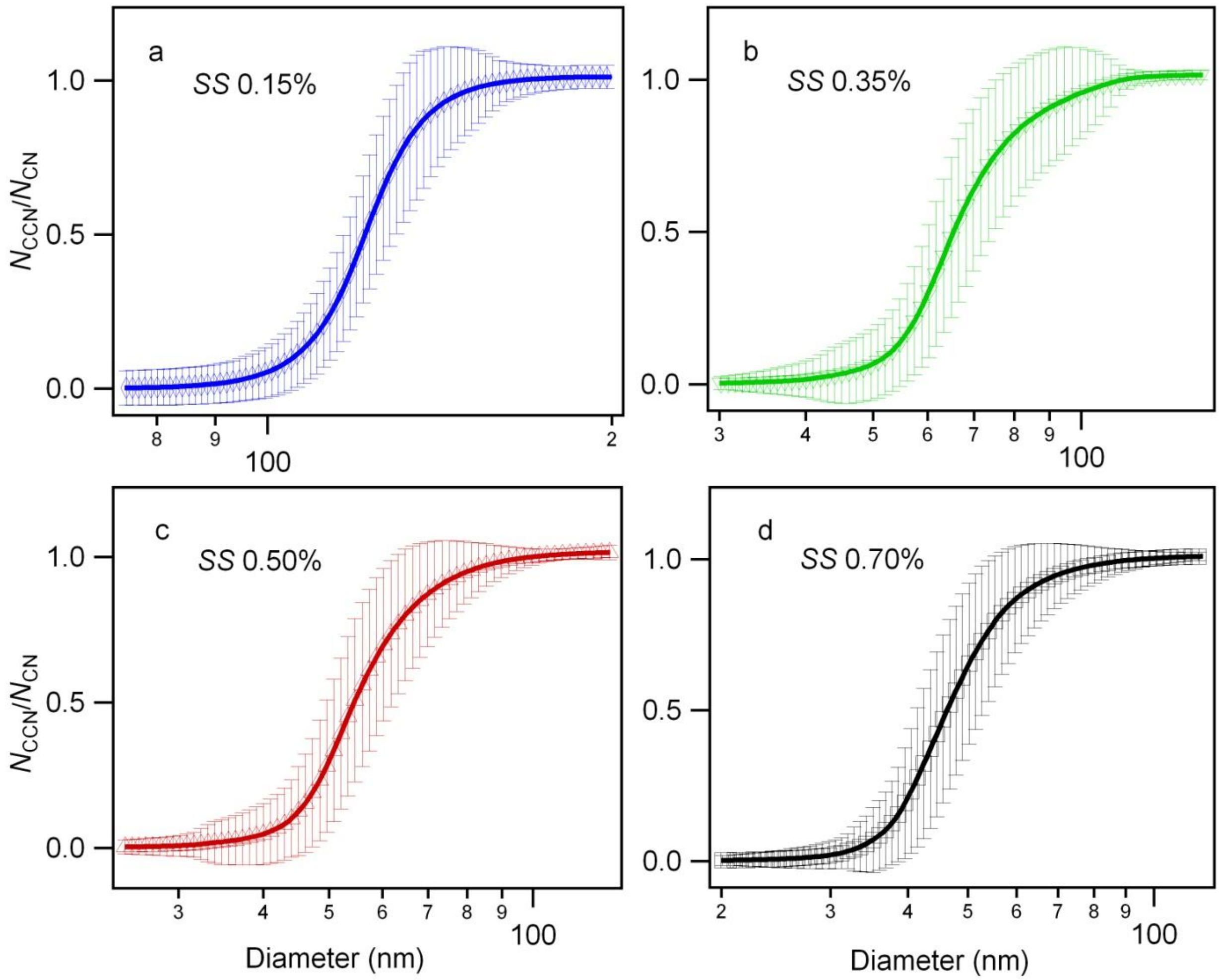

Fig.S6. The average size-resolved CCN activation ratio at *SS* (a) 0.15%, (b) 0.35%, (c) 0.50% and (d) 0.70%.

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