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## Rapid measurement of particle hygroscopicity and CCN activity using broad scanning supersaturation (BS2)-CCNC: calibration and intercomparison

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For understanding and assessing aerosol-cloud interactions and their impact on climate, reliable measurement data of aerosol particle hygroscopicity and cloud condensation nuclei (CCN) activity are required. Furthermore, aerosol liquid water, mainly controlled by hygroscopicity, affects heterogeneous and multiphase reactions of aerosol particles. The CCN activity of aerosol particles can be determined by scanning particle size and supersaturation ( $S$ ) in the CCN measurement. Compared to the existing CCN activity measurement, a broad supersaturation scanning CCN (BS2-CCN) system, in which particles are exposed to a range of  $S$  simultaneously, can measure particle hygroscopicity and CCN activity with a high-time resolution. Based on a monotonic relation between the activation supersaturation of aerosol particles ( $S_{\text{aerosol}}$ ) and the activation fraction ( $F_{\text{act}}$ ) of the BS2-CCN measurement, we can derive  $\kappa$ , a single hygroscopicity parameter, directly.

Here, we describe how the BS2-CCN system can be effectively calibrated and which factors can affect the calibration curve ( $F_{\text{act}} - S_{\text{aerosol}}$ ). For calibration, size-resolved CCN measurements with ammonium sulfate (AS) and sodium chloride particles are performed under the three different thermal gradient ( $dT$ ) conditions ( $dT=6, 8, \text{ and } 10$ ). First, the shape of the calibration curve is primarily influenced by  $S_{\text{max}}$ , maximum  $S$  in the activation tube. We need to determine appropriate  $S_{\text{max}}$  depending on particle size and  $\kappa$  to be investigated. To minimize the effect of double/multiple charged particles, small  $D_g$  and  $\sigma_g$  in number size distribution are recommended when generating the calibration aerosols. Sheath-to-aerosol-flow ratio (SAR) is the third factor to be considered. BS2-CCNC system uses a low SAR with a wider inlet compared to the typical CCN measurement, which can make a monotonic relation between  $F_{\text{act}}$  and  $S_{\text{aerosol}}$ . Lastly,  $F_{\text{act}}$  is affected by particle number concentration and has a decreasing rate of  $0.02/100 \text{ cm}^{-3}$  (within  $N_{\text{CN}} \sim 300 \text{ cm}^{-3}$  for AS) due to the water consumption in the chamber. For evaluating the BS2-CCN system, intercomparison experiments between typical DMA-CCN and BS2-CCN measurement were performed with the laboratory-generated aerosol mixture and ambient aerosols. Statistically good agreements of  $\kappa$  values between DMA-CCN and BS2-CCN measurements for both intercomparison experiments imply that the BS2-CCN system can measure particle hygroscopicity and

CCN activity well compared to the existing measurement. We expect that this new system can be applied to aircraft and ship measurements that require a high-time resolution as well as ground measurement for a broad range of hygroscopicity distribution. Because hygroscopicity is closely related to the fraction of organics/inorganics in aerosol particles, our method can also serve as a complementary approach for fast detection/estimation of aerosol chemical compositions.