Response to Reviewer #2

This manuscript presents a novel broad supersaturation scanning CCN (SB2-CCN) system, which can measure the CCN activity with a high time resolution. Overall, this manuscript clearly explained the set-up and calibration of the instrument, and applied it in the field measurement. This manuscript is well-written and easy to follow. The following comments must be addressed before consideration for publication.

A: We thank the reviewer for encouraging and helpful comments on our manuscript. We believe that the quality of our manuscript is improved as we reflect the reviewer's comments. Below each of the questions/comments is written first with the Italic font, and then our response is followed with the normal font. We marked newly added or revised sentences by highlighting them in the revised manuscript.

Comments

Q1: I recommend some of the figures can be moved from the main text to supplement, such as Fig. 7 and Fig. 8. Figures 3 and 4 can be combined into one figure. Figures 10 and 11 can be combined also. I prefer a compact and relatively short manuscript to introduce such a new instrument.

A: I combined Fig.3 and 4 (now 'Figure.4'), and Fig. 10 and 11 (now 'Figure.9') as you suggested. And, Figure 8 (now 'Figure S4') is moved to the supplement. Figure 8 is still in the main text to show the curve fitting procedure.

Q2: In Sect. 3.1.2, the double charged aerosols effect of the calibration of Fact – Saerosol relation was discussed. Did you consider the double charged effect in ambient measurement?

A: For ambient measurement in this study, we do not consider the double charged effect for ambient measurement. We added the sentence "The double charged effect is not considered in the inter-comparison experiment." (Line 293-294). We will discuss the correction of multiply charged particles in detail along with the field campaign results in the next paper. Instead, in section 3.1.2, we performed an additional calculation for ideal activation fraction with atmospheric relevant particle size distribution. We add the figure in Fig.S4 and add sentence. "Assuming the atmospheric relevant particle number size distribution with $N = 1000\ cm^{-3}, D_g = 80\ nm, and\ \sigma_g = 1.5$ from Rose et al. (2011), F_{act_double} is up to 0.05 (Fig. S4). It is noted that aerosols with κ of 0.3 is assumed to be internally mixed, and S_{max} is set to be 0.2%. Although the effect of doubly charged particle in Fig. S4 is not significant, the effect of doubly charged particle cannot be ignored if D_g or σ_g becomes large in specific environments or conditions." (Line 200-204).

Q3: In Sect. 3.1.3, the water depletion in the activation tube by high number concentration was discussed. I was wondering do you pre-humidify the particle for ambient aerosol measurement. What

is the total particle number concentration of the ambient measurement in this study? In Lines 217-218, it was mentioned that the aerosol concentration needs to be considered when it is high. Do you think is it worth testing the water depletion by high number concentration (such as >1000 cm-3) with lab-generated particles or in a polluted environment?

A: Pre-humidifier system is not included during the ambient aerosol measurement in this study. Particle number concentration at each selected diameter during the measurement ranged from 0 to 74 cm^{-3} and average number concentration was 18 cm^{-3}. In other words, pre-humidifier system is not necessary for this measurement. We also performed the test of water depletion by high number concentration (Maximum N_{CN} ~ 1100 cm^{-3}). Slope was similar to the experiment in the main manuscript. For example, F_{act} decreases 1.3 % (120 nm) per increase of 100 cm^{-3} in the number of particles (Figure R1). As BS2-CCN measurement system is based on the size-resolved measurement, we expect that particle number concentration of selected dry diameter will be within the range of concentration presented in the manuscript for the typical observations of ambient aerosols except for special high concentration cases (Figure R2).

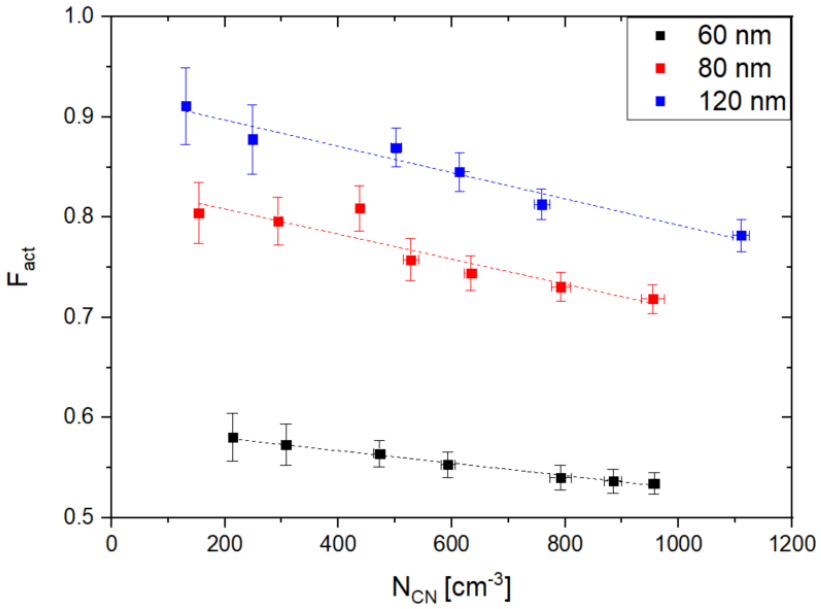

Figure R 1: Average and standard deviation (error bar) of F_{act} depending on the number concentration N_{CN} for 60 nm (black), 80 nm (red) and 120 nm (blue) of ammonium sulfate under the dT=7.7 K condition.

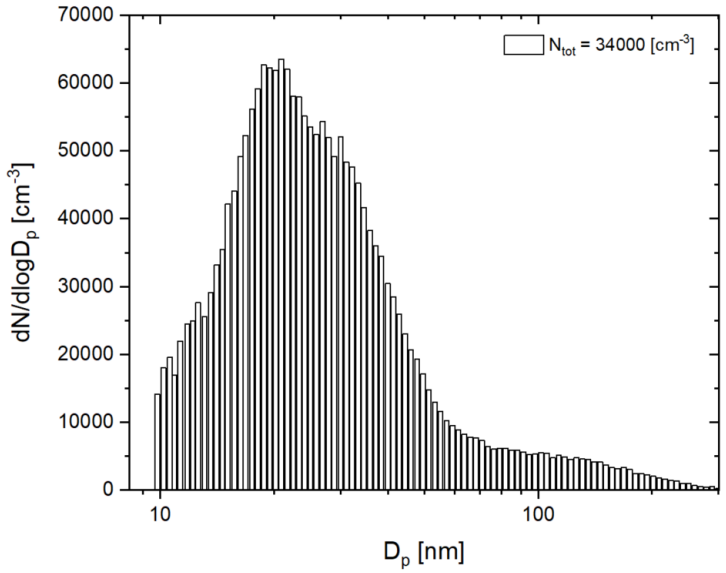

Figure R 2: Example of particle number size distribution data measured from SMPS in urban area during new particle formation event. Particle number concentrations at mode diameter are about 950 cm-3.

For calibration experiment with lab-generated conditions, water depletion without pre-humidifier system can affect the calibration curve if particle number concentration is too high. The slope between particle number concentration and F_{act} might be slightly different depending on the performance of CCNC itself. Therefore, we mention about recommended number concentration for compact instrumental setup without the pre-humidifier in the manuscript and the necessity of pre-humidifier system for special cases in the manuscript "In other words, a compact instrumental setup without the pre-humidifier system is sufficient for the BS2-CCNC calibration experiment as well as the measurement if aerosol particles are kept below ~ 3×10^2 cm^{-3}. Otherwise, we need a pre-humidifier system for high aerosol number concentration condition to avoid the decrease of F_{act}." (Line 225-227).

Q4: In Sect. 4.1, the AS and Su mixed particles are internally or externally mixed? If externally mixed, can it be seen in the Fact – Saerosol curves?

A: AS and Su mixed particle are internally mixed as we generated these aerosols by the solution of their mixtures. I follow the method that dissolving each component into the pure water to form dilute solution and generate atomized aerosol to generate internally mixed aerosols (Shi et al. 2012; Jing et al. 2016). Also, we get a smooth activation curve (no plateau in the middle of the curve, implying internally mixed) from DMA-CCN measurement. To be clear, I revise the sentence in the manuscript "We used an atomizer to generate internally-mixed nanoparticles with diameters of 30 – 160 nm by spraying a mixed solution of succinic acid and ammonium sulfate, which each pure component was completely dissolved in pure water obtained from a Mili-Q water purification system (Line 274-276).

$F_{act} - S_{aerosol}$ curve can be obtained from pure component with known aerosols as $S_{aerosol}$ is calculated based on κ-Köhler theory. For externally mixed aerosols, Su et al. (2016) suggest combining BS2-CCNC with complementary measurement (e.g. DMT-CCNC) to resolved the multiple κ mode by decoupling the mixed information and extracting the signal of each mode. If assuming aerosols with two log-normally distributed κ mode (κ of 0.3 and 0.01) with the same σ_κ (geometric standard deviation of κ), lower κ values are retrieved from model simulation with a BS2 approach itself ("apparent" in Fig.R3b) than those calculated with complementary measurement of the DMT-CCNC ("corrected" in

Fig.R3a). Below are equations, Eq. (R1) – (R3), used for calculation based on the assumptions, and the results are shown in the Fig.R3.

$$h(\kappa) = \frac{a_{\kappa 0.01}}{\sqrt{2\pi}log\sigma_\kappa} exp(-\frac{(log\kappa - log0.01)^2}{2(log\sigma_\kappa)^2}) + \frac{a_{\kappa 0.3}}{\sqrt{2\pi}log\sigma_\kappa} exp(-\frac{(log\kappa - log0.3)^2}{2(log\sigma_\kappa)^2}) \quad (R1)$$

$$a_{\kappa 0.01} = 0.3 \times log_{10}(D_d/30) \; and \; a_{\kappa 0.01} = 1 - a_{\kappa 0.3} \quad (R2)$$

$$F_{act} = a_{\kappa 0.3} \times F_{act,\kappa\,0.3} + a_{\kappa 0.01} \times F_{act,\kappa\,0.01} \quad (R3)$$

Where, $h(\kappa)$ is the fractional probability distribution function of hygroscopicity (Su et al. 2010), $a_{\kappa 0.01}$ and $a_{\kappa 0.3}$ present the number fraction of the two modes, and F_{act} represents an average of individual mode weighted by their number fraction

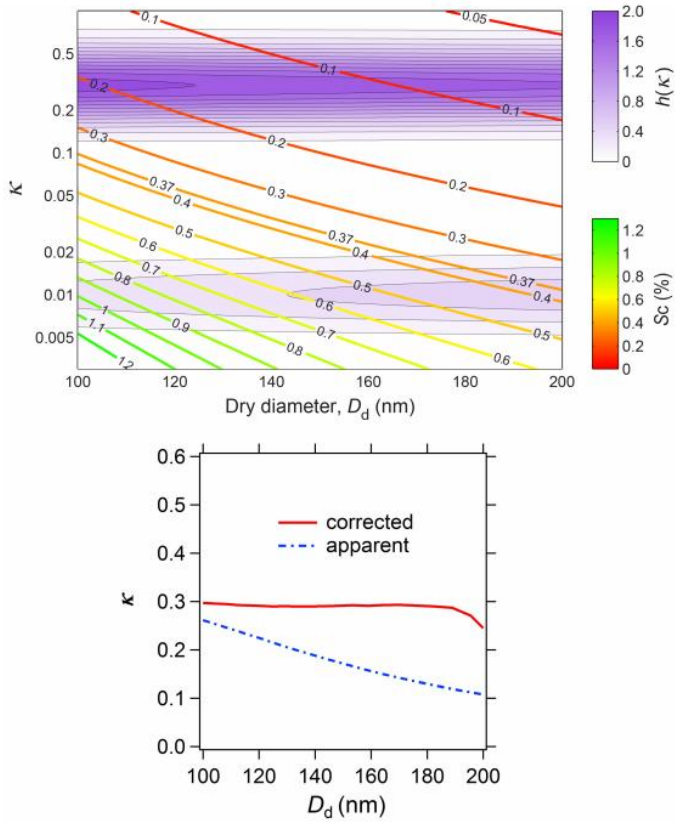

Figure R 3. (a) Supersaturation required to activate the less hygroscopic mode by the DMT-CCNC. The shaded isolines show the normalized size-resolved hygroscopicity distribution $h(\kappa)$, the probability density function of a two mode κ distribution (Rose et al. 2011). The colored isolines represent the supersaturation required to activate particles for a certain dry diameter D_d and κ. (b) Model simulation of κ retrieved for the more hygroscopic mode by BS-CCNC with (labeled as "corrected") and without (labeled as "apparent) complementary measurement of the DMT-CCNC. Reprinted from Su et al. (2016) under the Creative Commons Attribution 4.0 License.

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Q5: Line 50-60: When talking about the fast measurement of size-resolved κ, Zhang et al. (2021) introduced a novel method/technique to rapidly measure the size-resolved κ values under sub-saturation.

Zhang, J., Spielman, S., Wang, Y., Zheng, G., Gong, X., Hering, S., and Wang, J.: Rapid measurement of RH-dependent aerosol hygroscopic growth using a humidity-controlled fast integrated mobility

spectrometer (HFIMS), Atmos. Meas. Tech., 14, 5625–5635, https://doi.org/10.5194/amt-14-5625-2021, 2021.

A: We added the sentence "Zhang et al. (2021) introduce a novel measurement technique using a humidity-controlled fast integrated mobility spectrometer (HFIMS) to measure the size-resolved κ rapidly under the sub-saturated condition." (Line 60-62) as suggested.

Q6: Line 79: I realized that Fig. S1 was originally from Su et al. (2016). You probably need to mention that in the manuscript. I recommend moving Fig. S1 from supplement to main text because this figure is important for readers to understand the Fact.

A: Following the reviewer's suggestion, we have moved Fig. S1 to the main text. Besides in the figure caption, we also include another reference in the main text "Fig.1, Reprinted from Su et al., (2016) under the Creative Commons Attribution 4.0 License.".

Q7: Line 114: Why particle number concentration is the number concentration of condensation nuclei? NCCN and NCN are the same abbreviations?

A: N_{CCN} and N_{CN} are not the same abbreviation, number concentration of condensation nuclei for N_{CN} and number concentration of cloud condensation nuclei for N_{CCN}. Condensation nuclei are tiny particles in the air on which water vapor condenses and the term 'N_{CN}' is commonly used to describe the total particle number concentration (Rose et al. 2008; Pöhlker et al. 2018; Gao et al. 2020) and can be measured by condensation particle counter (CPC).

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