

Response to Interactive comment on “A broad supersaturation scanning (BS2) approach for rapid measurement of aerosol particle hygroscopicity and cloud condensation nuclei activity” by H. Su et al.

General comments

The authors present a new way of measuring the critical water vapour supersaturation required for activation of particles, termed broad supersaturation scanning (B2S) method. This new approach enables (according to the authors) fast measurements and easy data analysis, which can be implemented, with simple technical adjustments, to existing (and broadly used) instruments such as the DMT-CCNC. The method may be used for long-term measurements of atmospheric aerosols.

The novel approach for the B2S method is very interesting and the results from model simulations accompanied by measurements with a nano-CCNC give good support for the interpretations and conclusions made in the paper. As emphasized by the authors, there is a need for fast and well-controlled measurements of the critical supersaturation, both of ambient aerosols as well as for laboratory generated aerosols.

Response:

We thank Cerina Wittbom for the positive evaluation, very thorough and constructive comments/suggestions. We have revised the manuscript accordingly, with detailed responses as listed below.

Comments:

Why is the method termed broad supersaturation scanning (B2S)? As I understand, the measurement of the whole S distribution is instantaneous? What is the scanning part of the method? Could there be a more suitable name for the method?

Response:

Yes, the measurement of the whole S distribution is instantaneous. Under ideal conditions, we don't need to scan the supersaturation since a single scan will cover all supersaturation values between 0 and S_{\max} . In practice, supersaturation corresponding to $F_{\text{act}} < 0.2$ cannot be used (as explained in S.3.1 and Fig. 4), and multiple scans are needed. So we name each scan broad supersaturation scanning (BS2) to indicate its difference compared to the previous method.

Comments:

To simplify and make the concept of B2S more approachable for the reader, the authors exemplifies and makes comparisons with the commercially available CCNC from DMT, which I think is good in general. However, the DMT-CCNC is mentioned to a degree where one expects

to see results from experiments using the same instrument. Therefore, I suggest the authors to remind the reader that the B2S-method is tested only with the nano-CCNC (for example in the first paragraph in the Methodology section when the concept is introduced). I further suggest that the authors include a table of nomenclature and missing equations, the latter may be placed in the supplement.

Response:

Suggestions were taken. We have revised the text in the Methodology section to clarify the fact (inserted text in blue):

"For easy understanding, we introduce the BS2-CCNC in comparison with previous designs of DMT-CCNC (note that the experimental evaluation in Sect. 3.3 was performed with a nano-CCNC instead of the DMT-CCNC) "

We have included a table of notation. Can the referee specify which missing equations we should include?

Notation (frequently used symbols and acronyms)

Symbol	Unit	Quantity
<i>BS2</i>		Broad supersaturation scan
<i>CCN</i>		Cloud condensation nuclei
<i>CCNC</i>		Cloud condensation nuclei counter
<i>D_d</i>	m	Dry particle diameter
<i>F_{act}</i>		Number fraction of activated aerosols particles in the CCN counter
<i>HTDMA</i>		Hygroscopicity tandem differential mobility analyser
<i>M_w</i>	kg mol ⁻¹	Molar mass of water
<i>R</i>	J K ⁻¹ mol ⁻¹	Universal gas constant
<i>r</i>	M (unless specified)	Radial distance to the centerline of the activation unit
<i>S</i>	%	Water vapor supersaturation
<i>S_{aerosol}</i>	%	Critical activation supersaturation of aerosol particles
<i>S_{max}</i>	%	Maximum water vapor supersaturation at the centerline of a CCN counter
<i>S_{min}</i>	%	Maximum water vapor supersaturation at the edge of the aerosol flow in the

		activation unit
S_{tube}	%	Maximum water vapor supersaturation in the axial direction for a given r
T	K	Temperature
WO_x		Tungsten oxides
κ		Effective hygroscopicity parameter
σ_κ		Geometric standard deviation in a lognormal κ distribution
σ_{sol}	$J m^{-2}$	Surface tension of water
ρ_w	$kg m^{-3}$	Density of pure water

Comments:

In general, the paper is well written and concise, however in some sections a more descriptive text would be appreciated. I have a few comments that the authors should address prior to publication.

Specific comments

Page 9714, line 25: Its desirable to add references for the two methods for determining CCN activities.

Response:

We have added the following references in the revised manuscript (Dusek et al., 2006;Frank et al., 2006;Moore and Nenes, 2009;Snider et al., 2010;Kuwata et al., 2008;Gunthe et al., 2011;Rose et al., 2011;Petters et al., 2009;Wex et al., 2009).

Comments:

Page 9715, lines 3-16: The reason for developing and using the new approach is, as this paragraph declares, that a fast scanning of S is important for ambient measurements. However, the authors could also emphasize that there is a need for a method measuring rapid changing laboratory generated aerosols.

Response:

We have included such statement in the revised manuscript:

"Fast scan of S is important for measurements of ambient or laboratory generated CCN with high temporal variability. "

Comments:

Page 9715, line 17: Please clarify if the measurements are supposed to be performed for size-resolved measurements.

Responses:

We have specified that it is supported to be size-resolved:

" This study presents a new approach, termed broad supersaturation scanning (BS2) methods, which measures the activation of **size-resolved** CCN simultaneously over a continuous range of S ."

Comments:

Page 9715, line 19: Specify "infinitely small".

Responses:

Since we can fix S , the time for a single scan can be considered as zero. We have revised the text as " which measures the activation of **size-resolved** CCN simultaneously over a continuous range of S *by a single scan.*"

Comments:

Page 9715, line 22: The author states that "..., B2S-CCNC may be built with simple modifications of existing DMT-CCNC.". As I understand, no modifications are needed of the instrument. Instead, a change in settings of the aerosol-to-sheath flow ratio is implemented. Please, clarify if the instrument need a change in settings, or modifications of the hardware and/or software, either in this part of the manuscript or later (e.g. at page 9716, line 26).

Response:

For a moderate change of the aerosol-to-sheath flow ratio, the DMT-CCNC doesn't need to be modified while a very high aerosol-sheath flow ratio (e.g., up to 9:1) will introduce turbulence to the activation chamber and break the working principle of DMT-CCNC. We have revised the text accordingly as

" the measured aerosol particles are introduced with a wider inlet ~~of (or with~~ a high aerosol-to-sheath flow ratio) and distributed in a broad cross-section of the activation tube"

Comments:

page 9716, line 5: $Stube$ is stated as (and shown in the figure) a function of r . Due to the relevance of $Stube$ in the analysis as well as future use of the B2S-method it is desirable to include the function in the paper or in the supplementary material.

Response:

We have given the equation:

" $S=0.3069*\cos(0.1401*r)$, in which the unit of S is % and the unit of r is millimeter."

Comments:

page 9716, line 20: Please clarify what you mean by "The interval of $Stube$ scans determines

the uncertainty of S_{aerosol} .” Do you mean the number of S -steps in a spectrum?

Responses:

Yes, that's what we meant. We have clarified that by adopting the referee's suggestion.

"[The number of \$S\$ scans/steps in a spectrum](#) determines the uncertainty of S_{aerosol} ."

Comments:

page 9716, line 26: (1) The concept of the approach is to “make use of the whole S distribution inside the activation chamber”, changing the aerosol-to-sheath flow ratio. The sheath-to-aerosol flow rate is preferably set to 10 for the DMT-CCNC (or aerosol-to-sheath=1/10). During measurements this ratio can be adjusted, for low concentrations to get sufficient counts, and for high concentrations to minimize coincidence in the OPC. How will low vs. high concentrations affect the B2S-method performance? Are there wall-losses, buoyancy or other effects to account for when changing the aerosol-to-sheath ratio?

Responses:

Since the BS2 analysis requires size-resolved measurements, relatively small amounts of aerosol particles are sampled in each scan. Thus the coincidence of particles in the OPC under high aerosol concentrations is less problematic compared to that in bulk CCN measurements. On the other hand, a higher aerosol-to-sheath flow ratio will increase the counts and instrument sensitivity. Overall, we expect that the BS2 method will be less sensitive to the change of aerosol concentrations compared with the traditional method.

For a high aerosol-to-sheath ratio, the most critical effects may be the generation of turbulence in the mixing of aerosol and sheath flows. Since the generation of supersaturation in a DMT-CCNC requires a laminar flow, it won't work under turbulent conditions. But according to our FLUENT calculations, the stable laminar flow can be achieved under some modifications of the DMT-CCN instrument. The full development of the BS2-CCN instrument, calibration and implication are planned for a future study.

Comments:

Page 9717, lines 2-4: (2) Fact is here “calculated by integrating the activation fraction function $g(x)$ over the cross section of the aerosol flow...”. Particles in the centerline ($r=0$, subjected to a higher S) should have a different flow rate than particles closer to the wall (subjected to decreasing S) due to the parabolic velocity condition in the instrument. Has this been considered and how will this affect the results of Fact? Will it also affect the time resolution of the instrument, if central and peripheral parts of the flow, corresponding to different critical supersaturations, have different residence times in the instrument?

Responses:

This is a very good point. By considering the different flow speed $v(r)$, Equation (1) will be changed to

$$F_{\text{act}} = \frac{\int_0^r v g(S_{\text{aerosol}} - S_{\text{tube}}) r dr}{\int_0^r v r dr}, \text{ in which } g(x) = \begin{cases} 1 & \text{if } x \leq 0 \\ 0 & \text{if } x > 0 \end{cases} \quad (1)$$

Applying a flow velocity profile of $v(r) = v_m*(1 - r^2/R^2)$, the dependence of F_{act} on S_{aerosol} will change as shown in Fig. R1. Since the dependence is still monotonic and can be determined by the calibration, it will not influence the data analysis procedure of BS2-CCN measurements.

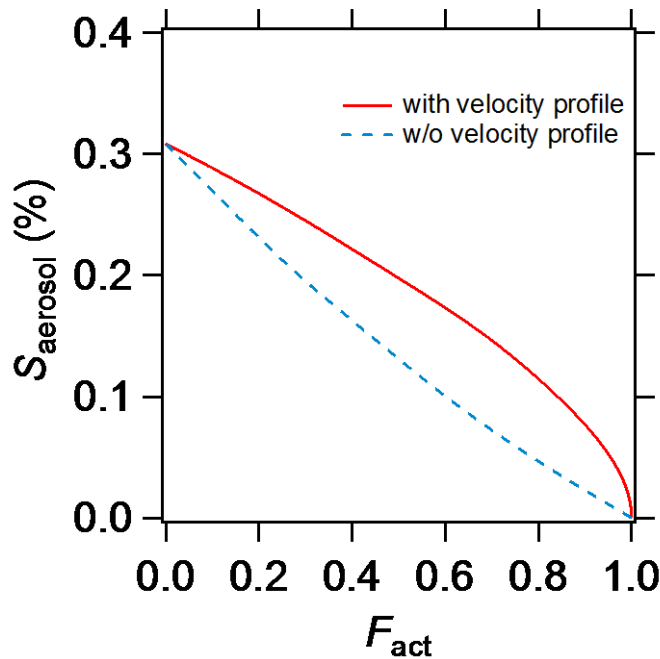


Figure R1: Plotting of the activation supersaturation of aerosol particles S_{aerosol} against the activation fraction F_{act} for a BS2-CCNC. One mistake was found in the calculation of the original $F_{\text{act}}-S_{\text{aerosol}}$ relation and was updated here (as labeled "w/o velocity profile").

Comments:

(3) How will F_{act} be affected by doubly/multiply charged particles? For the nano-CCNC the problem of doubly/multiply charged particles is minor. However, for measurements of ambient as well as laboratory-generated particles of larger particles this issue has to be taken into account. It would be desirable and give more credibility of the method to include a discussion and/or modelling results concerning this issue.

Response:

As the reviewer pointed out, the effect of the multiple charges is not negligible in BS2 measurement. However, the influence of the multiple charges and transfer function broadening can be corrected if multi-size (e.g., 20 diameters) measurements are performed. The size distribution of CN from CPC, and CCN from CCNC can be inverted separately with a standard SMPS/DMPS inversion algorithm. Then the size-resolved F_{act} can be calculated from those two inverted size distributions. Figure R2 shows an example of F_{act} derived from raw (blue) and inverted (red) size distributions, for lab-generated ammonium sulfate particles. We have included this information in the revised manuscript.

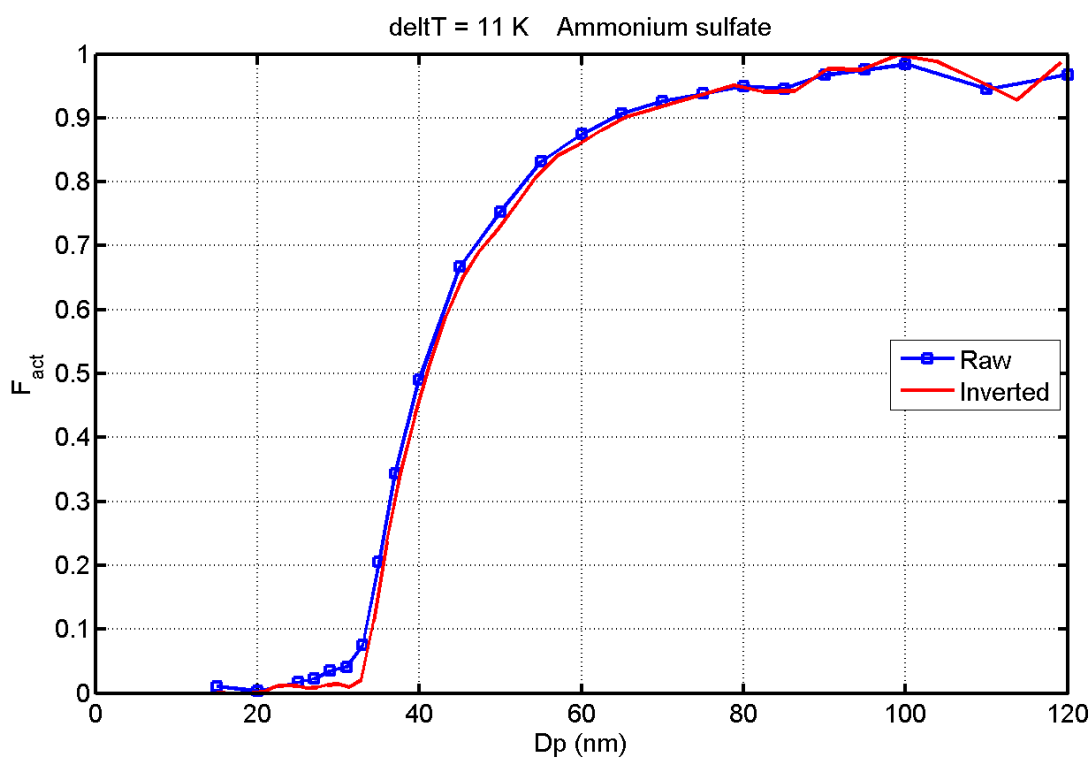


Figure R2. An exemplary F_{act} with (labeled as "inverted")/without (labeled as "raw") correction for doubly charged particles

Comments:

(4) Depending on the size chosen by a DMA, the broadening factor will be less or more. Usually, this is reflected in the resulting S-scan from the CCNC (by a more or less steep slope). How

will the broadening factor affect the results of the Fact – Saerosol relationship? Is there a certain size interval/range where the B2S method is applicable? Is there a limit in the sheath to aerosol flow ration, below which the assumption of the DMA broadening is insignificant? What would be the authors' recommendation?

Response:

This is a good point. DMA will also lead to a broadening. According to our experience, the broadening due to the DMA transfer function (aerosol/sheath flow ratio of 1:10) is smaller than that due to the hygroscopicity distribution of particles (Su et al., 2010). As shown in Fig. 4, the impact of broad hygroscopicity distribution has little effect on the retrieved κ at the $F_{act} > 20\%$.

According to Eq. (2), we have $d \ln D_d \approx -\frac{1}{3} d \ln \kappa$. Since $d \ln Z_p \approx -d \ln D_d$, we can estimate the

equivalent mobility bandwidth from the range of κ by $\frac{\Delta Z_p}{2Z_p} \approx \frac{1}{3}(3 \ln \sigma_k)$ in which $3 \ln \sigma_k$

represents almost the whole (99.7%) hygroscopic distribution and $\Delta Z_p/2$ is the half width. For a σ_k of 1.7 as in Fig. 4, the corresponding $\Delta Z_p/Z_p$ is 0.46. So we would recommend a threshold value of 0.46 for the aerosol to sheath flow ratio to ensure the quality of BS2-measurement.

$$\kappa \approx \frac{4A^3}{27(\ln(S_{aerosol} / 100\% + 1))^2 D_d^3} \quad (2)$$

Comments:

I lack a discussion and/or modelling results regarding sources of errors (including the issues listed above: (1) effect of changing the aerosol-to-sheath ratio, (2) different flow rates for particles in different parts of the laminar flow, (3) influence of doubly/multiply charged particles, and (4) effects of the broadening factor from the DMA). I suggest a section for discussing uncertainties and errors.

Response:

We have included a new section which summarizes our discussions on uncertainties and errors as presented above.

Comments:

Page 9717, line 15: Equation (2) is valid for $\kappa > 0.2$ derived from CCNC measurements (according to Petters and Kreidenweis, 2007), and if small numerical errors are acceptable it

can be used for lower numbers of κ . It would be useful for the reader if the authors could comment on this. For example, will this affect the results from the measurements (Figure 6 (c)).

Response:

In our nano-CCNC measurements (Fig. 6c), we did not use Equation (2) but a numerical iteration method to determine κ . Since the personal computer nowadays is fast enough for numerical analysis, we would rather not recommend using a simple approximation (Equation 2) for practical applications. We have included such statement in the revised manuscript. The difference between Equation (2) and the numerical method (more accurate) is shown in Fig. R3. It shows that the accuracy of the solution not only depends on κ but also on the particle size.

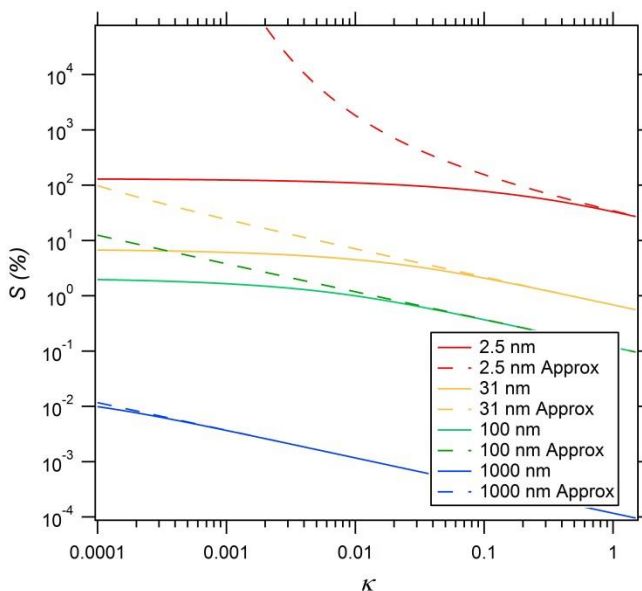


Figure R3: Comparison of supersaturation S determined by different methods. The dry particle size is labeled in the legend. The "Approx" represent the result determined by Eq. (2) while the other represent the result determined by numerical iteration.

Comments:

Page 9718, lines 8-9: According to the authors, the adjustment of S_{max} may be performed by changing the temperature gradient (ΔT) in the flow tube. Will the performance be affected by a change in ΔT ? I.e. is the performance the same for low and high ΔT ?

Response:

Comments:

Page 9718, lines 11-16: Here the authors state that size-resolved CCN measurements can be performed. Can polydispersed measurements also be performed and coupled with for example results from SMPS-measurements? Please comment.

Response:

We think it will probably not work for polydispersed measurements because there will be multiple solutions (n bins correspond to n κ , which can not be solved with only one equation). By assuming a uniform κ value for the whole size range, we can solve for κ but we are not sure how useful this parameter could be.

Comments:

Page 9722, lines 9-11: please clarify if the B2S-method is for size-resolved measurements or not.

Response:

We clarify that it is for size-resolved measurements.

Comments:

Page 9730, Figure 6 (b) and (c): Including error bars of the calibration curve and measurement results would add important information and credibility to the figures. Is the measurement point of tungsten oxides (WO) independent from the calibration?

Response:

We have included the error bars in Fig. 6 as suggested. The relative variation is ~ 2% to 5%. In Fig. 6, the point of tungsten oxides is part of the calibration data.

Technical corrections

*Page 9714, line 11: change "is" to "may ... be" (or similar) in the sentence: "Model simulations show that the B2S approach **may** also **be** applicable for measuring CCN activation of..."*

Response:

Corrected.

page 9714, line 2: change "budge" to "budget"

Response:

Corrected.

page 9715, line 20: change “the” to “a”, i.e. “...a fast determination of κ ,...”

Response:

Corrected.

page 9716, line 18: add “an infinite number of” inside the brackets, i.e. “(because it corresponds to an infinite number of values of Saerosol)”

Response:

Corrected.

Page 9717, line16: change “ σ is the surface tension of water” to “ σ' is approximated by the surface tension of water”

Response:

We see the point of the referee. In the paper of Petters and Kreidenweis (2007), σ was defined as the surface tension of the droplet (solution). However, it is difficult to know the real σ for practical CCN measurements. Since the value of κ depends on the choice of σ , it is important to treat the surface tension in the same way to make different CCN/ κ measurements comparable. Thus we defined an "effective" κ which is determined by the κ -Köhler approach by using the surface tension of water instead of that of the droplet/solution. Here “effective” means that the parameter accounts not only for the reduction of water activity by the solute but also for surface tension effects (Su et al. 2010; Rose et al., 2008; Gunthe et al., 2009). In the revised manuscript, we clarify that κ determined by Eq. (2) is an "effective" κ and we direct the readers to the reference Su et al. (2010).

Page 9718, line 19: change “For aerosol particles of the same composition (...), its Saerosol and κ values...” to “For aerosol particles of the same composition (...), **their** Saerosol and κ values...”

Response:

Corrected.

Page 9719, lines 3-5: The statement indicates more than one background site, please add more references.

Response:

Su et al. (2010) is a paper that introduces the concept of kappa distributions and use literature data at different sites to demonstrate the concept and the data retrieval methods. Since the

literature we cited does not give any kappa distribution, we cite only Su et al (2010) for such statement.

page 9719, lines 25-27: Please include some descriptive text concerning the probability density function of the κ distribution (κ -PDF, in Figure 5 (a)).

Response:

... have two lognormally distributed modes with the same σ_κ of 1.5, and a mode κ of 0.3 and 0.01 (Rose et al., 2011), respectively. The distribution of each mode can be described by the probability density function κ -PDF as

$$\kappa\text{-PDF} = \frac{a}{\sqrt{2\pi} \log \sigma_1} \exp\left(-\frac{(\log \kappa - \log \kappa_{g1})^2}{2(\log \sigma_1)^2}\right) + \frac{1-a}{\sqrt{2\pi} \log \sigma_2} \exp\left(-\frac{(\log \kappa - \log \kappa_{g2})^2}{2(\log \sigma_2)^2}\right) \quad \text{Eq (3)}$$

page 9720, lines 26-27: Is this adjustment of S_{min} the same as changing the aerosol-to-sheath ratio? Please clarify.

Response:

Changing the width of aerosol flow results in a change of the aerosol-to sheath flow ration. However, simply changing the aerosol to sheath ratio may disturb the flow conditions of CCN counter and breaks its working principle. From this aspect, it is not the same as adjusting the inlet design. We further clarify that in the revised manuscript "The adjustment of S_{min} can be done by changing the width of the aerosol flow with new design of inlets."

page 9721, line 13: add the figure character, "Figure 6 (a) ...", page 9721, line 20: add the figure character, "Figure 6 (b) and (c)..."

Response:

Corrected

Page 9722, line 6: change "can" to "may", i.e. "It may also be applicable..."

Response:

Corrected.

Page 9722, lines 13-14: change "...are especially well suited..." to "...may be well suited..." or similar. Due to the fact that no long-term measurements have been performed.

Response:

Corrected.

page 9729, Figure 5 (a) caption: include " κ -PDF" in the sentence "The shaded isolines describe the probability density function of a two-mode κ distribution." And add the reference

(Rose et al., 2011) of the figure.

Response:

Corrected.

page 9729, Figure 5 (b) caption: emphasize that these are modelled results and not retrieved from measurements.

Response:

Corrected.

Supplement, Figure S1 caption: Are the circles pure water droplets? Please clarify. This figure is exactly the same (including figure caption) as Figure 2 in Wang et al. (2015).

Response:

The circles are for $\kappa = 0$. We have clarified that in the revised manuscript. The figure is a re-use of Wang et al. (2015), we have included "reprint with permission" in the figure caption.

Reference:

Dusek, U., Frank, G. P., Hildebrandt, L., Curtius, J., Schneider, J., Walter, S., Chand, D., Drewnick, F., Hings, S., Jung, D., Borrmann, S., and Andreae, M. O.: Size Matters More Than Chemistry for Cloud-Nucleating Ability of Aerosol Particles, *Science*, 312, 1375-1378, 10.1126/science.1125261, 2006.

Frank, G. P., Dusek, U., and Andreae, M. O.: Technical note: A method for measuring size-resolved CCN in the atmosphere, *Atmos. Chem. Phys. Discuss.*, 6, 4879-4895, 2006.

Gunthe, S. S., Rose, D., Su, H., Garland, R. M., Achtert, P., Nowak, A., Wiedensohler, A., Kuwata, M., Takegawa, N., Kondo, Y., Hu, M., Shao, M., Zhu, T., Andreae, M. O., and Poeschl, U.: Cloud condensation nuclei (CCN) from fresh and aged air pollution in the megacity region of Beijing, *Atmospheric Chemistry and Physics*, 11, 11023-11039, 10.5194/acp-11-11023-2011, 2011.

Kuwata, M., Kondo, Y., Miyazaki, Y., Komazaki, Y., Kim, J. H., Yum, S. S., Tanimoto, H., and Matsueda, H.: Cloud condensation nuclei activity at Jeju Island, Korea in spring 2005, *Atmos. Chem. Phys.*, 8, 2933-2948, 2008.

Moore, R. H., and Nenes, A.: Scanning Flow CCN Analysis: Method for Fast Measurements of CCN Spectra, *Aerosol Science and Technology*, 43, 1192 - 1207, 2009.

Petters, M. D., Carrico, C. M., Kreidenweis, S. M., Prenni, A. J., DeMott, P. J., Collett, J. L., Jr., and Moosmüller, H.: Cloud condensation nucleation activity of biomass burning aerosol, *J. Geophys. Res.*, 114, 10.1029/2009jd012353, 2009.

Rose, D., Gunthe, S. S., Su, H., Garland, R. M., Yang, H., Berghof, M., Cheng, Y. F., Wehner, B., Achtert, P., Nowak, A., Wiedensohler, A., Takegawa, N., Kondo, Y., Hu, M., Zhang, Y., Andreae, M. O., and Pöschl, U.: Cloud condensation nuclei in polluted air and biomass burning smoke near the mega-city

Guangzhou, China – Part 2: Size-resolved aerosol chemical composition, diurnal cycles, and externally mixed weakly CCN-active soot particles, *Atmos. Chem. Phys.*, 11, 2817-2836, 10.5194/acp-11-2817-2011, 2011.

Snider, J. R., Wex, H., Rose, D., Kristensson, A., Stratmann, F., Hennig, T., Henning, S., Kiselev, A., Bilde, M., Burkhardt, M., Dusek, U., Frank, G. P., Kiendler-Scharr, A., Mentel, T. F., Petters, M. D., and Pöschl, U.: Intercomparison of cloud condensation nuclei and hygroscopic fraction measurements: Coated soot particles investigated during the LACIS Experiment in November (LExNo), *Journal of Geophysical Research: Atmospheres*, 115, D11205, 10.1029/2009jd012618, 2010.

Su, H., Rose, D., Cheng, Y. F., Gunthe, S. S., Massling, A., Stock, M., Wiedensohler, A., Andreae, M. O., and Pöschl, U.: Hygroscopicity distribution concept for measurement data analysis and modeling of aerosol particle mixing state with regard to hygroscopic growth and CCN activation, *Atmospheric Chemistry and Physics*, 10, 7489-7503, 10.5194/acp-10-7489-2010, 2010.

Wex, H., Petters, M. D., Carrico, C. M., Hallbauer, E., Massling, A., McMeeking, G. R., Poulain, L., Wu, Z., Kreidenweis, S. M., and Stratmann, F.: Towards closing the gap between hygroscopic growth and activation for secondary organic aerosol: Part 1 -Evidence from measurements, *Atmos. Chem. Phys.*, 9, 3987-3997, 2009.