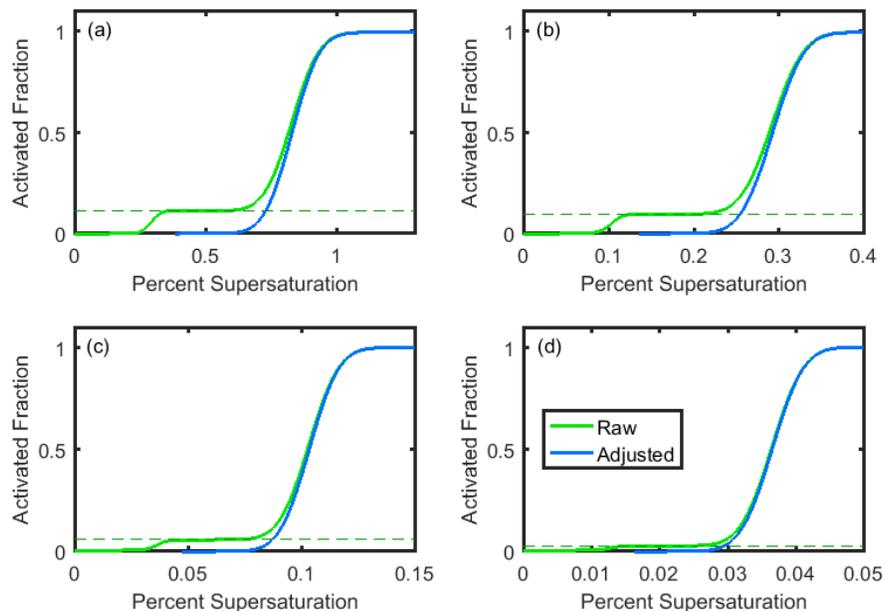


Authors' response to comments on "Instrument Artifacts Lead to Uncertainties in Parameterizations of Cloud Condensation Nucleation" Referee #2

Authors' response: We thank the Reviewer for her/his feedback, which we feel have improved the manuscript. New sections have been added to the manuscript in order to address the recommended revisions. While it is beyond the scope of this manuscript to address very possible source of uncertainty in detail, we have added significantly to the discussion of errors. In particular, to address the Reviewer's concern that we had not included uncertainties which arise due to multiple charging in the DMA, we have now performed calculations and added a new figure and addition text to the manuscript. Multiple charging uncertainties are incorporated into our overall analysis and conclusions. Additional specific modifications are discussed below.

- 1) Reviewer comment: Even though more discussion about artifacts and good practice in CCN measurements are needed, this is not the first paper dealing with the issue and with **the role of DMA flow ratio in determining the precision in fitting step functions** for evaluating number of CCN as a function of supersaturation. I would recommend that this literature is summarized and that results in this paper are discussed in perspective of this literature.

Authors' response: We thank the reviewer for this suggestion. The previous literature regarding the role of flow ratio in precision of fitting the step functions include the works of Petters et al. (2007b), Rose et al. (2008) and Zhao-Ze and Liang (2014) which are now discussed in the text. To summarize, the DMA sample/sheath flow ratio increases, the DMA transfer function broadens, allowing particles that area both larger and smaller than the selected diameter to exit the DMA. This in turn broadens the CCN activated fraction curve (Rose et al., 2008). DMA transfer function broadening may be significant enough to also increase the concentration of multiply-charged particles downstream of the DMA, resulting in the appearance of "plateaus" in the activated fraction curves. An example of a plateau is shown in the figure below, where the green lines correspond with raw activated fractions and the dashed lines on each plot correspond with the activated fraction plateau for 25, 50, 100, and 200 nm diameter particles. This figure is further discussed in a new section in the manuscript, 3.1.3 Effect of double and triple charges on particles. The effects of multiple charges are addressed in more detail in the response to Question 4. A new section has also been added to the manuscript to address particles with multiple charges, 3.1.3 Effect of double and triple charges on particles.



Authors' changes to the manuscript: Page 21 Lines 379-390 the text now reads: "The aerosol/sheath ratio within the DMA also modulates the effect of multiple charges on κ_{app} . As the aerosol/sheath ratio increases, the transfer function broadens, allowing particles that are both larger and smaller than the selected diameter to exit the DMA. This in turn broadens the CCN activated fraction curve (Rose et al., 2008). The larger particles will activate as CCN at lower supersaturations than particles with the selected diameter, resulting in an increase in the activated fraction plateau due to multiple-charged particles and a further decrease in the determined SS_{crit} . Petters et al. 2007b showed that CCN activated fraction curves are significantly skewed by multiply-charged particles when the mode diameter of the aerosol population upstream of the DMA exceeds the critical diameter of the size-selected particles. In an example CCN activated fraction curve, Rose et al. 2008 demonstrated that a 1:6 ratio of doubly-to-singly charged particles resulted in an underestimation of the critical activation diameter by 2%. Zhao-Ze and Liang, 2014 also showed that multiply-charged particles can introduce significant uncertainty in values of kappa (~17%) into hygroscopicity calculations."

- 2) Reviewer comment: Also, the use of hygroscopicity parameterizations goes much further back in time than indicated in this manuscript. The parameter epsilon is originally adopted from the work by Fitzgerald (1975) and in 1982 Fitzgerald et al. suggested a single hygroscopicity parameter (Bc). There might be even older literature.

Authors' response: This is a good point, thank you. The Reviewer is referring to hygroscopicity parameterizations which precede the "kappa" notation but are nonetheless about hygroscopicity, and yes, we agreed that these references should be included. A number of references to earlier works have been added to the manuscript's introduction. Specific changes are described in the revised text below.

Authors' Changes to the manuscript:

Pages 4-5 Lines 49-66 the text now reads: "In earlier works on the hygroscopic growth of aerosol particles, particle composition was found to influence the relationship between particle growth and relative humidity. Covert et al., 1972 posited that the effect of relative humidity on the light-scattering properties of an aerosol could be used to determine its chemical composition, in conjunction with other chemical analysis techniques; this concept was employed in Charlson et al., 1974 to quantify the sulfate content in aerosol particles. Winkler 1973 developed an equation for approximating the growth of an aerosol particle with relative humidity, based on the quantity and physical characteristics of the soluble species in the particle. Another approximation for the relationship between the equilibrium size of a particle and relative humidity was derived by Fitzgerald in 1975, in which the soluble fraction and composition of the soluble component(s) are taken into account.

Parameterizations which use hygroscopicity to predict CCN activation that pre-date Petters and Kreidenweiss 2007 exist as well. Fitzgerald et al., 1982 derived a particle composition parameter using the mass fraction and physical properties of soluble material in a particle. Kreidenweiss et al., 2005 determined that the critical activation diameter of dry aerosol particles can be calculated from simplified Köhler theory using the physical properties of water and the solute in a solution droplet. This parameterization has been used in CCN closure studies (Bougiatioti et al., 2009; Moore et al., 2011; Moore et al., 2012). The earliest prediction of CCN concentrations for specific particle diameters and hygroscopicity used this parameterization as well (Mochida et al., 2006)."

- 3) Reviewer comment: I also recommend that the authors state the limitations of the paper clearly:
- A) All CCN measurements are not done with the set up indicated in figure 1. Lab studies can be performed with an SMPS instead of CPC if evaporation from the particles is suspected.

B.) Field studies at low particle concentrations are made without a DMA upstream the CCNC. These measurements have their own issues.

A. Authors' response: We now state the scope of limitation of the paper clearly. The scope of this manuscript includes only CCN measurements that are used to calculate apparent hygroscopicity from critical supersaturation, which in turn is determined from activated fraction curves for monodisperse (or near-monodisperse) aerosol. Other setups can be used for CCN measurements, as the reviewer correctly points out. For example, if evaporation is a concern, an SMPS can be used to determine the size distribution of the aerosol particles. An SMPS can also be used in conjunction with unsized CCN measurements. A clarification has been added to section 3. Artifacts derived from CCN measurements (now titled "Artifacts derived from size CCN measurements").

Authors' changes to the manuscript:

Page 11 lines 173-178 the text now reads (additions are bolded): "CCN measurements **used for calculating apparent hygroscopicity from monodisperse aerosol** require accurate operation of three instruments: the CCN, the differential mobility analyzer (DMA), and the condensational particle counter (CPC). The scope of this manuscript includes only CCN measurements that are used to calculate apparent hygroscopicity from critical supersaturation, which in turn is determined from activated fraction curves for monodisperse (or near-monodisperse) aerosol. Other setups can be used for CCN measurements, as the reviewer correctly points out. For example, if evaporation is a concern, an SMPS can be used to determine the size distribution of the aerosol particles. An SMPS can also be used in conjunction with unsized CCN measurements."

B. Authors' response: The Reviewer is correct that a large number of field studies have been conducted without a DMA to size-select the aerosol prior to CCN measurements, Although these are somewhat different than the sized CCN measurements which are the focus of this paper, it is a good idea to mention them here. Examples which we now cite in the text include Jennings et al., 1996; Hudson and Xie, 1998; Modini et al., 2015; Duan et al., 2017; Schmale et al., 2018; Leng et al., 2013. However, our manuscript focuses on sized CCN measurements which may be used for the determination of κ_{app} . This is now clearly stated on page 10.

Authors' Changes to the manuscript:

Page 11 Lines 174-181 the text now reads: "This study considers sized CCN measurements which may be used for the determination of κ_{app} . In contrast, CCN studies are often conducted on the full ambient aerosol population without sizing the aerosol (Jennings et al., 1996; Hudson and Xie, 1998; Modini et al., 2015; Duan et al., 2017; Schmale et al., 2018; Leng et al., 2013). Instrument artifacts will first be assessed separately for the DMA, CPC, and CCN counter. In the concluding section of the paper (and Fig. 10), the overall uncertainty due to the combination of these is presented and discussed."

4). Reviewer comment: Another limitation is that the paper does not treat all the important artifacts.

A. An example is uncertainty in sizing due to evaporation of particle material or residual water in the particles while sized in the DMA. I can understand if this is out of scope for this article, but they could be mentioned.

B. Other sources of errors are closer to the focus of this paper and could be included or at least references made to papers discussing them. I am mainly thinking about three effects. The first is the role of doubly charged particles, especially in lab studies in which atomized aerosols can be overcharged in comparison to equilibrium charge distribution and radioactive sources normally used are not strong enough to neutralize the aerosol.

C. Voltage offset in the DMA is sometimes an issue, especially when working with high supersaturations and small aerosol particle sizes.

D. The role of counting variability due to sampling statistics at low concentrations and how it influences the determination of SSc in different cases (for example different flow ratios).

A. Authors' response: While we cannot address all possible artifacts in this manuscript, a new section has been added to the manuscript to address sources of artifacts that are beyond the scope of the study, but should still be considered (3.1.4 Additional artifacts resulting from DMA measurements). We have included a short discussion of particle evaporation and growth due to volatility and hygroscopicity, respectively.

Authors' Changes to the manuscript:

Pages 21-22 Lines 394-402 the text now reads: "Several other factors that may impact experimental κ_{app} are beyond the scope of this study, but are worth mentioning as they represent additional potential sources of error in some cases. First, volatile aerosols may partially evaporate inside the DMA, resulting in a decrease in particle size exiting the DMA. DMA sizing error due to aerosol volatility (defined as the ratio of sampled diameter to the selected diameter) increases with volatility, though sizing error can be decreased by increasing the sheath flow rate in the DMA. Conversely, hygroscopic aerosols may grow inside the DMA, resulting in larger particles existing the DMA. Operationally, errors in DMA sizing due to hygroscopic growth can be mitigated if aerosols entering the DMA inlet are in wet metastable states (higher aerosol RH at DMA inlet), and if DMA sheath flow rates are kept low (Khlystov, 2014)."

B. Authors' response: The reviewer has a valid point. On further consideration, we now include full consideration of one additional significant artifact, effects of multiple charging, in our analysis. We have added an analysis of multiple particle charges to the manuscript in a new section (3.1.3 Effect of double and triple charges on particles). In addition, 3 new figures have also been added (Figure S2 in supplement, Figures 5 and 6 in manuscript, all shown below).

Authors' Changes to the manuscript:

Pages 18-21 Lines 319-390 the text now reads: "The Grimm DMA employs a bipolar charger (also known as a neutralizer) to charge aerosol particles through the capture of gaseous ions. The analysis in Section 3.1.2 assumes that each particle carries a single (+1) charge. In reality, the methods used to charge particles prior to entering a DMA may impart two, three, or more charges to individual particles (**Fuchs, 1963**). The charge distribution resulting from a bipolar charger is roughly approximated using the Boltzmann law (**Keefe et al., 1959**). Additionally, the Boltzmann law assumes symmetric aerosol particle charging (equal concentrations of negatively and positively charged particles). Deviation from symmetric charging is observed in regions of high ionizations, and this deviation becomes more pronounced as particle size increases (**Hoppel and Frick, 1990**).

A more accurate estimation of stationary charge distribution has been calculated using an approximation formula for the charge distribution produced by a bipolar charger:

$$f(k) = 10^{\left[\sum_{i=0}^{i=5} a_i(k)(\log_{10} D_{nm})^i\right]} \quad (13)$$

where $f(k)$ is the fraction of particles carrying k charges, $a_i(k)$ are approximation coefficients determined using a least-squares regression analysis, and D_{nm} is the particle diameter in nanometers (Wiedensohler, 1988). The approximation coefficients only apply to particles with 0, ± 1 , and ± 2

charges. In a separate study, **Maricq et al., 2008** determined approximation coefficients for poly (α -olefin) oligomer oil droplets with ± 1 , ± 2 , and ± 3 charges. The approximation coefficients reported by these two studies were in excellent agreement for particles with ± 1 and in weak agreement for ± 2 charges (+2 and -2 charging efficiencies were overestimated by 50% and 100%, respectively). Therefore, this analysis will use the approximation coefficients from Wiedensolher, 1988 for particles with +1 and +2 charges, and the approximation coefficient for particles with +3 charge from Maricq et al., 2008.

In order to assess the impact of multiple charges on κ_{app} , Eq. (13) and the approximation coefficients from Wiedensolher, 1988 and Maricq et al., 2008 were used to calculate the charge distribution of the representative aerosol population shown in Fig. 3a. The resulting charge distribution is shown in Fig. S2a). The particle diameters in Fig. S2a were chosen to represent particles that would be present due to single, double, and triple charging for the DMA selected diameters 25, 50, 100, and 200 nm. The ratio of the concentration of double and triple charged particles to single charged particles is shown in Fig. S2b. An increase in multiple charging is observed as particle diameter increases, though this is offset somewhat by the decrease in concentration with particle size above 50 nm.

It follows that aerosols incorrectly sized due to double and triple charging will be passed from the DMA to the CCN and result in an additional uncertainty in the CCN measurements. To illustrate this, activated fraction curves, were generated for 25, 50, 100, and 200 nm sodium chloride particle selection by the DMA. The activation of sodium chloride is represented by sigmoid curves, where the midpoint of each activation curve is the κ -Köhler-derived critical supersaturation of sodium chloride, and the standard deviation of each curve is one-tenth of this value (consistent with the standard deviation/midpoint ratio observed from our instrument's ammonium sulfate CCN calibration data).

For each particle diameter D , the observed activated fraction, $AF_{D,weighted}^{SS}$, for each percent supersaturation SS was determined by weighting the activated fraction $AF_{D,i}^{SS}$ of each particle diameter/charge at that percent supersaturation, by the fraction of particles of that diameter:

$$AF_{D,weighted}^{SS} = \sum_{i=1}^3 \frac{\text{concentration of particles with charge } i \text{ and diameter } D}{\text{concentration of particles with charge } +1,+2,+3, \text{ and diameter } D} AF_{D,i}^{SS} \quad (14)$$

The raw data shown in Fig. 5 (green curves) can be corrected for multiple charging by determining the fraction of particles with $> +1$ charge from the lower plateau in each plot (dashed lines). The adjusted activated fraction for each percent supersaturation, $AF_{adjusted}$, is calculated using the equation:

$$AF_{adjusted} = \frac{AF_{raw} - AF_{plateau}}{1 - AF_{plateau}} \quad (15)$$

Where AF_{raw} is the raw activated fraction at that percent supersaturation, and $AF_{plateau}$ is the activated fraction corresponding to the lower plateau (**Rose, 2008**). The adjusted activated fraction curves are shown in Fig. 5 (blue curves), and agree with the theoretical κ -Köhler-derived activation curves for sodium chloride (not shown due to overlap with adjusted activated fraction curves). Critical supersaturation was determined for each diameter by calculating the percent supersaturation at which the raw $AF_{D,weighted}^{SS} = 0.5$. These critical supersaturations are shown in Fig. 6a, and the theoretical critical supersaturations calculated from κ -Köhler theory are shown for comparison. Eq. 4 was used to calculate apparent hygroscopicity for each particle diameter, shown in Fig. 6b. A dashed line in Fig. 6b indicates the literature value for κ_{app}^{NaCl} . It is apparent that failing to account for multiply-charged particle in the activated fraction curves shown in Fig. 5 leads to an overestimation of κ_{app} . Artifacts in κ_{app} are shown in Fig. 6c.

For the theoretical aerosol distributed used in this analysis (Fig. 3a), small, positive deviations from κ -Köhler theory and the literature value for κ_{app}^{NaCl} were observed ($0.01 \leq \kappa_{app,artifact}^{NaCl} \leq 0.04, 1 -$

3 % of κ_{app}^{NaCl}). As shown in the figure, κ_{app} artifacts resulting from unaccounted-for multiple charges decrease with particle diameter for this theoretical aerosol population. Greater κ_{app} artifacts would be expected for aerosol populations with more prevalent accumulation modes.”

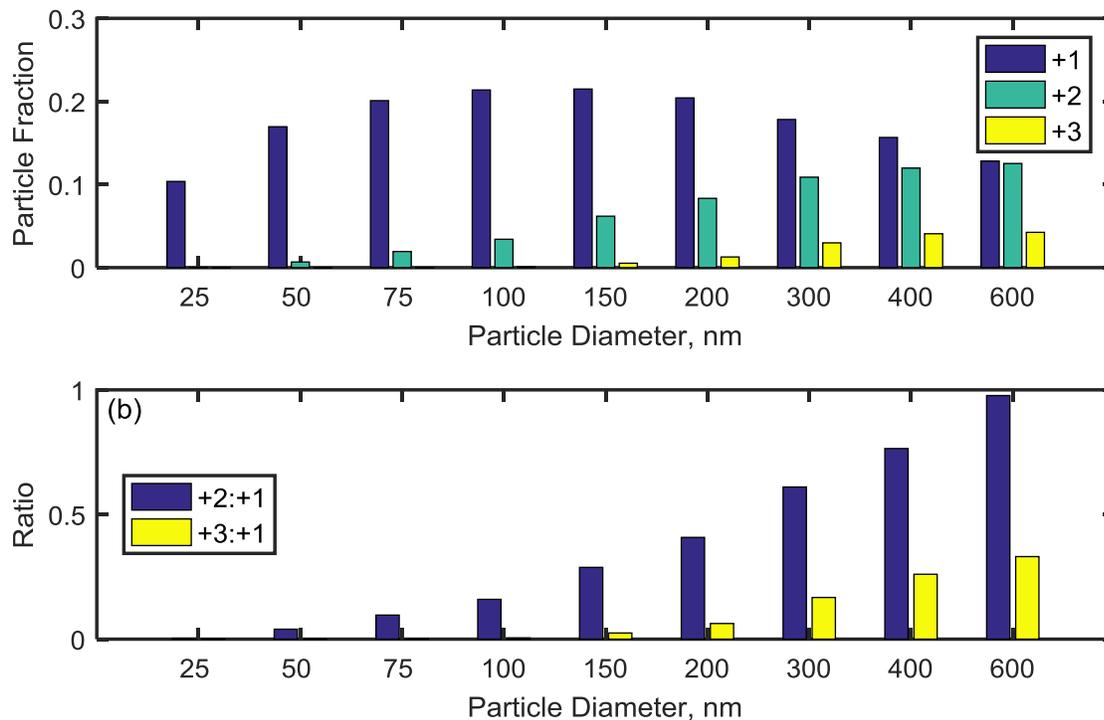


Figure S2 (a) Stationary charge distribution on particles shown in Fig. 3a. The particle diameters were chosen to represent particles that would be present due to single, double, and triple charging for the DMA selected diameters 25, 50, 100, and 200 nm. (b) Ratio of multiple charged particles to single charged particles.

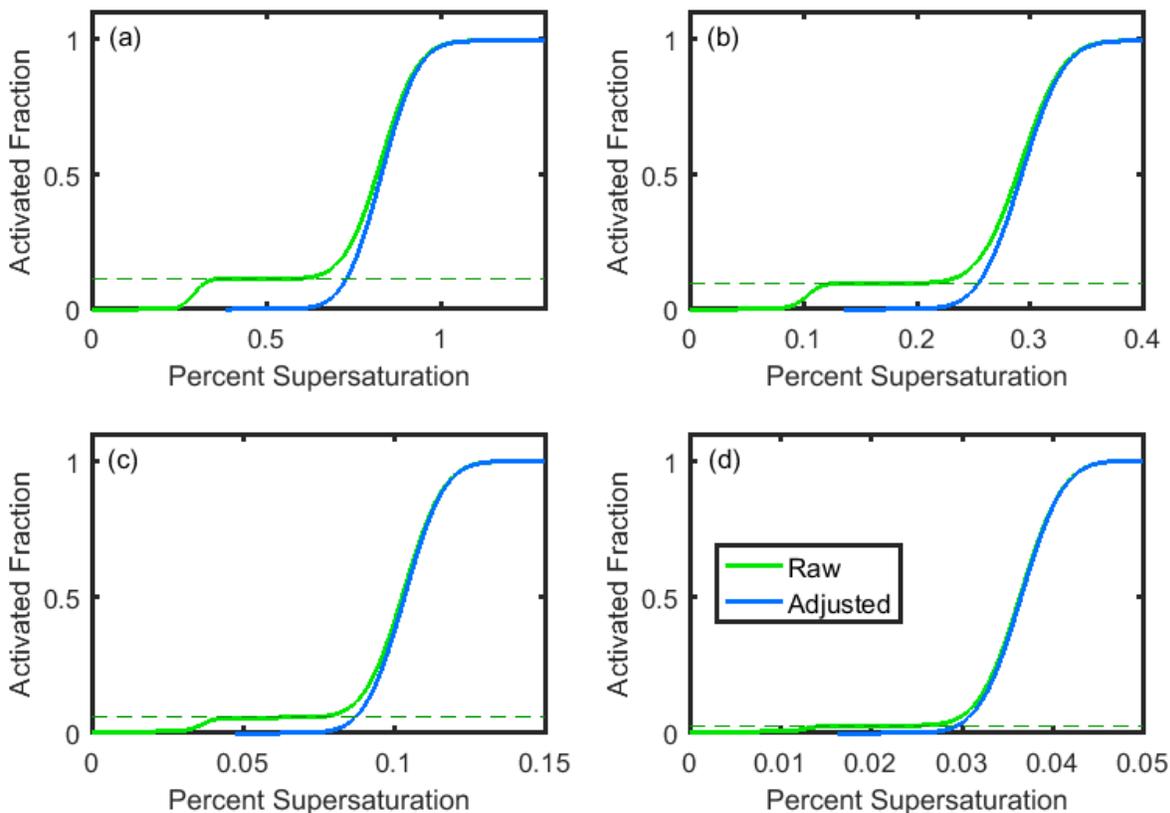


Figure 5 (a) Theoretical raw and adjusted activated fraction curves for single charged 25 nm particles, double charged 50 nm particles, and triple charged 75 nm particles; (b) single charged 50 nm particles, double charged 100 nm particles, and triple charged 150 nm particles; (c) single charged 100 nm particles, double charged 200 nm particles, and triple charged 300 nm particles; (d) single charged 200 nm particles, double charged 400 nm particles, and triple charged 600 nm particles. All particles are pure sodium chloride.

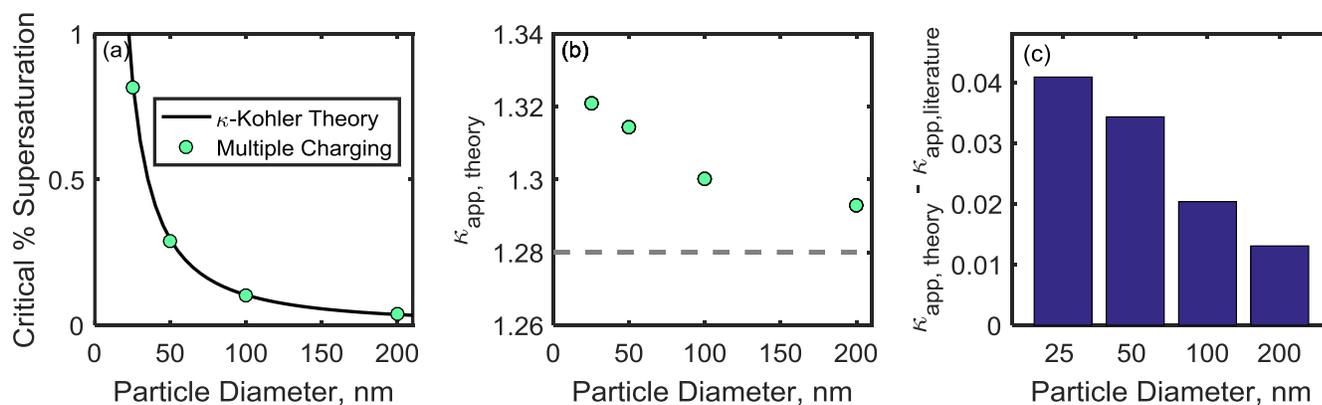


Figure 6 (a) The critical supersaturation of sodium chloride particles determined from the activated fraction curves in Fig. 5. A sodium chloride curve calculated using κ -Köhler theory is shown for comparison. (b) κ_{app} was calculated for the charge distribution; the gray dashed line indicates the literature value for the κ_{app} of sodium chloride. (c) κ_{app} artifacts resulting from multiple particle charges.

C. Authors' response: A new section has been added to the manuscript (3.1.4 Additional artifacts resulting from DMA measurements) that includes a short discussion of voltage shifts that may result from the space-charge field observed due to separation of the charged aerosol particles.

Authors' Changes to the manuscript:

Page 22 Lines 404-412 the text now reads: “Voltage shifts within the DMA (differences between the selected voltage and the actual voltage inside the DMA) can lead to discrepancies between selected and sampled particle diameters. Voltage shifts may result from a space-charge field generated by the motion of charges within the DMA. Particles charged by the neutralizer will either be attracted towards or repelled away from the inner column of the DMA, depending on whether they are positively or negatively charged. This charge separation creates a space-charge field which shifts the actual voltage within the DMA from the selected voltage. The impact of the space-charge field on the midpoint and spread of the DMA transfer function increases as particle mobility increases (as particle size decreases), and as particle concentration increases (Alonso and Kousaka, 1996; Alonso et al., 2000; Alonso et al., 2001).”

D. Authors' response: New text has been added to the manuscript in order to address this issue (6. Counting statistics in CCN and CPC measurements).

Authors' Changes to the manuscript:

Pages 29 Lines 542-544 the text now reads: “Sampling at very low particle concentrations ($< 200 \text{ cm}^{-3}$ total particle concentration) can introduce additional error into CCN and CPC measurements. This error can be mitigated by increasing scan times. For example, Moore et al., 2010 averaged CCN and particle concentrations over 5-second intervals for monodisperse particle concentrations $< 10 \text{ cm}^{-3}$, and increased averaging time to 20-second intervals when the monodisperse particle concentration reached $< 6 \text{ cm}^{-3}$.”

- 5) Reviewer comment: I also have a comment that might sound nerdy, but I find it important that we stick to the definition of an aerosol as a population of solid and/or liquid particles and the surrounding gas. Thus, we should not talk about aerosol size, when referring to the size of the particles. Please check the manuscript in line with this.

Authors' response: the reviewer is correct about the technical difference between aerosols and particles, but we respectfully disagree that the term needs to be changed in the manuscript. “Aerosol” is often used synonymously with “aerosol particle”. In keeping with convention, we will not change the current wording. Doing so would likely make the text less clear.

- 6) Reviewer comment: It is not clear how the critical supersaturation is determined: is it defined as the supersaturation at which $\#CCN/\#CN$ is 0.5 or when its value is 50% of the level reached at high supersaturation (the later often being used in experimental work)? This will in some of the examples make a large difference, and need to be discussed. An example is line 346-348.

Authors' response: Activated fraction data is fit with a sigmoid error function to determine the supersaturation at which 50 % of the particles have activated as CCN (activated fraction = 0.50), which is considered the operationally defined critical supersaturation SS_{crit} (Rose et al., 2008). An alternative approach is to define critical supersaturation as the value of supersaturation corresponding to 50% of the maximum observed value. However, this alternative method has a caveat in cases of external aerosol mixtures which include non-hygroscopic members. In such cases, the highest CCN/CN reached will be much lower than 1 (due to the inactive member aerosols) and thus the resulting value of critical supersaturation would be lower than the SS required to the active aerosol members to become CCN. A clarification has been added to the Background section of the manuscript.

Authors' changes to the manuscript: Page 8 Lines 125-127 the text now reads (clarification bolded): "Activated fraction data is fit with a sigmoid error function to determine the supersaturation at which

50 % of the particles have activated as CCN (**activated fraction = 0.50**), which is considered the operationally defined critical supersaturation SS_{crit} (Rose et al., 2008)."

Additional clarification was added to Section 4.1 CPC operation at low concentration.

Page 21 Lines 410-411 the text now reads: "Critical supersaturation was determined for each CPC case by finding the percent supersaturation at which activated fraction = 0.50."

- 7) Reviewer comment: Please check the plots with particle size distributions. The y-axis should be dN/dDp (with the unit $\text{cm}^{-3} \text{nm}^{-1}$) if a linear diameter scale is used. Why are you using a linear diameter scale and not a logarithmic? Also, see my comments to figure 6 and 7 below. I think that these are critical for the quality of the paper and the conclusions!

Authors' response: We see the Reviewer's point here. Employing dN/dDp is one of the standard way to plot an aerosol size distribution and readers are likely to be comfortable viewing such graphs. It is advantageous to view instrumental data in $dN/d\log Dp$ format, in which the normalization accounts for differences in the widths of size bins employed by various instruments and in various size ranges. However, our analysis is based on a theoretical aerosol distribution in which there are no disparate ranges in size bins to account for, and thus normalization is not necessary. For the purposes of this analysis, it is more straightforward to plot concentration in lieu of $dN/d\log Dp$.

In the original text, logarithmic scales were used for particle size distribution plots in Figures 8a and 8b (previously Figures 6a and 6b) and in Figure 9b (previously Figure 7b). A linear scale is used in Figures 3a and 3c because we feel that it clearly demonstrates "visual multiplication" (combining the distribution in 3a with the transfer function for 100 nm particles in 3b) performed in our analysis. In addition, given the small range of concentrations observed in Figure 3c, this is the clearest presentation of the data.

- 8) Reviewer comment: Also, make sure that the figure captions and legends are sufficient.

Authors' response: Several figure captions have been edited for clarity as specified below.

Authors' changes to the manuscript: figure captions have been changed as indicated below (changes are shown in bold).

"Figure 1 Experimental setup **used for obtaining sized** CCN and particle concentration measurements **from an aerosol sample.**"

"Figure 2 Simplified flow diagram of a DMA with an inner electrode radius r_1 , outer electrode radius r_2 , distance between aerosol inlet and sample outlet L , **clean sheath air flow Q_{sh} , aerosol flow Q_a , excess air flow Q_e , and sample air flow Q_s .**"

"Figure 3 (a) A theoretical aerosol distribution generated using a lognormal function centered at 50 nm. (b) The transfer function calculated using Eq. (7). (c) **Multiplying the distribution by the transfer function gives the downstream aerosol concentration (cm^{-3}).**"

"Figure 4 (a) Apparent hygroscopicity κ_{app} for **DMA cases 1-7 for sodium chloride (triangles) and ammonium sulfate (circles) [see legend in (b)].** (b) Critical supersaturation of ammonium sulfate and sodium chloride particles calculated using κ_{app} values derived in (a). Ammonium sulfate and sodium chloride curves from κ -Köhler theory are shown for comparison. **Legend colors apply to both salts.** (c,d) DMA-flow-derived artifacts in κ_{app} are shown for each DMA case and both salts."

"Figure 7 (a) Counting efficiency curves for CPC **Cases 1-6 (shown in Table 3).**

(b-e) CCN activated fraction curves for 25, 50, 100, and 200 nm NaCl, respectively. (f) Critical supersaturation calculated for each **particle diameter**. (g) Theoretical κ_{app} for each CPC case **and particle diameter**. (h) Artifacts in κ_{app} for each CPC case **and particle diameter**.”

“**Figure 8** (a) Theoretical relationships between the reference aerosol concentration and CPC concentration. (b) Concentration-dependent counting efficiencies **from (a)** were applied to four theoretical aerosol distributions. (c-f) Activated fraction curves for **CPC Distribution 1 and** particle diameters 25, 50, 100, and 200 nm NaCl aerosol, respectively. (g,h) Critical supersaturation and κ_{app} for each case. (i) Artifacts in κ_{app} for each case.”

“**Figure 9** (a) Counting efficiencies of the DMT CCN-100 **for specific supersaturations**. (b) Lognormal aerosol distributions used to study CCN undercounting at high concentrations. (c-f) Activated fraction curves for 25, 50, 100, and 200 nm NaCl particles. Supersaturation-specific counting efficiencies from (a) applied to theoretical sigmoid curves for NaCl CCN activation. Activated fraction in the case of 100 % counting efficiency is shown for comparison. (g) Critical supersaturation for each case. (h) Theoretical κ_{app} calculated for each case. (i) Artifacts in κ_{app} artifacts for each case.”

9) Reviewer comment: Figure 6 b. As I understand it, the CPC counting limitations relevant here relate to the number concentration after the DMA.

A. Are the DMA transfer function and the charge distribution taken into account when determining these curves? And if so, for which aerosol to sheath flow ratio are they made?

B. Is it just a coincidence that the size distribution is cut at the same value of dN/dDp (in $\text{cm}^{-3} \text{nm}^{-1}$?) as the CPC concentration saturates (in the unit cm^{-3}). And how can the “saturated size distribution” be a horizontal line? Both charging probability and transfer function width (in a linear scale) are size dependent.

A. Authors’ response: Yes, the CPC instrument is positioned after the DMA. The DMA transfer function is not taken into account in the CPC concentration analysis here.

Page 11 Lines 179-181 the text now reads: “Instrument artifacts will be assessed separately for the DMA, CPC, and CCN counter. In the concluding section of the paper (and Fig. 10), the overall uncertainty due to the combination of these is presented and discussed.”

B. Authors’ response: This is not a coincidence; this concentration is the concentration at which the CPC can no longer count additional particles (and is not related to charging probability or the transfer function). An additional sentence has been added to the manuscript for clarification.

Page 26 Lines 477-479 the text now reads: “It should be noted that the CPC concentration (the concentration that would be measured and recorded by the CPC in Cases 7-10) levels off at the saturation concentration for each case.”

10) Reviewer comment: Figure 7 and the calculations behind them: How is dN/dDp transferred into a concentration after the DMA? Which flows are used?

Author’s response: To clarify, we have only used concentration in this manuscript, not dN/dDp . If we understand this question correctly, the reviewer is referring to the fact that particles are lost in the DMA which leads to error in particle concentration measured by the CPC placed downstream of the DMA. Indeed, particle losses within the DMA are substantial. If aerosol concentration were the

measurement objective then a more accurate measurement of concentration would simply use a CPC, thus minimizing particle losses. However, for the purposes of this manuscript, we consider activated fraction (the ratio of particles measured by the CCN instrument to those measured by the CPC for a given particle diameter and supersaturation). In this case, losses in the DMA impact the CPC and CCN concentrations exactly the same way, so no error is introduced in the resulting activated fraction. In this analysis, the actual CCN and aerosol particle concentrations are only used for determining how activated fraction is effected in each CPC or CCN case, if at all. We acknowledge that estimation of particle losses in the DMA, and between the DMA and CPC/CCN, are worth considering but the treatment of these particle losses is beyond the scope of this manuscript.

11) Reviewer comment: You use both saturation ratio and supersaturation in the theoretical discussion. As I can see you are using them correctly, but sometimes you use only saturation for saturation ratio. I would recommend that you stick to saturation ratio in order to avoid confusing the readers.

11. Authors' response: We thank the reviewer for finding this error. All mentions of "saturation" have been revised to "saturation ratio".

12) Reviewer comment: Line 184. With a truly monodisperse aerosol the concentration would also be 0.

12. Authors' response: This is true, and the sentence indicated has been changed for clarification.

Page 13 Lines 212-213 the text now reads: "In other words, Q_s would ideally consist only of aerosols with diameters equal to, or very nearly equal to, the selected diameter."

13) Reviewer comment: The discussion and the conclusion section is mainly a repetition of the results (which might well be a part of these sections), but I would have liked to see a discussion on what should be considered good practice in CCN measurements, based on this work and the literature.

Authors' response: Good practice in CCN measurements was originally addressed in the Conclusion section of the original manuscript, in terms of minimizing kappa artifacts. Based on the reviewer's comment, we have expanded on the discussion and conclusions, including references to DMA sample/sheath ratios recommended in other studies for comparison.

Authors' changes to manuscript:

Page 32 Lines 580-584 the text now reads: "These demonstrate that limiting Q_a/Q_{sh} to ≤ 0.10 will result in a narrow particle size distribution downstream of the DMA. Other studies have recommended employing DMA sample/sheath ratios of 0.2 (Petters et al., 2007; Carrico et al., 2008; Moore et al., 2010) or 0.1 (Moore et al., 2010; Zhao-Ze and Liang, 2014) in order to minimize measurement aerosols due to transfer function broadening."

Page 32 Lines 586-591 the text now reads: "The effects of multiply-charged particles on κ_{app} calculations were also quantified. Small, positive κ_{app} artifacts (1 – 3 % of κ_{app}^{NaCl}) were observed when particles with +2 and +3 charges were not accounted for. This analysis considered a theoretical aerosol distribution in which most of the particles measure less than 100 nm in diameter. Actual aerosol distributions vary temporally and spatially, and often include accumulation and coarse modes that would result in larger κ_{app} artifacts."

Page 33 Lines 611-615 the text now reads (additions are bolded): The combined artifacts for the cases where the highest artifacts were observed (DMA Case 4, **multiple particle charging**, CPC Case 4, CPC Case 10, CCN Case 1) are 0.21, 0.24, 0.32, and 0.21 for 25, 50, 100, and 200 nm particles respectively, as shown in Fig. 10. The combined artifacts for the lowest-artifact cases (DMA Case 2, CPC Case 3, and CCN Case 4) are < 0.001 except for 200 nm particles, where $\kappa_{app,artifact} = 0.0013$.

Page 36 Lines 670-672 the text now reads (**additions are bolded**): “Under optimal operating conditions, where the DMA sample/sheath ratio is 0.10 and excess/sheath ratio is 1.0, and in the absence of undercounting by the CPC or CCN, uncertainties in κ_{app} are within $\pm 1.1\%$ for 25 to 200 nm particles. When the DMA sample/sheath ratio drops to 0.05, κ_{app} uncertainties decrease to $\pm 0.01\%$. Additionally, errors in activated fraction (and therefore κ_{app}) resulting from the bipolar charge distribution can be corrected by determining the fraction of particles with multiple charges.