General Comments

This article presents a new algorithm for processing measurements from a Broad Supersaturation Scanning Cloud Condensation Nuclei (BS2) system. The outcome of this algorithm is to improve retrieval of aerosol hygroscopicity parameters, namely the particle hygroscopicity, κ. The article claims that this algorithm provides a unique solution to a known problem in determining κ: namely, that multiply-charged particles that pass through the differential mobility analyzer (DMA) in a BS2 system result in misshapen particle activation curves which degrade the retrieval of κ. Despite the claim of novelty, the algorithm bears a rather close resemblance to the proposed methodology of Moore et al (2010).

In general, the Methods section is missing sufficient detail for their method to be utilized and reproduced by other researchers. Some issues are purely technical: the authors need to re-work the notation of the Methods section. There are several instances where the notation is not appropriate, misleading, or definitions are missing altogether. Other issues are pragmatic: further descriptions of their BS2 system should be included (rather than referenced) such as impactor size, DMA size detection range, etc. This is, essentially, a methods paper and the Methods section is perhaps the weakest point of the current manuscript. It should be spelled out to the letter what a researcher needs to do to implement this method.

The results demonstrate a fulfillment of the original promise. The appearance of multiply charged particles in the activation curve have disappeared. However, the assessment of this methodology is fairly qualitative. The case study approach is not sufficient enough argument for researchers to understand when this correction needs to be applied to their measurements. It is clear, for example, that the correction algorithm need not be applied to calibration experiments. A revision of this manuscript should include a more quantitative laboratory-based study with ammonium sulfate rather than the qualitative field-based study that is currently used. The revision should also include a full uncertainty analysis to determine confidence intervals on derived hygroscopicity. This would allow other researchers to better understand when they should apply this correction and the magnitude of the effect on hygroscopicity retrieval (so that they can troubleshoot their implementation). The authors should also make it more apparent in the abstract and conclusions that the proposed algorithm assumes that the particle size distribution is monomodal. The algorithm has not been tested for more complicated PSDs.

Finally, the authors should support their claim that their methodology is a necessary improvement on previous approaches. A revision of this manuscript should also include a side-by-side comparison of this methodology to previously proposed methods in the literature, e.g. Moore, Nenes & Medina (2010) to which the proposed method bears an uncanny resemblance.

Specific Comments

Introduction. The introduction does a good job of introducing source material and identifying where this manuscript is positioned within the scientific literature. However, the importance of obtaining the hygroscopicity parameter, k, should be introduced much earlier so as to better guide the narrative flow of the rest of the introduction. Currently, the introduction seems very technical with no obvious goal.

Methods. There are many minor and major technical issues in the Methods section that must be fixed. In general, \mMore detail is needed such that others can apply this method to their work. By section & subsection:

Section 2.1 The use of "υ" as the number of elementary charges is unusual and confusing. The variable "υ" ought to be reserved for kinematic viscosity in this context. Some authors use φ to represent the number of charges, e.g. Collins, Flagan & Seinfeld (2003). Next, the authors omit the definition of the set mobility, Z^*_p, which is not at all equivalent to Z_p. This might not be obvious to a reader at first pass, so Z*p must be defined. Third, it may be more correct to say that Wiedensohler (1986) developed an empirical model based on charging theory of Fuchs (1963). Additionally, Wiedensohler's model is only valid up to ±2 charges. Beyond this number of charges, it is common to use Gunn (1956). As you evaluated charging probabilities for +3 charges, you would need to apply the formulae described by Gunn.

To avoid confusion G_v should be redefined:

$$G_v(D_p, x) = F(x, v)\Omega(x, v, D_p)$$

Without the summand. It should also be specified how $\Omega(x, v, D_p)$ was calculated. What processes/efficiencies are involved in its calculation? Penetration efficiency? Impactor Efficiency? Diffusive losses? This is a methods paper. More detail is needed such that others can use this tool.

Section 2.2. I am wondering if it needs to be specified if the data are collected simultaneously on a single computer or if two separate computers are used and the data are analyzed offline. If the latter is the case, there must have been some need to make sure that the clocks were aligned for 1 Hz measurement. If so, specify!

Step 1. Are these variables named correctly? The use of C implies that these are counts, not concentrations (N). Further, you should be more specific about what algorithm is used to compute the inversion. It is not sufficient to say that private software is used (do you mean the AIM software?). In Section 3, you also mention that a lognormal size distribution is assumed in this step. If it is, specify this here.

Step 2. As written, this is open to misinterpretation. Is the DMA set at a "single" particle size for 40 s, you omit the first 15 s and average the latter 25 s? Or, are you saying that the DMA is set at a "single" particle size for 25 s, you omit the first 15 s, and average the last 10 s? Clarify please.

Step 3. There is more than one way to interpolate between two points… linear? Polynomial? Spline? Be specific! It is also worth asking: what is the goal of interpolation at this step? Is it to determine h(x, v, D_p)? If so, specify. Additionally, it's not obvious that the activated fraction should depend on the number of charges. In my later comments, I rearrange your integral.

Step 4. Did you actually integrate Equation (4) out to infinity? I am guessing not… It is common to integrate up to some factor of the impactor cut-off, D_{50}. Specify the actual upper bounds of your integration. Additionally, I believe that Gv is incorrectly defined. Based on what this step says, I assume that there was an intermediary step of calculating the following:

$$G_{+2}(D_p, x) = F(x, +2)\Omega(x, +2, D_p)$$

This is different than the current definition of G_v, which would include all charges as defined in the manuscript (See my comments on Section 2.1). After calculating G_{+2}, I assume that one would multiply G_{+2} by the proposed solution spectrum n(x) and integrated to retrieve the number of doubly-charged particles of size Dp, $N_{+2}(D_p)$, i.e.:

$$N_{+2}(D_p) = \int_0^\infty n(x) G_{+2}(D_p, x) dx$$

Either re-define G_v or be more explicit.

Step 5: Again… What is the upper limit of charges that were considered? You state that no more than 3 charges are calculated, but it should be specified in this step. I think this is a justifiable cut-off step given the limited range of 300 nm, but you should be more quantitative in your argument for why you terminated the sum at +3 (e.g. less than X% of particles have charge >3 at the impactor cur-off diameter). Additionally, charge is discrete, so integral notation is inappropriate. This should read:

$$N_{CCN}(D_p) = \int_0^\infty n(x) h(x, D_p) \sum_{v=1}^{+3} G_v(D_p, x) dx$$

At this stage in my review, I am beginning to wonder how different this procedure is from the SMCA proposed by Collins et al (2010). Is it unique because you are using slightly different flow rates and supersaturation settings? Or is it unique according to Step 5?

Application
As noted above, the log-normal fitting procedure should be described in the Methods, not the results.

Section 3.2. What is dT? Spell it out, please. You should also be more descriptive/quantitative of your test distribution. What was the value of the geometric mean diameter and **geometric** standard deviation? Even if the reader can go read your other paper (Kim, 2021), re-iterate the suggested limits of D_g and σ_g for a "well-performed" calibration experiment here. Finally, Table 1 could probably be added to a Supplemental Information document, I don't think there is any benefit to it appearing in the main text.

Section 3.3. It should be noted that that ambient aerosol size distributions are rarely monomodal, like this retrieval assumes. The authors should thus comment on what environments, if any, it is safe to assume that the distribution is monomodal. In the marine environment, the particle size distribution is often at least tri-modal.

The referral of the reader to another paper for information about your campaign is frustrating. Basic information like the duration of the campaign should be specified. The selection of cases should also be detailed and better motivated. It is clear after reading this section that you want to highlight the degree to which D_g determines the magnitude of the effect your multiple-charge correction will have, but this should be spelled out at the beginning of the section.

I also believe that this effect could have been better studied directly in the lab with particles of known hygroscopicity, e.g. ammonium sulfate, by modifying the atomizer pressure & solution concentration. This would allow you to directly relate the deviation in k as a function of Dg. Uncertainty analysis could then allow you to determine a threshold Dg below and above which your correction should/shouldn't be applied. This would provide your readers with a much more satisfying quantitative assessment than the relatively qualitative case study that is presented.

Next, F_{act} is a function of particle size. Are you saying that the average value of F_{act} decreased from 0.01 to 0.07? Or, are you referring to a specific value of F_{act} at a diameter of 60 nm? Follow-up question: How can something decrease from 0.01 to 0.07?

I would like to see a proper error analysis added to this section. It is clear that your algorithm results in changes to retrieved values of hygroscopicity, k. It also seems to complete the task as advertised, removing multiply charged particles from the minor plateau, but are these changes significant? Add 95% C.I. to your estimates of k in Figure 6. This will help quantitatively reinforce your claim that multiple-charge corrections need only be considered when the peak of the observed size distribution is >100 nm.

Finally, this manuscript is staking a claim that the algorithm uniquely solves the multiple-charge correction in the context of SMPS+CCN measurements. It would be useful to actually demonstrate that this is true, since the algorithm bears such close resemblance to the Collins, Nenes & Medina (2010). The algorithm seems to predict the greatest change when Dg is >100 nm. It would be useful to do a side-by-side comparison of data processed by the proposed algorithm and existing algorithms in the literature.

Discussion. Uncertainty/Error analysis should also inform the discussion of Figure 7. This would help support your claim that these deviations are meaningful by demonstrating that they are statistically significant.

Final Note: As it currently stands, you have demonstrated that your correction algorithm works for monomodal aerosol. It is a rather large stretch that you finish this manuscript by saying it should be applied to "a variety of particle number size distributions." Remove this statement or prove it.

Technical Corrections

Line 28: "~~the~~ **a** key element". I disagree with the notion that clouds are the sole element controlling climate change.

Line 29: Change to "Despite the scientific importance of CCN,"

Line 30: "aerosol-cloud interaction**s**". There are many types of aerosol-cloud interactions.

Line 31: Citation needed at the end of "composition and processes." Perhaps the updated version of this figure: https://archive.ipcc.ch/publications_and_data/ar4/wg1/en/tssts-2-5.html.

Line 33: "~~over the past~~" → "in recent"

Line 40: "under the simple assumption" Are you referring to a specific assumption here? ZSR? Or, did you mean to say "under **this** simple assumption."

Line 51: On first introduction, explain what D_c is.

Line 52: "Constant fraction ~~by~~ **of** doubly charged particle**s"**

Lines 54: Not sure if it is worth mentioning that the process starts from the largest aerosol size bin and iterates towards smaller bins.

Line 55: Suggested rephrasing: "Ultimately, each of the methods introduced above are designed to determine the critical activation diameter, Dc, of the test aerosol and thus the hygroscopicity, k, of the aerosol." Additionally, it should be outlined at the beginning of this paragraph that the hygroscopicity parameter, k, is the desired outcome.

Line 57: You should introduce the theme of this paragraph in the first sentence. "Whereas previous studies have improved hygroscopicity retrieval through the development of post-processing algorithms, modern studies have focuses on directly manipulating the sampling parameters (e.g. sample flow rate, sheath flow rate, supersaturation, etc.) to allow direct retrieval of k. Examples of this approach include…"

Line 58: What is the calibration experiment? Either describe or omit.

Line 68: Do you mean continuous as opposed to discrete? Or continuous as in "temporally continuous". It might be beneficial to describe as "continuously variable".

Line 82: What is Dp? Be thorough and describe.

Line 84: This equation is valid, but what is Z*p?
$Z^*_p = (Q_{sh}/(2*\pi*V*L))*\ln(r_2/r_1)$