Review of:

Characterization of tandem aerosol classifiers for selecting particles: implication for eliminating multiple charging effect

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General comments:

The manuscript by Song et al. examines the problem of multiple charging when using tandem aerosol measurement methods. Specifically, the authors investigated the combinations of a differential mobility analyzer (DMA) with either a centrifugal particle mass analyzer (CPMA) or aerodynamic aerosol classifier (AAC); DMA-CPMA and DMA-AAC, respectively. The authors modeled the transfer function of all instruments to derive a limiting case below which it would not be possible to resolve the contributions of particles bearing multiple charges and refer to this as PP0. Using flame generated soot, the authors then demonstrate the ranges of mobility diameters (D_m) where the particle contributions as a function of charge can be isolated.

The technical quality of this manuscript is not very good; the authors appear to have performed their transfer function calculations incorrectly.

Specific AMT review criteria:

1. The manuscript could represent a substantial contribution to scientific progress within the scope of Atmospheric Measurement Techniques and this is specifically highlighted by the derivation of Equations 25 and 27 for the limiting cases for the complete separation of multiply charged particles. In my opinion, the authors need to refer to these cases as something better than PP0 and make mention them in the abstract since this seems to be the most important takeaway. However, the clarity of the presented results is lacking, and it appears the presented transfer functions are not correct, which severely detracts from the manuscript. Rating: poor

2. The scientific approach and applied methods seem valid, but not necessarily the calculations, with limited discussion of the results but in an appropriate and balanced way. Rating: fair

3. The presentation of the scientific results and conclusions needs significant improvement. The use of English language is not fluent or precise in places and this distracts from the information the authors are trying to convey. Additionally, there are sections of the discussion that should be significantly expanded, and this expansion should aid in clarity. Rating: fair

This manuscript is likely publishable after significant revision since the topic is of interest to the community.

Specific comments:

1. I’m really struggling to understand the figures because of the rainbow color scale used and I strongly recommend using a different color scheme. Additionally, this color scheme is not visually accessible to all readers; *e.g.* (Nuñez, Anderton and Renslow 2018).

2. Uncertainties are missing on reported values throughout.

3. The authors investigated the pairwise combinations of DMA-CPMA and DMA-AAC but some mention of their expectations when utilizing other orderings (AAC-CPMA, AAC-DMA, DMA-CPMA) is needed. This is especially true for an AAC-DMA since the ratio of the β's may depend upon this ordering. (i.e. does it matter whether the transfer function of the first instrument is narrower/similar/wider than the second?)

4. It appears that the calculated transfer functions do not include the effect of the mass to charge ratio on the resolution of the CPMA in Figures 2, 5 and S2; effective masses, not absolute masses are the key quantity being measured.

For example: in Figure 5a, the authors state that D_m = 80 nm, m = 0.16 fg and D_{fm} = 2.28. Solving the $\underline{D}_{fm}$ relationship yields $\rho_f = 7.3 \times 10^{-6}$ fg nm$^{-2.28}$. At D_m = 120 nm, m ≈ 0.40 fg which agrees with the data shown in the figure. Unfortunately, because q = 2, the effective mass would be a factor of 2 lower and should be ≈ 0.2 fg. It appears that a similar error is present in Figures 2 and S2.

5. Figure 4 appears to correspond to utilizing an AAC-DMA combination rather than a DMA-AAC as is discussed in the text. In the DMA-AAC, you'd get two populations of particles in D_{ae}-space since the distributions of particles have equivalent Z_p exiting the DMA but are physically different sizes with different q. In the AAC-DMA, you'd only have one population in D_{ae}-space, the AAC selects one physical size, and then that distribution would have multiple charge states exiting the DMA.

5. The authors fit the distributions of aerodynamic diameter (D_{ae}) utilizing multiple log-normal distributions, but it doesn't seem to me that they have the resolution to constrain these values even though we know that multiple charges are present *a priori*. More discussion of the fitting routine is necessary. For example, in Fig. 5b, a) How does the fit compare to using just a single log-normal distribution? Or a single Gaussian or summation of multiple Gaussian distributions? b) Were the central values of D_{ae} constrained prior to the fit? Or were they allowed to float? c) Were the widths of the distributions constrained in any way prior to the fit? d) What are the magnitude of the uncertainties in each of the fit coefficients? e) Were the uncertainties in particle number densities included in the fits? f) The peak of the distribution is significantly underfit. Is it possible that more than q = 1 and 2 are contained within the primary peak and what was identified as q = 3 is 4 or higher?

Technical corrections:

Line 15: "effect"

effects

Line 16: "technique"

techniques

Line 18: "the potential multiple charging effect"

Elaborate.

Line 19: "remove"

Resolve?

Line 20: "instruments setups of DMA-CPMA system"

What is meant by instrument setups? Elaborate.

Line 23: "DMA-AAC can eliminate multiple charging effect"

This is not strictly correct as written since you can only "remove" the multiple charging artifact when used in a static configuration. In a scanning mode, the contributions would be resolvable.

Line 24: "while particles with multiple charges can be selected when decreasing resolutions of DMA and AAC"

Confusing as written.

Line 25: "We propose that the multiple charging effect should be reconsidered when using DMA-CPMA or DMA-AAC system in estimating size and mass resolved optical properties in the field and lab experiments."

This statement is not clear as written. How should the effects be "reconsidered"?

Line 35: "is the most commonly used size classifier"

If the DMA is the most commonly used classifier, why is only the original Knutson and Whitby reference provided?

Line 38: "particles are required to be pre-charged"

In what sense do they need to be pre-charged?

Line 39: "resulting in that particles"

Delete "that".

Line 41: "subsequence"

Subsequent

Line 49: "This conclusion implies that it can hardly to achieved that all the multiply charged particles are effectively excluded for aspherical particles, especially for soot particles."

Grammar makes this sentence confusing.

Line 51: "conducted"

Investigated?

Line 52: "ammonia"

Ammonium

Line 60: "dynamic shape factor (χ), can be inferred…"

The measured χ may depend upon the combination of instruments used. See (Yao et al. 2020) and potentially Table 2 here.

Line 80: “particle”
Elementary

Line 89: “The transfer function is an isosceles triangle with value of 1 at Zp * and going to 0 at (1 ± βDMA)·Zp*.”

I think it is important to mention that this translates to asymmetric distributions in Dm and mp since their relationship with Z_p is nonlinear.

Line 106: “is much simpler and more robust”

Elaborate.

Line 107: “radical”

Radial

Line 113: “should”

would

Line 134 and Line 328: “nominated”

Nominal?

Line 142: “80, 100, 150, 200”

Missing units.

Line 146: “while the condensation particle counter”

Was only a single CPC used during the soot characterization experiments? In the previous sentences, the author make it seem like the measurements were made simultaneously and in parallel. Also, please include flow rates.

Line 147: “concentration”

Number density of particles. Concentration is assumed to have units of mole per unit volume.

Line 148: “fitted to log-normal distribution”

Please include the equation that you used to fit your distribution since there are many ways to define the same relationship. Also, considering the shapes of the distributions shown in Fig. 5 and S2, why was a log-normal distribution utilized? The distributions appear symmetric, and they’re plotted on a linear axis, so some justification is warranted.

Line 149: “70, 150”

Missing units.

Line 151: “density of PSL”

Please enumerate.

Line 153: “effect of particles selected by DMA-CPMA system, the *d*ae distribution of twice classified particles”.

Please provide additional information about this portion of the procedure.

Line 167: “we explain the transfer functions of DMA-CPMA and DMA-AAC utilizing the literature data of soot particles”

When the transfer functions were calculated, what range of parameters were used? And how exactly were the transfer functions solved? Iteratively or something else? If iteratively, what was the Δt for each step and the number of individual trajectories considered? These details need to be provided somewhere in the manuscript.

Line 178: “representing pre-exponential factor (*ρf*)”

Having the pre-exponential factor share a variable with effective density (ρ_{eff}) is confusing since I would expect them to share units when they do not; the units are (g nm^{-Dfm}) and (g nm^{-3}) for ρ_f and ρ_{eff}, respectively. Additionally, I recommend including a normalization factor in the Dm term to avoid having fractional units, *e.g.*, D_{fm} = 2.28 will have ρ_f with units of g $nm^{-2.28}$.

Line 182: “however,”

However is used twice in the same sentence.

Line 184: “In the exemplary case, the derived *D*fm of premixed flame generated soot particles was 2.28,”

What study does this refer to? Reference?

Line 189: “The DMA-CPMA system can eliminate the multiply charged particles only if the *D*fm of particles is larger than the slope of a line connecting line connecting (*d*m, *m*) = (*d*m2,min, *m*2,max)(*d*m1,,*m*1) (as PP0 shown in Fig. 2).”

The point (dm2, m2) appears to correspond to the actual mass and dm of a particle bearing 2 charges instead of the effective mass (m/2). This is unclear and has significant implications for the calculated transfer functions and resultant discussion since the effective mass is ultimately what affects instrument resolution.

Line 190: “line connecting (*d*m, *m*) = (*d*m2,min, *m*2,max)(*d*m1,,*m*1) (as PP0 shown in Fig. 2).”

I’m assuming that the location of (dm1, m1) is point P0 and is at the center of the q = 1 transfer function? This is not clear in the figure. There’s an extra “,” after dm1 in the text.

Line 201: “are necessary to reduce the potential of multiply charged particles”

By increasing the resolution of the measurement?

Line 205: “PP0 of 3.55 was derived when DMA-APM selects the same example soot particles”

Compared to what?

Line 208: “critical slope of PP0”

Z-axis in Figure is labelled as “The slope of PP0”.

Line 210: "when the *D*fm of particles is larger than the slope of PP0 which is represented as background color."

So, the color of the data point will be red shifted relative to the background when multiple charging is not affecting the output distribution? This is unclear and the rainbow color scheme isn't helping.

Line 213: "Fig. 3, the *d*m, *m* and corresponding *D*fm were taken from literature"

In the caption, the authors mention that the shapes correspond to the individual studies. Please elaborate which is which here?

Line 214: "Generally, multiple charging effect can be avoided for DMA-CPMA to select soot particles with diameter larger than 200 nm.,"

I don't think the authors can conclude this without providing more data to support the claim. For fresh soot, my experience has been that multiple charging can be a problem at almost all Dm; e.g. see Figure S1 of (Radney et al. 2014). Also, there's a "," after the "." at the end of the sentence.

Line 215: "diameter"

To which diameter metric are you referring?

Line 216: "eliminate"

Resolve?

Line 220: "Therefore, the multiple charge effect could be avoided theoretically."

Measurements by just an AAC will avoid multiple charging. Multiple charging only becomes a problem again when the tandem measurement is a DMA or PMA.

Line 226 to end of paragraph: "In order to simulate the transfer function of DMA-AAC selecting the same particles as that used in calculations of DMA-CPMA. The corresponding *d*ae…"

See Specific Comment 5.

Line 242: "resulting in the minimum D_{fm} of 1.41, which is the case for most atmospheric aerosol particles."

Is this the black dashed line drawn as PP0? If so, please label in figure. Also, please differentiate these dashes from the vertical and horizontal ones.

Line 245: "sheath flow rate of classifier is restricted by the instrument design"

It's also important to note that sheath flow restricts the maximum size ranges.

Line 252: "uncertainties"

Uncertainties does not seem to be the correct word here. Biases?

Line 255: "*D*fm was 2.28"

What was the value of ρ_0?

Line 258: "along"

Delete word.

Line 261: "80, 100, 150, 200"

Units?

Line 263: "which suggested that multiply charged particles are still classified in this circumstance."

This suggests that the contributions from the multiply charged particles can't be resolved.

Line 268: "the multiply charged particles can be resolved in aerodynamic size distribution"

I disagree that the q = 1 and 2 can be resolved. Please provide more evidence to support this claim.

Line 270: "PNSDae was fitted using log-normal distributions and three peaks which correspond to singly, doubly and triply charged particles were identified."

What does the peak at Dae < 40 nm correspond to? Please mention.

Line 296: "Mie theory was used to calculate the theoretical absorption coefficient at the wavelength of 550 nm."

Mie theory probably isn't the "best" method to use here since soot particles are aspherical agglomerates. Realistically though, the Mie comparison is only being used to prove a point about the impact of multiple charging. So, in this instance any errors in the calculated optical properties are somewhat inconsequential. Some mention of this nuance should be mentioned.

Line 301: "integral concentration for particles"

Integrated number density of particles. Concentration is assumed to have units of mole per unit volume.

Line 308: "70"

Units?

Table 2: It'd be interesting (but not necessary) to include a comparison of the derived shape factors (χ) for each method.

Figure 2: "Example of DMA-CPMA transfer function"

Transfer function for what? Soot?

Figure 3: The coloration of this figure is very hard to understand.

Figure 4: Z-axis is labelled "Transfer function of DMA-CPMA" and the caption says DMA-AAC.

Figure S1: Having the Z-axis go all the way to zero is confusing.

References:

Nuñez, J. R., C. R. Anderton, R. S. Renslow. 2018. Optimizing colormaps with consideration for color vision deficiency to enable accurate interpretation of scientific data. *PLOS ONE* 13:e0199239. doi: 10.1371/journal.pone.0199239.

Radney, J. G., R. You, X. Ma, J. M. Conny, M. R. Zachariah, J. T. Hodges, C. D. Zangmeister. 2014. Dependence of soot optical properties on particle morphology: Measurements and model comparisons *Environ. Sci. Technol.* 48:3169-3176. doi: 10.1021/es4041804.

Yao, Q., A. Asa-Awuku, C. D. Zangmeister, J. G. Radney. 2020. Comparison of three essential sub-micrometer aerosol measurements: Mass, size and shape. *Aerosol Sci. Technol.*:1-13. doi: 10.1080/02786826.2020.1763248.