Review of:

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

Yao Song, Xiangyu Pei, Huichao Liu, Jiajia Zhou, Zhibin Wang

General comments:

The manuscript by Song et al. examines the problem of multiple charging when using two tandem systems: a differential mobility analyzer (DMA) and centrifugal particle mass analyzer (CPMA) and a DMA and an aerodynamic aerosol classifier (AAC). These systems are collectively referred to as a DMA-CPMA and DMA-AAC, respectively. The manuscript analyzed the transfer function of these systems theoretically, under static conditions, using data from the literature and then analyzed the presence or absence of multiply charged particles when working with flame generated soot. They find that the ability of the DMA-CPMA and DMA-AAC to remove multiply charged particles depends upon the resolution of the classifiers, the morphology of the particles and the mobility diameter (D_m) under investigation; smaller Dm are more likely to be contaminated with multiply charged particles.

The manuscript has been greatly improved since the first review and can be accepted pending a minor revision.

Specific AMT review criteria:

1. Scientific significance: The manuscript represents a substantial contribution to understanding the limits of nanoparticle separation when using a tandem DMA-CPMA or DMA-AAC. Rating: good

2. The scientific approaches and applied methods are valid. While the discussion of the results has been improved since the initial submission, there are places where further discussion would improve manuscript clarity. Rating: good

3. The presentation of the results and conclusions has improved greatly since the initial submission, but there are still many places where improvements to clarity would be beneficial to the reader. Rating: good

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

Specific comments:

1. The authors put quite a few equations inline in the text. It may improve readability to have them offset like the remainder, especially when the values are used multiple times throughout.

2. The authors need to double check that they are defining each variable to represent a single quantity. For example, m is used as mass in general or as the mass axis in a distribution and as the mode mass of the particle distribution.

3. It is still not clear to me how the charge (when > 1) is being included in the calculated transfer functions (*e.g.*, paragraph starting on line 230 and Figure 2) since the authors are presenting everything as absolute mass instead of effective mass. Is the charge term somehow “baked into” the calculated transfer functions? Some clarification is warranted.

In the author response document, their response to technical comment 27 was “The particles are shown in figure 2 in actual dm and m, but when we calculate the resolution of DMA and CPMA, the mobility and effective mass are used. The resolution of CPMA can be calculated by Eq. R2, where m1 is the mass of singly charged particles which can be selected by the CPMA, i.e. effective mass.”

Including this would be sufficient.

4. In the reviewer response, the authors include the following paragraph: “In our study, we also use scanning mode of CPMA after DMA selection to determine the mode mass of the selected particles, then we use the tandem setup of DMA and CPMA both at fixed mode to select particle at fixed mobility and mode mass i.e. DMA and CPMA are used in a static configuration, no scanning for either instrument is used. In Figure 5a, DMA-CPMA is set to select singly charged particles with dm of 80 nm and m of 0.16 fg, while the doubly charged particles with dm of 119.3 nm and m of 0.32 fg will also be selected and the transfer function is presented as upper right region. Soot particles curve (red line) goes through the upper-right region which doubly charged particle can penetrate (dm of 113 nm~118 nm, m of 0.35 fg~0.39 fg). As a result, we conclude that multiple charging effect still exists when DMA-CPMA select soot particles with dm of 80 nm and m of 0.16 fg.”

This is one of the best and most concise paragraphs describing the DMA-CPMA data in the manuscript. I highly recommend integrating it into the body of the manuscript.

5. In the reviewer response, the authors provide a discussion on the ordering of the DMA-AAC and the impact on static measurements in response to my comment 5. (Starts at “This population has one physical size (dae) but the dm”). This is an excellent discussion and I strongly recommend integrating it into the manuscript.

Technical corrections:

Line 15: “specific size or mass.”

Size is a nebulous term with respect to mobility (D_m) or aerodynamic (D_{ae}) diameter. Maybe rewrite to “specific mobility diameter, aerodynamic diameter or mass, respectively.”?

Line 17: “demonstrate”

Calculate?

Line 18: “in static configurations.”

For flame generated soot particles.

Line 22: “resolutions”

How is resolution defined in this sense?

Line 24: “Otherwise, our results indicate…”

Repeated sentence from line 21.

Line 36: "size dependence"

size-dependent

Line 44: "particles must be precharged"

Unclear what this means since, depending upon particle size, most aerosols possess a net charge. Instead, I think the authors are referring to bringing the particles to a known charge distribution by passing through a charge neutralizer or similar?

Line 45: "mass-to-charge ratio,"

mass-to-charge ratio, respectively,

Line 55: "of DMA-APM"

of the DMA and APM

Line 64: "is that no charging process is needed"

Should this be "the charge state of the particles does not need to be known"?

Line 71: "(Johnson et al., 2018)."

This is not a peer-reviewed manuscript but rather a conference presentation.

Line 73: "APM(Yao"

Missing a space between APM and (Yao

Line 97: "elemental"

elementary

Line 98: "Zp*"

Z_p* needs to be defined. Also, the symbol used for Zp* on line 98 and in the table of symbols is different than that used in Eq. 2.

Line 102: "DMA, respectively."

DMA electrodes, respectively.

Line 109: "$d_{m\ n,max}$ and $d_{m\ n,min}$"

Is there supposed to be a space or a , between m and n?

Line 111: "Zp"

Z should be italic.

Line 113: "The construction of the CPMA is similar to the APM,"

How so? No discussion on the construction of the APM has been provided.

Line 123: α and β, in this usage, should be explicitly defined to avoid confusion with other quantities (e.g., αabs, βDMA, etc.)

Line 136: "Assuming a plug flow,"

Delete "a".

Line 139 and 140: Formatting on functions should be consistent throughout. Min, max, exp should all be formatted similarly. Also, min (and max, preferably) should be explicitly defined after use as the minimum (and maximum) of the quantities in the brackets since min is also the abbreviation for minutes.

Line 161: "τ"

τ should be italic.

Line 162: "denote"

denoted

Line 165 and 166: "SCPM" and "SLPM" are non-standard units and should be explicitly defined; *e.g.* SLPM (standard L per min, flow in L min^{-1} converted from ambient to T = and P =).

Line 171 and 173: "Qsh/Qa = 10"

the β_{DMA} and β_{AAC} formulation should be used for consistency throughout.

Line 171: "monodisperse"

This is not technically correct as written and the authors should remind the reader that the stream is monodisperse in Z_p.

Line 174: "The particle mass (m) and aerodynamic diameter (d_{ae}) were determined by the scanning mode"

This is not exactly true as written. The distributions of particle number density as a function of particle mass and aerodynamic were measured and mode particle mass was then determined from a fit of that distribution.

Line 175: "CPMA and AAC, while"

CPMA and AAC, respectively, while

Line 178: "SMPS"

Needs to be defined.

Line 179: "soot particles did not change during the whole experiment."

It is unclear to what change the authors are referring.

Line 180: "The m and d_{ae} distributions were fitted to log- normal distributions; thus, the modes m and d_{ae} for the mobility-selected particles were determined"

According to this sentence, m and d_{ae} represent multiple quantities, both the axis and the modal value. Separate variables should be used for each quantity to avoid confusion.

Line 181: “equation of log-normal distribution used in this study is expressed as”

This equation only applies to $N(d_p)$ and not to the m or d_{ae} fits that the authors are referring.

Line 183: “where σ is the geometric standard deviation and μ is the geometric mean”

Did the distributions of m and Dae have the same σ and μ? That is what is implied.

Line 194: “electrical diameter”

electrical mobility diameter

Line 196: “ρeff”

The mathematical relationship for ρ_{eff} has not been defined anywhere.

Line 202: “where Φ and Ω are the transfer functions of each classification system expressed by subscripts.”

Should be “where Φ and Ω are the transfer functions of the combined and individual classification systems expressed by subscripts, respectively.”

Line 206: “dm = 80 nm”

Why is the dm, m and dae different in this sentence than in the previous?

Line 208: “included 600 points, respectively”

600 points each?

Line 210: “>m2,max, from” How far < or > the respective values were investigated?

missing “and”

Line 213: “The DMA-CPMA transfer function is calculated in log(dm)-log(m) space, as shown in Fig. 2.”

“DMA-CPMA transfer function ($\Phi_{DMA\text{-}CPMA}$)”. What is the transfer function calculated for?

Line 215 and 216: “*nm*”

nm should not be italicized when used as a unit.

Line 217: “and smaller than 3 for aspherical particles”

Dfm can be larger than 3 for particles that are non-spherical at small D_m and approach spherical as D_m increases.

Line 219: “Under this specific operation condition”

What specific operating condition? Please explain.

Line 220: “spherical particle population (black line)”

Is this a “theoretical” spherical particle population? What would be the ρ_{eff} of these particles?

Line 220: “classification region”

What is the classification region? Please elaborate.

Line 231: "than the slope of a line connecting (dm, m) = (dm2,min, m2,max)(dm1,m1) (as PP0 shown in Fig. 2)"

PP0 is not clearly shown in Fig. 2 and this was **the** source of my confusion. From the figure, it appears that PP0 is drawn as the Dfm = 2.28 line. So, while the Dfm = 3 discussion seems reasonable, the Dfm = 2.28 does not. In contrast, for Figure 4 the PP_0 line is clearly visible making the discussion much easier to understand. My recommendation is to either switch the ordering of the DMA-CPMA and DMA-AAC sections or to add an additional plot to Figure 2 at a larger dm1 where PP0 is clearly visible.

Line 236: ". Accordingly, the ideal condition…"

Under static operation at this set point

Line 240: "Eq. (26) gives instructions in actual operation"

It is unclear how Eq. 26 gives instructions.

Line 244: "and the slope of PP0 derived from the actual condition"

This is unclear as written. How is the slope of PP_0 derived from actual conditions? Weren't the transfer functions from which PP_0 is determined theoretically calculated?

Line 246: "According to the theoretical calculation described in Kuwata (2015), the slope of PP0 of 3.55 was derived when the DMA-APM selects the same…"

Kuwata did not calculate a PP_0, so it is unclear where this value is coming from. What was the slope of PP_0 for the DMA-CPMA for reference? Should the value of 3.55 be 2.55? If not, are the authors claiming that the DMA-APM would be unable to separate spherical particles (Dfm = 3) under these theoretical conditions? I completely agree that the DMA-APM is more susceptible to multiple charging, but this comparison to the APM needs to be clarified or removed.

Line 251: "Rm = 8"

What is Rm? This is the first instance of it in the manuscript.

Line 254: "the slope"

the critical slope

Line 251: "contour lines in Fig. 3"

How were these contours calculated? From Eq. 26? If so, how was dm2 determined?

Line 259: "mobility diameters larger than 200 nm, while it fails to eliminate …"

As shown by the circles and squares in Figure 3.

Line 263: "to charge aerosol particles"

To Boltzmann distribution or a known charge state?

Line 264: "AAC cannot constrain the properties of aspherical particles as monodisperse as DMA or CPMA classification"
Unclear as written.

Line 267: "selecting the same representative particles"

The same as what? Please give values.

Line 268: "aspherical particles can be expressed as follows"

The log(Dae) is expressed on the next line. Not aspherical particles.

Line 273: "the same particles"

Please give values as a reminder.

Line 276: "are in parallel for the DMA-AAC"

Unclear as written. I think the authors are referring to the fact that transfer function will have the same D_{ae} and different D_m?

Line 277: "the example setups"

What example setups?

Line 290: "which is the case for most atmospheric aerosol particles."

What is the case? This D_{fm} is smaller than for most aerosols.

Line 292: "is required"

"may be required"

Line 295: "When increasing β_{AAC} to 0.3"

Increasing is a misnomer here since an increase in β_{AAC} is a decrease in resolution. Please remind the reader of this distinction.

Line 302: "the corresponding dae and m were determined using the AAC and CPMA scan modes"

This isn't exactly true as written. The distributions of number density as a function of D_{ae} and m were determined by the scans. These distributions were then fit to a log-normal to determine the modal values and from these values the ρ_{eff} were determined.

Line 304: "measured spectral density"

Measured distributions?

Line 305: "The results are summarized in Table 2."

How were the uncertainties in Table 2 determined? 1σ standard deviation of multiple measurements? Or something else? Please describe.

Line 306: kf has units of mass.

Line 308: "two methods"

Which two methods?

Line 308: "the deviation"

What deviation?

Line 316: "the corresponding transfer function"

DMA-CPMA transfer function?

Line 317: "The particle population"

Shown by the red D_{fm} = 2.28 line?

Line 320: "particles number aerodynamic size distribution"

Should be "particles number density aerodynamic size distribution"

Line 324: "The mean dae values"

For particles with dm = 80 nm?

Line 327: "In contrast, …"

In contrast to what?

Line 332: "PP0"

Subscript 0.

Line 363: "26.7±3.0%"

What is the unit on 26.7? Is it %? If so, should be written as (26.7±3.0) % or 26.7 % ± 3.0 % to avoid the confusion of 3.0 % being a relative value and the absolute being 26.7 ± 0.8. Other values in this paragraph need to be similarly corrected.

Line 387: "Under the same setups"

Same as what?

Table A1: Thank you for including this table. But, can you please sort values alphabetically to assist the reader in locating values and include the corresponding units where appropriate.

Table 2: "*M* (fg)"

Is this the modal mass from the log-normal fit? This is the wrong symbol.

Table 3: fN and fabs are not mathematically defined in the body of the manuscript. Can the authors provide the calculated MAC for each size and the overall? And should "MAC overestimation" have a units associated with it?

Figure 2 caption: Please note that this plot is in log-log space as a reminder for the reader. It is not clear from just looking at the axes.

Line 533: “DMA and CPMA.”

DMA and CPMA, respectively.

Figure 3: What are the minimum values on the dm and m axes?

Line 537: “The contour lines denote the slope”

Critical slope?