Response to James Radney

We thank the reviewer for the constructive suggestions and comments concerning our manuscript entitled "Characterization of tandem aerosol classifiers for selecting particles: implication for eliminating multiple charging effect" (ID: amt-2021-436). Those comments are valuable and very helpful for improving our paper, as well as the important guiding significance to our studies. Below, we provide a point-by-point response to individual comment (Reviewer comments in italics, responses in plain font; page numbers refer to the AMTD version; Tables used in the response are labeled as Table S1, Table S2,…, figures used in the response are labeled as Fig. R1, Fig. R2,…)

General Comments and suggestions:

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

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

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

Responses and Revisions:

Thank you for the advice. In summary, we are very sorry for the misunderstanding due to the poor expression. The revisions could be found in individual comment in details.

For our main contribution, Equations 26 and 28 demonstrate the relationship between D_{fm} and the instrument resolutions when using DMA-CPMA to eliminate multiple charging effect. We have included a brief description of these equations in the abstract: "We propose an equation that constrains the resolutions of DMAs and CPMAs to eliminate the multiple charging effect when selecting particles with a certain mass–mobility relationship using the DMA-CPMA system. The equation for the DMA-AAC system is also derived"

For transfer functions of tandem system of DMA-CPMA and DAM-AAC, we think our calculation is credible, while the expression leads to the misunderstanding. We calculated the transfer functions of DMA-CPMA and DMA-AAC both in a static configuration. The transfer function of DMA-CPMA was derived by multiplying the transfer functions of DMA and CPMA. The transfer function of DMA-AAC was derived by multiplying the transfer functions of DMA and AAC. We have reorganized our language in the revised manuscript. The details could be found in Comments 4.

Our English writing has been promoted by native speaker.

1. ***Comments and suggestions:***

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).*

Responses and Revisions:

We have revised Fig.3 to contour plot. It is more straightforward to compare the reported D_{fm} and critical slope PP_0 values. The selected particles with multiple charging effect are resented as squares. The colors are used to distinguish literature data. Moreover, we have included the black and white version in the supplement (Fig. S4). According to the Color BLIndness Simulator, this figure should be readable for all readers.

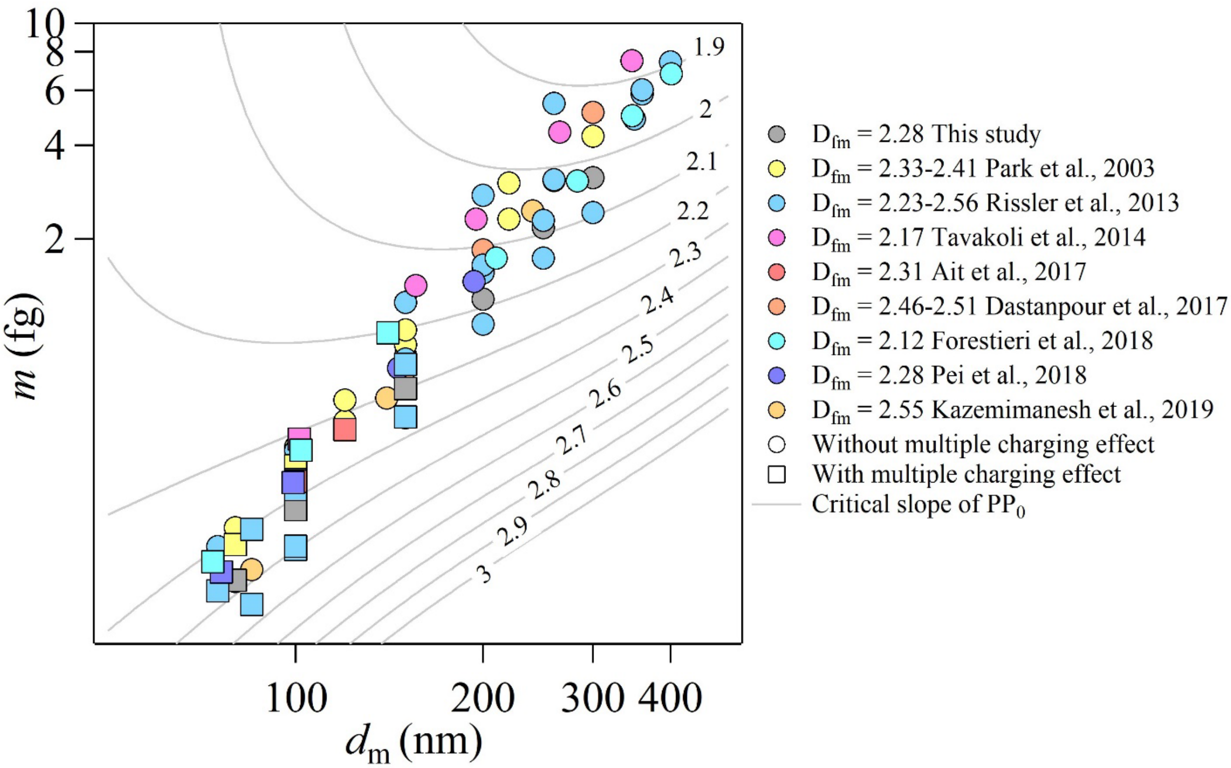


Figure 3: Variations of the slope of PP_0 as a function of classified d_m and m. The following parameter set was employed for the calculations: β_{DMA} = 0.1, Q_{CPMA}=0.3 L min^{-1}, R_m = 8. The contour lines denotes the slope of PP_0 with values labeled on them. The data points are soot particles measured in literatures (Park et al., 2003; Rissler et al., 2013; Tavakoli et al., 2014; Ait Ali Yahia et al., 2017; Dastanpour et al., 2017; Forestieri et al., 2018; Pei et al., 2018; Kazemimanesh et al., 2019) and generated in this study (See details in section 3.2).The D_{fm} of these data points are listed in the legend. The data points become square when the D_{fm} is smaller than the critical slope of PP0 in the background, i.e. the potential multiple charging effect may exist.

In the revised manuscript, the figures (Fig. 2, Fig. 4, Fig. 5a and Fig 6a) with rainbow color scale are kept to compare with the previous study representing the DMA-APM transfer function (Kuwata et al., 2015), we hence kept them likewise.

2. *Comments and suggestions:*

Uncertainties are missing on reported values throughout.

Responses and Revisions:

We have added the uncertainties in Table 2 and Table 3.

3. *Comments and suggestions:*

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?)

Responses and Revisions:

This is a very good suggestion. The transfer function of AAC-DMA is calculated with the transfer functions of two classifiers (Tavakoli and Olfert, 2014):

$$\Phi_{\mathrm{AAC-DMA}} = \Omega_{DMA}\Omega_{AAC} \ , \qquad \text{R1}$$

we think the transfer function of DMA-AAC in a static configuration is independent on their ordering. An example can be found in comment 5.

As for DMA-CPMA, the resolution of CPMA is calculated assuming that all the particles have exactly the same mobility:

$$R_m = \frac{m_1}{m_{1,max}-m_1} = \frac{2\pi B_{1,max} L_{CPMA} r_c^2 \omega^2 m_1}{Q_{CPMA}} \qquad \text{R2}$$

where m_1 and $m_{1,\mathrm{max}}$ are the nominal mass and the maximum mass that can be selected by CPMA, respectively. $B_{1,\mathrm{max}}$ is the mobility of particles with mass of $m_{1,\mathrm{max}}$. We assume that $B_{1,\mathrm{max}}$ is equal to the mobility of nominal particles. This assumption is valid when the particles are mobility selected before fed into the CPMA, so the ordering of DMA-CPMA can't be changed.

We didn't include the transfer function of AAC-CPMA or CPMA-AAC in our study. We attempted to calculate the transfer function of AAC-CPMA, but we found it was difficult since particle mobility should be provided first to connect the calculations of AAC and CPMA. The relationship between τ selected by AAC and m selected by CPMA is shown as follows (Yao et al., 2020):

$$\tau = m_{sp}B = \chi m B, \qquad \text{R3}$$

in which m_{sp} is mass of spherical particles. CPMA determines real mass of particles without any assumption of χ. Mobility cannot be derived from Eq. R3 since χ is unknown.

4. *Comments and suggestions:*

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 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.

Responses and Revisions:

Thank you for the comment. We thought that we did not clearly clarify our instruments configuration. Usually, the tandem setup of DMA-CPMA is that DMA firstly selects a fixed mobility, while CPMA is used in scanning mode to derive the corresponding mass . For example, in Figure 2 in Radney et al. (2013), they fixed the DMA to select particles at d_{m} of 200 nm and particles bearing higher-order charges were also selected because of multiple charging effect. The mobility-selected particles included particles bearing 1, 2 and 3 charges with d_{m} of 200 nm, 321 nm and 434 nm, respectively. The corresponding m of 7.4 fg, 30.7 fg and 75.8 fg can be calculated using d_{m} and effective density, respectively. The scanning mode of APM was used downstream to resolve multiple charging effect of these mobility-selected particles. The APM selects particles according to their mass-to-charge ratio. As a result, the effective masses measured by APM were 7.4 fg, 15.3 fg, 25.3 fg, respectively, which is shown as grey circles in Figure 2 (Radney et al., 2013)

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 d_m of 80 nm and m of 0.16 fg, while the doubly charged particles with d_m 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 (d_m 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 d_m of 80 nm and m of 0.16 fg.

5. *Comments and suggestions:*

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.

AND

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.

Responses and Revisions:

Yes, we agree that AAC selects only one population. This population has one physical size (d_{ae}) but the d_m range of this population is wide since soot particles have different densities. Kazemimanesh et al. (2022) demonstrated that AAC does not constrain the properties of nonspherical particles as monodisperse as DMA or CPMA classification. The AAC selects relaxation time, not directly aerodynamic diameter. The relationship between the relaxation time and mobility diameter can be expressed as follows,

$$\tau = \frac{\rho_{eff} d_m^2 Cc(d_m)}{18\mu}, \quad \text{R4}$$

which indicates the AAC selects monodispersed particles when particles have the same effective density. In our study, the effective density of soot particles decreases with increasing d_m. Particles with different d_m but the same relaxation time will be selected.

We think the transfer functions of DMA-AAC or AAC-DMA are identical regardless of the order of DMA and AAC. For example, we use AAC-DMA to select particles with d_{ae} of 68 nm and d_m of 100 nm. In figure 4a in this study, the transfer function of AAC is the region between the horizontal lines of $d_{ae,max}$ (75 nm) and $d_{ae,min}$ (63 nm). The soot particles population (red line) goes through this region will be selected by AAC. The mobility diameter distribution of these relaxation time selected particles is around 80 nm to 120 nm. Then the DMA is fixed to select particles with d_m of 100 nm, the particles with double charges and the same mobility (d_m of 150 nm) have been excluded by AAC. As a result, AAC-DMA select monodispersed particles with d_{ae} of 68.3 nm and d_m of 100 nm. In Fig. 4b, the resolution of AAC is lower and transfer function of AAC is broader than that in Fig. 4a. The soot particles population (red line) goes through the transfer function region between the horizontal lines at d_{ae} of $d_{ae,max}$ (50 nm) and $d_{ae,min}$ (86 nm). The mobility diameter distribution of these relaxation time selected particles is very wide from less than 80 nm to about 158 nm. Then these relaxation time selected particles were charged and selected by DMA at d_m of 100 nm, singly charged particles with d_m of 95 nm~106 nm and doubly charged particles with d_m of 142 nm~158 nm will be selected.

If we use the DMA-AAC, the particles are selected by DMA first. For example, in Figure 4b, the transfer function of DMA is shown as two vertical regions which particles with single and double charges can penetrate. The soot particles (red line) goes through it and two populations of soot particles with mode d_m of 100 nm and 150 nm will be selected. The corresponding d_{ae} distributions of these singly and doubly charged particles are

66 nm~70 nm and 81 nm~87 nm. These mobility-selected particles are selected at d_{ae} of 68.3 nm by AAC and the transfer function of AAC shows that particles with d_{ae} of 50 nm~86 nm can penetrate. As a result, singly charged particles with d_{ae} of 66 nm ~70 nm and doubly charged particles with d_{ae} of 81 nm ~86 nm can be selected.

As a summary, the transfer functions of DMA-AAC and AAC-DMA in a static configuration are the same no matter the ordering of DMA and AAC.

6. *Comments and suggestions:*

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 containedwithin the primary peak and what was identified as q = 3 is 4 or higher?

Responses and Revisions:

Thank you for raising this question. First of all, the size distribution of aerosols is often found to be a log-normal distribution. Then, the d_{ae} distribution is asymmetrical on a linear axis and we found that d_{ae} distribution was fitted well with log-normal distribution.

a) We used DMA-CPMA in a static configuration to select soot particles with specific d_m and m. Particles with higher order charges can also be selected and the values of d_m and m can be calculated. According to Eq. (1) and Eq. (16), the d_{ae} of particles with different charges can be calculated by:

$$\frac{\pi}{6}\rho_0 Cc(d_{ae})d_{ae}^2 = \frac{Cc(d_m)}{d_m}k_f[d_m(nm)]^{D_{fm}} \cdot 10^{-18}, \quad \text{R5}$$

The corresponding d_{ae} of the selected particles with different charges can be calculated with selected d_m and m according to Eq. R5. The d_{ae} for different sizes soot particles with single, double and triple charges are shown in Table R1. The d_{ae} distribution for different sizes particles was fit well with single log-normal distribution and the values of mode d_{ae} were determined, which denoted as d_{ae_sd}. The deviations between the calculated d_{ae} and fitted d_{ae} for particles with d_m of 150 nm, 200 nm and 250 nm are within 0.31% while the d_{ae_sd} for particles with 80 nm and 100 nm are much larger than the calculated d_{ae}. The deviations for 80 nm and 100 nm particles are 7.38% and 6.85%, respectively. We think that the peaks shift right because of multiple charged particles, so we use the summation of multiple log-normal distributions. Although the deviation for 150 nm particles is 0.29%, the summation of multiple log-normal distributions is used because the points with d_{ae} larger than 90 nm can't be fitted well with single log-normal distribution.

Table R1. The calculated d_{ae} for mobility and mass selected particles with single, double or triple charges and the single log-normal fitted d_{ae_sd}. The deviations between the calculated d_{ae_1q} of particles with single charge and the fitted d_{ae_sd} for different size soot particles.

d_m(nm)	m (fg)	Calculated d_{ae} (nm)	d_{ae_sd} (nm)	Deviation

		d_{ae_1q}	d_{ae_2q}	d_{ae_3q}	d_{ae_4q}		
80	0.16	51.5	62	70.7	78.5	55.3	7.38%
100	0.27	56.9	70.4	81.8	92.3	60.8	6.85%
150	0.66	70.1	91.8	111.2	129.4	70.3	0.29%
200	1.28	82.9	113.9	141.8	167.7	83.1	0.24%
250	2.17	95.6	157.8	172.2	205.5	95.9	0.31%

b) The DMA-CPMA selected particles with specific d_m and m, and the corresponding d_{ae} of selected particles with different charges can be calculated with Eq. R5. The calculated values are set as the start points of central values of d_{ae}. For example, the central values of d_{ae} used in 80 nm particles fitting are 51.5 nm, 62 nm and 70.7 nm.

c) The constraint of peak width of particles with single, double and triple charges were the same. The start point of the coefficient σ (two times of peak width) was e^0.5, and the lower and upper bounds were e^0 and e^1, respectively.

d) The fitting coefficients with 95% confidence bounds are summarized in Table R2.

Table R2. The fitting coefficients with 95% confidence bounds for different mobility and mass selected particles with single, double and triple charges.

	N (95% confidence bounds)	μ (95% confidence bounds)	σ (95% confidence bounds)
80	1718.8 (1476.9, 1961.9)	53.9 (fixed at bound)	1.07 (1.05, 1.08)
	637.1 (337.7, 936.5)	60.65 (fixed at bound)	1.06 (1.02, 1.10)
	25.1 (906.7, 957.0)	70.9 (fixed at bound)	1.02 (0.07, 15.30)
100	5985.0 (5412.2, 6557.9)	59.5 (fixed at bound)	1.09 (1.08, 1.10)
	1279.7 (746.7, 1812.6)	68.6 (fixed at bound)	1.08 (1.04, 1.12)
150	8361.9 (7997.1, 8726.7)	69.9 (69.5, 70.4)	1.11 (fixed at bound)
	329.3 (-31.4, 690.0)	93.5 (78.6, 111.3)	1.11 (fixed at bound)
200	8086.6 (7864.7, 8308.4)	83.1 (82.9, 83.3)	1.099 (1.096, 1.102)
250	9835.5 (9603.9, 10067.1)	95.9 (95.7, 96.1)	1.095 (1.093, 1.098)

e) We have included the uncertainties in particles number concentration. We scanned the d_{ae} distributions at least three times for each size particles. The average of the number densities was used and error bar is shown in the figures.

f) The d_{ae} of particles with higher order of charges (q>3) can also be calculated using Eq. R5. The values are very large (e.g. 78.5 nm for 80 nm particle with 4 charges) and close to upper limit of scanning range (35 nm~80 nm for 80 nm particles). Nonetheless, we think that particles with higher order of charges do not exist since the number density already tends to zero at the end of the scanning.

Technical corrections:

7. *Comments and suggestions:*

Line 18: *"the potential multiple charging effect" Elaborate.*

Responses and Revisions:

Thank you for the comment. The potential multiple charging effects of Tandem System, such as DMA-CPMA and DMA-AAC et al., have been discussed in the following text from Line 21 to Line 28: "Our results show that the ability to remove multiply charged particles mainly depends on particles morphology and instruments resolutions of DMA-CPMA system. Using measurements from soot experiments and literature data, a general trend in the appearance of multiple charging effect with decreasing size when selecting aspherical particles was observed. Otherwise, our results indicated that the ability of DMA-AAC in a static configuration to eliminate particles with multiple charges is mainly related to the resolutions of classifiers. In most cases, DMA-AAC can eliminate multiple charging effect regardless of the particle morphology in a static configuration, but multiply charged particles will be selected when decreasing resolution of DMA or AAC".

8. *Comments and suggestions:*

Line 19: *"remove"*

Resolve?

Responses and Revisions:

Thank you for your advice. We think "remove" should be used since we discuss DMA-CPMA in a static configuration.

9. *Comments and suggestions:*

Line 20: *"instruments setups of DMA-CPMA system" What is meant by instrument setups? Elaborate.*

Responses and Revisions:

We have changed this in the revised manuscript:

"Our results show that the ability to remove multiply charged particles mainly depends on particles morphology and instruments resolutions of DMA and CPMA".

10. *Comments and suggestions:*

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 usedin a static configuration. In a scanning mode, the contributions would be resolvable.

Responses and Revisions:

Sorry we didn't make it clear that we used DMA-AAC in a static configuration. We have changed it in the revised manuscript:

"In most cases, DMA-AAC in a static configuration can eliminate multiple charging effect regardless of the particle morphology".

11. *Comments and suggestions:*

Line 24: *"while particles with multiple charges can be selected when decreasing resolutions of DMA and AAC"*
Confusing as written.

Responses and Revisions:
We have changed this in the revised manuscript:
"In most cases, DMA-AAC in a static configuration can eliminate multiple charging effect regardless of the particle morphology, but multiply charged particles will be selected when decreasing resolutions of DMA or AAC".

12. ***Comments and suggestions:***
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"?

Responses and Revisions:
We have changed this in the revised manuscript:
"We propose that the potential influence of the multiple charging effect should be considered when using DMA-CPMA or DMA-AAC systems in estimating size- and mass-resolved optical properties in field and lab experiments".

13. ***Comments and suggestions:***

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?

Responses and Revisions:
Park et al. (2008) reviewed the tandem techniques of DMA and other measurements. We have included it and other references of application of DMA in laboratory and field studies therein.

14. ***Comments and suggestions:***

Line 38: *"particles are required to be pre-charged"In what sense do they need to be pre-charged?*

Responses and Revisions:
We have changed this in the revised manuscript:
"However, particles must be precharged when classified by a DMA or PMA because DMA and PMA classify particles based on electrical mobility and mass-to-charge ratio".

15. ***Comments and suggestions:***

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.

Responses and Revisions:
We have changed this in the revised manuscript:

"This conclusion implies that multiply charged particles cannot be effectively excluded for aspherical particles, especially for soot particles"

16. ***Comments and suggestions:***

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.

Responses and Revisions:

We have added the sentence: "The derived ρ_{eff} and χ depend upon the combination of instruments used, while the nonphysical values of χ and ρ_{eff} for aspherical particles can be determined by the AAC-APM(Yao et al., 2020) and AAC-CPMA (Kazemimanesh et al., 2022)".

17. ***Comments and suggestions:***

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 Zp is nonlinear.

Responses and Revisions:

We have added the sentence: "It translates to asymmetric in d_m since the relationship between d_m and Z_p is nonlinear".

18. ***Comments and suggestions:***

Line 106: *"is much simpler and more robust"*
Elaborate.

Responses and Revisions:

We have changed this in the revised manuscript:
"They considered the Taylor series expansion about the center of the gap (r_c=(r_{2_CPMA}+r_{1_CPMA})/2) instead of the equilibrium radius to avoid problems with the scenario in which the equilibrium radius does not exist. This method is much simpler and more robust"

19. ***Comments and suggestions:***

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.

Responses and Revisions:

Sorry for the misunderstanding. In this study, we only use one CPC. We have revised the manuscript and Figure 1. In the previous sentence, we have revised it to "For the soot characterization, the monodisperse aerosol flow was switched between two parallel lines and fed into the CPMA (Cambustion Ltd., UK) and AAC (Cambustion, Ltd., UK, $Q_{sh}/Q_a = 10$); meanwhile, the condensation particle counter (CPC, Model 3756, TSI, Inc., USA, 0.3 L·min^{-1}) was switched between the CPMA and AAC. The particle mass (m) and aerodynamic diameter (d_{ae}) were determined by the scanning mode of the CPMA and AAC, while the CPC recorded their corresponding number concentrations at each setpoint. For each d_m, the m and d_{ae} distributions were measured three times. Between measurements of each d_m, the CPC was used behind the DMA, and the number size distribution of the generated soot particles was measured by SMPS to ensure that the generated soot particles did not change during the whole experiment".

20. ***Comments and suggestions:***

Line 147: *"concentration"*
Number density of particles. Concentration is assumed to have units of mole per unit volume.

Responses and Revisions:
We have revised it to "number concentration".

21. ***Comments and suggestions:***

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 thesame 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.

Responses and Revisions:
The equation $N(d_p) = \frac{N_0}{\sqrt{2\pi}\ln\sigma}\exp\left(\frac{-(\ln(d_p)-\ln(\mu))^2}{2(\ln\sigma)^2}\right)$has been added. In Fig. 5 and S2, the distributions are plotted on a log axis, not a linear axis. This distribution is asymmetric on linear axis, hence we use log-normal distribution.

22. ***Comments and suggestions:***

Line 151: *"density of PSL"Please enumerate.*

Responses and Revisions:
We have added the density value of 1050 kg m^{-3} for PSL.

23. ***Comments and suggestions:***

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.

Responses and Revisions:
We have changed this in the revised manuscript:
"To quantify the multiple charging effect of particles selected by the DMA-CPMA system, the soot particles were initially selected by the DMA-CPMA at different d_m and the corresponding m. Then, the d_{ae} distribution

of mobility and mass selected particles was obtained by stepping the AAC rotation speed of the cylinder with simultaneous measurement of the particle concentration at the AAC outlet using a CPC".

24. ***Comments and suggestions:***

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.

Responses and Revisions:

This is a very useful suggestion. We have included the details in the revised manuscript: . "We explain the transfer functions of DMA-CPMA and DMA-AAC utilizing the literature data of soot particles (Pei et al., 2018). The d_m and m of the representative particles are 100 nm and 0.33 fg, respectively, and the corresponding d_{ae} is 68.3 nm according to Eq. (19). In the calculation, the following parameter set was employed for the calculations: d_m = 80 nm, Q_{CDMA} = 0.3 L min^{-1}, β_{DMA} = 0.1, m = 0.16 fg, Q_{CPMA}=0.3 L min^{-1}, R_m = 8, d_{ae} = 68.3 nm, Q_{AAC} = 0.3 L min^{-1}, β_{AAC} = 0.1".
The maximum and minimum values of d_m for particles with n charges can be derived and denote as $d_{m\ n,max}$ and $d_{m\ n,min}$, respectively. The maximum and minimum m of particles bearing single and double charges which are derived from Eq. 15 denote as $m_{1,\ max}$, $m_{1,min}$, $m_{2,max}$ and $m_{2,min}$. The maximum and minimum values of d_{ae} that can be selected by AAC denote as $d_{ae,max}$ and $d_{ae,min}$. The transfer functions of DMA-CPMA and DMA-AAC were solved iteratively using logarithmically spaced d_m, m and d_{ae}, which included 600 points, respectively. the ranges of d_m, m and d_{ae} used in the calculations were from < $d_{m1,min}$ to > $d_{m2,max}$, from < $m_{1,min}$ to > $m_{2,max}$, from < $d_{ae,min}$ to > $d_{ae,max}$, respectively.

25. ***Comments and suggestions:***

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$^{-2.28}$.

Responses and Revisions:

Equation 22 has been changed to $m = k_f \frac{(d)_m}{nm}^{Dfm}$, then k_f will have the unit of "g". Equation 23 has been changed to $\log(m) = D_{\mathrm{fm}} \log\left(\frac{d_m}{nm}\right) + \log(k_f)$.

26. ***Comments and suggestions:***

Line 184: *"In the exemplary case, the derived Dfm of premixed flame generated soot particles was 2.28,"*
What study does this refer to? Reference?

Responses and Revisions:

The reference (Pei et al., 2018) has been added.

27. ***Comments and suggestions:***

Line 189: *"The DMA-CPMA system can eliminate the multiply charged particles only if the Dfm of particlesis larger than the slope of a line connecting line connecting (dm, m) = (dm2,min, m2,max)(dm1,,m1) (as PP0 shown in Fig. 2)."*
The point (dm2, m2) appears to correspond to the actual mass and d_m *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.*

Responses and Revisions:
Thank you for your comment. We use the DMA-CPMA in a static configuration. When particles are selected by the DMA-CPMA with certain d_m and m, the corresponding doubly charged particles of d_{m2} and m_2 would also be selected.
The particles are shown in figure 2 in actual d_m 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 m_1 is the mass of singly charged particles which can be selected by the CPMA, i.e. effective mass.

28. ***Comments and suggestions:***

Line 190: *"line connecting (dm, m) = (dm2,min, m2,max)(dm1,,m1) (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.

Responses and Revisions:
We have removed the extra "," and the PP_0 has been marked out in Fig. 2.

29. ***Comments and suggestions:***

Line 201: *"are necessary to reduce the potential of multiply charged particles" By increasing the resolution of the measurement?*

Responses and Revisions:
We have changed this in the revised manuscript:
"When selecting particles of certain d_m and m, by decreasing Q_{CPMA}, or increasing ω and β_{DMA} i.e. by increasing the resolution of the measurement, the potential of multiply charged particles is reduced".

30. ***Comments and suggestions:***

Line 205: *"PP0 of 3.55 was derived when DMA-APM selects the same example soot particles" Compared to what?*

Responses and Revisions:
We have changed this in the revised manuscript:
"the slope of PP_0 of 3.55 was derived when the DMA-APM selects the same example soot particles from Pei et al. (2018) (d_m of 100 nm and m of 0.33 fg) with a D_{fm} of 2.28".

31. ***Comments and suggestions:***

Line 208: *"critical slope of PP0"*
Z-axis in Figure is labelled as "The slope of PP0".

Responses and Revisions:
We have revised it to "the critical slope of PP_0" in Fig. 3, consistent with the text.

32. ***Comments and suggestions:***

Line 210: *"when the Dfm 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.

Responses and Revisions:
The color scheme has been changed. The data points become square when D_{fm} is smaller than the critical slope of PP0 in the background, i.e., the potential multiple charging effect may exist.

33. ***Comments and suggestions:***

Line 213: *"Fig. 3, the dm, m and corresponding Dfm were taken from literature"*
In the caption, the authors mention that the shapes correspond to the individual studies. Please elaboratewhich is which here?

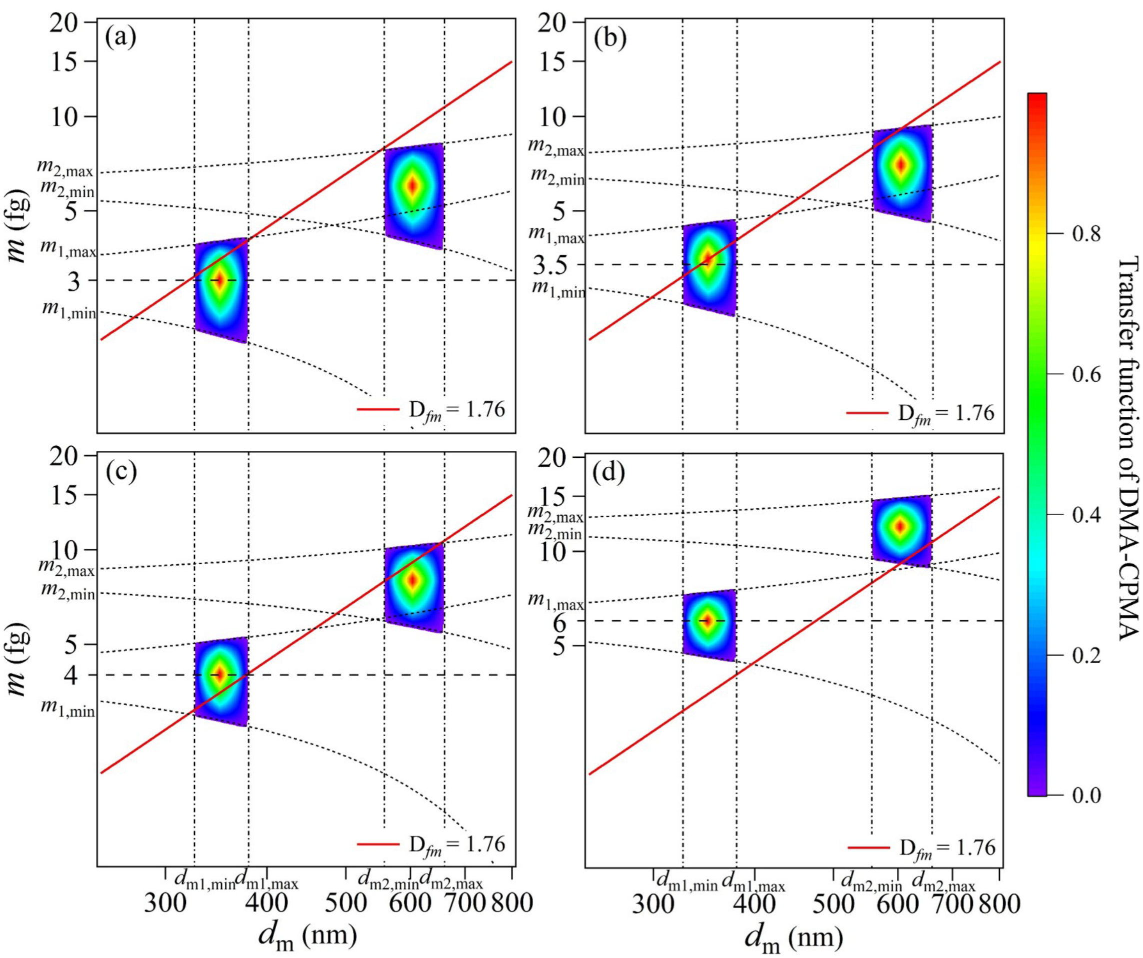
Responses and Revisions:
We have added more data to Fig. 3 and the references have been labeled in the figure.

34. ***Comments and suggestions:***

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 S1of (Radney et al. 2014). Also, there's a "," after the "." at the end of the sentence.

Responses and Revisions:
Thank you for your comment. More data has been added to Fig. 3. Generally, we think that the D_{fm} of soot particles is around 2.2~2.4. For these soot particles, we conclude that multiple charging effect can be avoided for DMA-CPMA to select particles with mobility diameter larger than 200 nm. We have revised the conclusion to "Generally, for soot particles with d_m of 2.2~2.4, multiple charging effect can be avoided for DMA-CPMA to select soot particles with mobility diameter larger than 200 nm".
We think the conclusion is consistent with the study of Radney et al (2014). The main difference between these two studies is the instruments configuration. In Figure S1 in Radney et al. (2014), DMA was used in static configuration and particles with d_m of 350 nm were selected. The APM was used in scanning mode and the mass distribution of mobility selected particles was determined. However, we used the static configuration of both DMA and CPMA in this study. Particles were sequentially selected by their mobility and mass. Usually we select a certain d_m and the corresponding mode mass. In the bottom of Figure S1 in Radney et al. (2014), three regions are determined and singly charged are selected at mass < 4 fg. In the top of Figure S1 in Radney

et al. (2014), the mode mass of particles with d_m of 350 nm is around 3.5 fg, as a result, no multiple charging exist when selecting these particles. Nonetheless, we demonstrate different result. The d_m of lacy soot particles in Radney et al. is 1.79, which is smaller than common soot particles mentioned above. Applying particles with d_m of 350 nm and m of 3.5 fg to Figure 3 in this study, the d_m of 1.79 is smaller than the critical PP0 of 2.02, i.e. multiple charging effect still exists after mobility and mass selection. We also apply other particles with smaller mass and the result shows that singly charged particles are selected at mass < 3 fg. In the bottom of Figure S1 (Radney et al., 2014), the slope of red line doesn't change obviously at mass of 3 fg maybe because the quantity of particles with higher charges is too small compared with particles with single charge since the peak of number concentration is at 3.5 fg. Moreover, we apply particles with d_m of 350 nm and mass of 6 fg to Figure 3 and the result shows that doubly charged particles are selected by DMA-CPMA, which is consistent with the green line in the bottom of Figure S1 (Radney et al., 2014).



RC-Figure 1: The transfer functions of soot particles with d_m of 350 nm and m of (a) 3 fg, (b) 3.5 fg, (c) 4 fg and (d) 6 fg from Figure S1 in Radney et al. (2014).

35. *Comments and suggestions:*

Line 215: *"diameter"*

To which diameter metric are you referring?

Responses and Revisions:

It has been changed to “mobility diameter”.

36. ***Comments and suggestions:***

Line 216: *“eliminate”*
Resolve?

Responses and Revisions:

Thank you for your comment. We think it should be ‘eliminate’. ‘Resolve’ is used when APM or CPMA is used in scanning mode and multiply charged particles can be identified from the mass distribution. We use the static configuration of DMA-CPMA and we are discussing if multiple charging can be avoided totally after mobility and mass selection. Hence, we think ‘eliminate’ is more proper.

37. ***Comments and suggestions:***

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.

Responses and Revisions:

We have changed this in the revised manuscript:
“Measuring solely with an AAC will avoid multiple charging. However, AAC cannot constrain the properties of aspherical particles as monodisperse as DMA or CPMA classification (Kazemimanesh et al., 2022). Multiple charging becomes a problem when the tandem measurement is made with a DMA or PMA”.

38. ***Comments and suggestions:***

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.

Responses and Revisions:

The label of the black dashed line drawn as PP_0 has been added. The figure has been replotted.

39. ***Comments and suggestions:***

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.*

Responses and Revisions:

Agree. We have added the sentence: “In addition, the maximum size ranges are also restricted by the sheath flow, so in some cases, a lower sheath flow rate is required to select larger particles.”.

40. ***Comments and suggestions:***

Line 255: *“Dfm was 2.28” What was the value of ρ_0?*

Responses and Revisions:

We have added the value of ρ_0 and this sentence has been revised to "The fitted value of D_{fm} and k_f were 2.28 and 7.49×10^{-6}, respectively**"**.

41. ***Comments and suggestions:***

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.

Responses and Revisions:
Thank you for your advice. We have changed this in the revised manuscript:
"which suggests that the contributions from the multiply charged particles can't be eliminated". We use "eliminate" here instead of "resolve" because we use the DMA-CPMA in a static configuration.

42. ***Comments and suggestions:***

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.

Responses and Revisions:
We have changed this in the revised manuscript:
"Since the classification of AAC is different from DMA and CPMA, the aerodynamic size distributions of mobility and mass selected particles were characterized."

43. ***Comments and suggestions:***

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.

Responses and Revisions:
Some small particles remaining in the AAC induced the peak at d_{ae} <40 nm. These residual particles were measured even if the sample flow is filtered. This reason has been added in the text.

44. ***Comments and suggestions:***

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.

Responses and Revisions:
Agree. The following text has been added in the text:
"Mie theory is probably not the "best" method to use here since soot particles are aspherical agglomerates. Realistically, however, the Mie comparison is only being used to prove a point about the impact of multiple charging. Therefore, in this instance, any errors in the calculated optical properties are somewhat inconsequential."

45. ***Comments and suggestions:***

Line 301: *"integral concentration for particles"*
Integrated number density of particles. Concentration is assumed to have units of mole per unit volume.

Responses and Revisions:

It has been changed to "number concentration".

46. ***Comments and suggestions:***

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

Responses and Revisions:

The derived shape factors have been added. The shape factors calculated from two methods are consistent. We think the values of χ are not necessary in this study, so we didn't include them in the text.

d_m(nm)	m(fg)	d_{ae}(nm)	$\rho_{DMA\text{-}AAC}$ (kg m^{-3})	$\rho_{DMA\text{-}CPMA}$ (kg m^{-3})	Deviation	$\chi_{DMA\text{-}AAC}$	$\chi_{DMA\text{-}CPMA}$	Deviation
80	0.16	48	551.2	596.8	7.65%	2.05	1.96	5.05%
100	0.27	55	488	515.7	5.38%	2.17	2.10	3.42%
150	0.66	67	359. 1	373.5	3.86%	2.49	2.43	2.34%
200	1.28	82	303.2	305.6	0.77%	2.62	2.60	0.44%
250	2.17	96	262.8	265.2	0.90%	2.71	2.70	0.50%

47. ***Comments and suggestions:***

*Figure **2:** "Example of DMA-CPMA transfer function"*
Transfer function for what? Soot?

Responses and Revisions:

The reference has been added in the sentence. It has been changed to "Example of DMA-CPMA transfer function of flame-generated soot particles (Pei et al., 2018)".

48. ***Comments and suggestions:***

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

Responses and Revisions:

Agree. The figure has been changed.

49. ***Comments and suggestions:***

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

Responses and Revisions:

The Z-axis label has been changed to "Transfer function of DMA-AAC".

50. ***Comments and suggestions:***

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

Responses and Revisions:

Agree. The figure has been changed.

51. *Comments and suggestions:*

Grammar mistakes.

Responses and Revisions:

Grammar mistakes have been corrected.

References:

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Tavakoli, F., and Olfert, J. S.: Determination of particle mass, effective density, mass–mobility exponent, and dynamic shape factor using an aerodynamic aerosol classifier and a differential mobility analyzer in tandem, J. Aerosol Sci., 75, 35-42, https://doi.org/10.1016/j.jaerosci.2014.04.010, 2014.

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Yao, Q., Asa-Awuku, A., Zangmeister, C. D., and Radney, J. G.: Comparison of three essential sub-micrometer aerosol measurements: Mass, size and shape, Aerosol Sci. Technol., 1-18, https://doi.org/10.1080/02786826.2020.1763248, 2020.