

## ***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 revised version submitted last time)

### **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.*

#### **Responses and Revisions:**

Thank you for the advice. We have tried to move the equations to Appendix, but it seems this change makes difficulty to follow the text. Therefore, we didn't make changes in the current version, but we can modify if the reviewer insists.

*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.*

#### **Responses and Revisions:**

Thank you for the advice. We have rechecked the variables in the manuscript, and revised the ambiguous ones.

*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  $d_m$  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.”

Including this would be sufficient.

#### **Responses and Revisions:**

Thank you for the advice. We have added it after the description of Fig. 2:

“The DMA-CPMA transfer function ( $\Phi_{\text{DMA-CPMA}}$ ) for particles mentioned above, i.e. particles with  $d_m$  of 100 nm and  $m$  of 0.33 fg, is calculated in  $\log(d_m)$ - $\log(m)$  space, as shown in Fig. 2. 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. (15), where  $m_1$  is the mass of singly charged particles which can be selected by the CPMA, i.e., effective mass”.

*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  $d_m$  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.”*

*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.*

### **Responses and Revisions:**

Thank you for the advice. We have included this description in the revised manuscript:

“When selecting particles with  $d_m$  of 80 nm and  $m$  of 0.16 fg, the corresponding DMA-CPMA transfer function is shown in Fig. 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. 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  $d_m$ ”). This is an excellent discussion and I strongly recommend integrating it into the manuscript.*

### **Responses and Revisions:**

The relevant discussion has been added after the discussion of multiple charging effect of DMA-AAC.

## **Technical corrections:**

### **6. Comments and suggestions:**

*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.”?*

### **Responses and Revisions:**

Thank you for the comment. We have revised it as “Differential mobility analyzer (DMA), centrifugal particle mass analyzer (CPMA) and aerodynamic aerosol classifier (AAC) are commonly used to select particles with a specific size mobility diameter, aerodynamic diameter or mass, respectively”.

### **7. Comments and suggestions:**

*Line 17: “demonstrate”*

*Calculate?*

### **Responses and Revisions:**

Revised.

### **8. Comments and suggestions:**

**Line 18:** *“in static configurations.” For flame generated soot particles.*

**Responses and Revisions:**

Revised.

**9. Comments and suggestions:**

**Line 22:** *“resolutions”*

*How is resolution defined in this sense?*

**Responses and Revisions:**

Resolutions refer to the electrical mobility resolution and mass resolution settings of DMA and CPMA, respectively. We have revised it to “our results show that the ability to remove multiply charged particles mainly depends on the particle morphology and resolution settings of the DMA and CPMA”.

**10. Comments and suggestions:**

**Line 24:** *“Otherwise, our results indicate...” Repeated sentence from line 21.*

**Responses and Revisions:**

Sorry we didn’t make it clear. The line 21 is about the ability to eliminate multiple charging effect for DMA-CPMA in a static configuration, while the Line 24 is about the DMA-AAC in a static configuration. We have revised the Line 21 and the Line 24 to “For DMA-CPMA in a static configuration, our results show that the ability to remove multiply charged particles mainly depends on the particle morphology and resolutions of the DMA and CPMA.” and “As for DMA-AAC in a static configuration, the ability to eliminate particles with multiple charges is mainly related to the resolutions of classifiers”, respectively.

**11. Comments and suggestions:**

**Line 36:** *“size dependence” size-dependent*

**Responses and Revisions:**

Revised.

**12. Comments and suggestions:**

**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?*

**Responses and Revisions:**

Thank you for the comment. We have revised it to “The charge distribution of particles must be known by passing through a neutralizer or similar when classified by DMA or PMA. However, particles with higher-order charges and identical apparent mobility or mass-to-charge ratio can be selected simultaneously, which are referred to as the multiple charging effect.”.

**13. Comments and suggestions:**

**Line 45:** *“mass-to-charge ratio,” mass-to-charge ratio, respectively,*

**Responses and Revisions:**

Revised.

**14. Comments and suggestions:**

**Line 55:** “of DMA-APM”  
of the DMA and APM

**Responses and Revisions:**

Revised.

**15. Comments and suggestions:**

**Line 64:** “is that no charging process is needed”  
Should this be “the charge state of the particles does not need to be known”?

**Responses and Revisions:**

Thank you for the comment. We have revised it to “The advantage of utilizing an AAC is that the charge state of the particles does not need to be known in particle classification compared with the aforementioned classifiers”.

**16. Comments and suggestions:**

**Line 71:** “(Johnson et al., 2018).”  
This is not a peer-reviewed manuscript but rather a conference presentation.

**Responses and Revisions:**

Thank you for the comment. We have deleted it and cited another reference.

**17. Comments and suggestions:**

**Line 73:** “APM(Yao”  
Missing a space between APM and (Yao

**Responses and Revisions:**

Revised.

**18. Comments and suggestions:**

**Line 97:** “elemental”  
elementary

**Responses and Revisions:**

Revised.

**19. Comments and suggestions:**

**Line 98:** “ $Z_p^*$ ”  
 $Z_p^*$  needs to be defined. Also, the symbol used for  $Z_p^*$  on line 98 and in the table of symbols is different than that used in Eq. 2.

**Responses and Revisions:**

Thank you for the comment. It has been revised to “the centroid mobility,  $Z_p^*$ , selected by the DMA is defined

as”. All the symbols have been changed to “ $Z_p^*$ ”.

## 20. Comments and suggestions:

**Line 102:** “DMA, respectively.” DMA electrodes, respectively.

### Responses and Revisions:

Revised.

## 21. Comments and suggestions:

“ $d_{mn,max}$  and  $d_{mn,min}$ ”

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

### Responses and Revisions:

Thank you for the comment. We have revised it to “ $d_{mn,max}$  and  $d_{mn,min}$ ”.

## 22. Comments and suggestions:

**Line 111:** “ $Z_p$ ”

Z should be italic.

### Responses and Revisions:

Revised.

## 23. Comments and suggestions:

**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.

### Responses and Revisions:

Thank you for the comment. The construction of APM, “The APM consists of two coaxial electrodes which are rotating at an equal angular velocity and a voltage is applied between these electrodes to create an electrostatic field”, has been added.

## 24. Comments and suggestions:

**Line 123:**  $\alpha$  and  $\beta$ , in this usage, should be explicitly defined to avoid confusion with other quantities (e.g.,  $\alpha_{abs}$ ,  $\beta_{DMA}$ , etc.)

### Responses and Revisions:

Thank you for the comment. We have added “ $\alpha$  and  $\beta$  are the azimuthal flow velocity distribution parameters” and the symbols of  $\alpha$  and  $\beta$  have also been added into the table A1. The ratio of flow rates of sample flow and sheath flow of DMA and AAC have been specifically denoted as  $\beta_{DMA}$  and  $\beta_{AAC}$ , respectively.

## 25. Comments and suggestions:

**Line 136:** “Assuming a plug flow,” Delete “a”.

### Responses and Revisions:

Revised.

## 26. Comments and suggestions:

**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.

### Responses and Revisions:

Thank you for the comment. We have made the format consistent throughout and added the descriptions of min and max:

$$r_a = \min\{r_{2\_CPMA}, \max\{r_{1\_CPMA}, G_0(r_{1\_CPMA})\}\}, \quad (12)$$

$$r_b = \min\{r_{2\_CPMA}, \max\{r_{1\_CPMA}, G_0(r_{2\_CPMA})\}\}, \quad (13)$$

$$G_0(r_L) = r_c + \left(r_L - r_c + \frac{c_3}{c_4}\right) \exp(-C_4 L \bar{v}) - \frac{c_3}{c_4}, \quad (14)$$

where  $G_0(r)$  is the operator used to map the final radial position of the particle to its position at the inlet and  $\bar{v}$  is the average flow velocity.  $\min\{\}$  and  $\max\{\}$  are the minimum and maximum values of the quantities in the brackets.”.

## 27. Comments and suggestions:

**Line 161:** “ $\tau$ ”

$\tau$  should be italic.

### Responses and Revisions:

Revised.

## 28. Comments and suggestions:

**Line 162:** “denote” denoted

### Responses and Revisions:

Revised.

## 29. Comments and suggestions:

**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  $P =$  ).

### Responses and Revisions:

Thank you for the comment. We have revised it to “with a propane flow of 74.8 SCPM (standard mL per minute, flow in  $mL \min^{-1}$  converted from ambient to  $T = 298.15$  K and  $P = 101.325$  kPa) and an air flow rate of 12 SLPM (Standard L per minute, flow in  $L \min^{-1}$  converted from ambient to  $T = 298.15$  K and  $P = 101.325$  kPa)”.

## 30. Comments and suggestions:

**Line 171 and 173:** “ $Q_{sh}/Q_a = 10$ ”

the  $\beta_{DMA}$  and  $\beta_{AAC}$  formulation should be used for consistency throughout.

### Responses and Revisions:

Thank you for the comment. We have revised them to “DMA (Model 3081, TSI Inc., USA,  $\beta_{DMA} = 10$ )” and “AAC (Cambustion, Ltd., UK,  $\beta_{AAC} = 10$ )”, respectively.

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

**Line 171:** “monodisperse”

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

#### **Responses and Revisions:**

Thank you for the comment. It has been revised to “the mobility-selected aerosol flow was switched between two parallel lines and fed into the CPMA (Cambustion Ltd., UK) and AAC (Cambustion, Ltd., UK,  $\beta_{AAC} = 10$ )”.

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

**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.*

#### **Responses and Revisions:**

Thank you for the comment. We have revised it to “The distributions of particle number concentration as a function of particle mass ( $m$ ) and aerodynamic diameter ( $d_{ae}$ ) were measured by the scanning mode of the CPMA and AAC”.

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

**Line 175:** “CPMA and AAC, while” CPMA and AAC, respectively, while

#### **Responses and Revisions:**

Revised.

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

**Line 178:** “SMPS”

*Needs to be defined.*

#### **Responses and Revisions:**

Thank you for the comment. We have revised it to “the number size distribution of the generated soot particles was measured by a scanning mobility particle sizer (SMPS)”.

### **35. Comments and suggestions:**

**Line 179:** “soot particles did not change during the whole experiment.” *It is unclear to what change the authors are referring.*

#### **Responses and Revisions:**

Thank you for the comment. We have revised it to “to ensure the number size distribution of generated soot particles did not change during the whole experiment”.

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

**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.

### Responses and Revisions:

Thank you for the comment. We have revised it to “The  $m$  and  $d_{ae}$  distributions were fitted to log-normal distributions; thus, the modal values denoted as  $m_c$  and  $d_{ae,c}$  for the mobility-selected particles were determined”.

### 37. Comments and suggestions:

**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.

### Responses and Revisions:

Thank you for the comment. We have revised it to

$$\begin{aligned} N(m) &= \frac{N_0}{\sqrt{2\pi}\ln\sigma_m} \exp\left(-\frac{(\ln(m)-\ln(m_c))^2}{2(\ln\sigma_m)^2}\right) \\ N(d_{ae}) &= \frac{N_0}{\sqrt{2\pi}\ln\sigma_{dae}} \exp\left(-\frac{(\ln(d_{ae})-\ln(d_{ae,c}))^2}{2(\ln\sigma_{dae})^2}\right), \end{aligned} \quad (20)$$

where  $\sigma_m$  and  $\sigma_{dae}$  are the geometric standard deviation of  $m$  and  $d_{ae}$  distributions, respectively.  $m_c$  and  $d_{ae,c}$  are the geometric mean of  $m$  and  $d_{ae}$ , respectively”.

### 38. Comments and suggestions:

**Line 183:** “where  $\sigma$  is the geometric standard deviation and  $\mu$  is the geometric mean”

Did the distributions of  $m$  and  $Dae$  have the same  $\sigma$  and  $\mu$ ? That is what is implied.

### Responses and Revisions:

No, different distributions have different  $\sigma$  and  $\mu$ . Sorry for the confusing expression. We have revised it to “ $\sigma$  is the geometric standard deviation and  $\mu$  is the geometric mean which are fitted from  $m$  or  $d_{ae}$  distributions, respectively.”.

### 39. Comments and suggestions:

**Line 194:** “electrical diameter” electrical mobility diameter

### Responses and Revisions:

Revised.

### 40. Comments and suggestions:

**Line 196:** “ $\rho_{eff}$ ”

The mathematical relationship for  $\rho_{eff}$  has not been defined anywhere.

### Responses and Revisions:

Thank you for the comment. The mathematical relationship “ $\rho_{eff} = \frac{6m}{\pi d_m^3}$ ” has been added for  $\rho_{eff}$ .

### 41. Comments and suggestions:

**Line 202:** “where  $\Phi$  and  $\Omega$  are the transfer functions of each classification system expressed by subscripts.”

Should be “where  $\Phi$  and  $\Omega$  are the transfer functions of the combined and individual classification systems expressed by subscripts, respectively.”



## Responses and Revisions:

Revised.

### 42. Comments and suggestions:

**Line 206:** “ $dm = 80 \text{ nm}$ ”

Why is the  $dm$ ,  $m$  and  $d_{ae}$  different in this sentence than in the previous?

## Responses and Revisions:

Sorry for the mistake. We have revised it to “ $d_m = 100 \text{ nm}$ ,  $Q_{DMA} = 0.3 \text{ L min}^{-1}$ ,  $\beta_{DMA} = 0.1$ ,  $m = 0.33 \text{ fg}$ ,  $Q_{CPMA} = 0.3 \text{ L min}^{-1}$ ,  $R_m = 8$ ,  $d_{ae} = 68.3 \text{ nm}$ ,  $Q_{AAC} = 0.3 \text{ L min}^{-1}$ ,  $\beta_{AAC} = 0.1$ ”.

### 43. Comments and suggestions:

**Line 208:** “included 600 points, respectively”

600 points each?

## Responses and Revisions:

We have revised it to “which included 600 points each”.

### 44. Comments and suggestions:

**Line 210:** “ $>m_{2,max}$ , from” How far  $<$  or  $>$  the respective values were investigated?  
missing “and”

## Responses and Revisions:

We have revised it to “The ranges of  $d_m$ ,  $m$  and  $d_{ae}$  used in the calculations were from 0.8 times of  $d_{m1,min}$  to 1.2 times of  $d_{m2,max}$ , and from 0.8 times of  $m_{1,min}$  to 1.2 times of  $m_{2,max}$ , from 0.8 times of  $d_{ae,min}$  to 1.2 times of  $d_{ae,max}$ , respectively.”.

### 45. Comments and suggestions:

**Line 213:** “The DMA-CPMA transfer function is calculated in  $\log(dm)$ - $\log(m)$  space, as shown in Fig. 2.”  
“DMA-CPMA transfer function ( $\Phi_{DMA-CPMA}$ )”. What is the transfer function calculated for?

## Responses and Revisions:

Thank you for the comment. It is calculated for particles mentioned in Line 206 and 207, i.e. particles with  $dm$  of 100 nm and  $m$  of 0.33 fg. It has been revised to “The DMA-CPMA transfer function ( $\Phi_{DMA-CPMA}$ ) for particles mentioned above, i.e. particles with  $d_m$  of 100 nm and  $m$  of 0.33 fg, is calculated in  $\log(d_m)$ - $\log(m)$  space, as shown in Fig. 2”.

### 46. Comments and suggestions:

**Line 215 and 216:** “nm”

nm should not be italicized when used as a unit.

## Responses and Revisions:

Thank you for the comment. It has been revised.

### 47. Comments and suggestions:

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

$D_{fm}$  can be larger than 3 for particles that are non-spherical at small  $D_m$  and approach spherical as  $D_m$  increases.

#### Responses and Revisions:

Thank you for the comment. We have revised it to “In general,  $D_{fm}$  equals 3 for spherical particles and smaller than 3 for aspherical particles, although  $D_{fm}$  can be larger than 3 for particles that are non-spherical at small  $d_m$  and approach spherical as  $d_m$  increases”.

#### 48. Comments and suggestions:

**Line 219:** “Under this specific operation condition” What specific operating condition? Please explain.

#### Responses and Revisions:

Sorry we didn’t make it clear. This specific operating condition refers to “ $d_m = 100$  nm,  $Q_{DMA} = 0.3$  L min<sup>-1</sup>,  $\beta_{DMA} = 0.1$ ,  $m = 0.33$  fg,  $Q_{CPMA} = 0.3$  L min<sup>-1</sup>,  $R_m = 8$ ” which mentioned in Line 206 and Line 207.

#### 49. Comments and suggestions:

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

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

#### Responses and Revisions:

Yes, this is a “theoretical” spherical particle population. The effective densities of this particles are the same as nominal particles with  $d_m$  of 100 nm and  $m$  of 0.33 fg, i.e. 630.3 kg m<sup>-3</sup>.

#### 50. Comments and suggestions:

**Line 220:** “classification region”

What is the classification region? Please elaborate.

#### Responses and Revisions:

Thank you for the comment. The classification region is presented as the rainbow color blocks. We have revised it to “no overlap was observed between the spherical particle population (black line) and the classification region (the colored blocks) for doubly charged particles”.

#### 51. Comments and suggestions:

**Line 231:** “than the slope of a line connecting  $(d_m, m) = (d_{m2,min}, m_{2,max})(d_{m1}, m_1)$  (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  $D_{fm} = 2.28$  line. So, while the  $D_{fm} = 3$  discussion seems reasonable, the  $D_{fm} = 2.28$  does not. In contrast, for Figure 4 the PP<sub>0</sub> 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  $d_{m1}$  where PP0 is clearly visible.

#### Responses and Revisions:

Sorry we didn’t make it clear. We have added a dashed line to indicate PP<sub>0</sub>. It is easier to compare the  $D_{fm} = 2.28$  and PP<sub>0</sub>.

## 52. Comments and suggestions:

**Line 236:** “. Accordingly, the ideal condition...” Under static operation at this set point

### Responses and Revisions:

Thank you for the comment. We have revised it to “Accordingly, the ideal condition under static operation to completely eliminate the multiply charged particles is”.

## 53. Comments and suggestions:

**Line 240:** “Eq. (26) gives instructions in actual operation” It is unclear how Eq. 26 gives instructions.

### Responses and Revisions:

Thank you for the comment. We have added an equation of  $R_m$ :

“The mass resolution ( $R_m$ ) of CPMA is related to particles mobility. When selecting the particles with mass of  $m_1$  and mobility of  $B_1$ , the  $R_m$  can be calculated by

$$R_m = \frac{2\pi B_1 L_{CPMA} r_c^2 \omega^2 m_1}{Q_{CPMA}}, \quad (15)$$

where  $\omega$  is the equivalent rotational speed calculated by  $\omega = \alpha + \frac{\beta}{r_c^2}$ ,  $m_1$  is the nominal mass that the CPMA can select. The limiting mass can be calculated by

$$m_{n,min}^{n,max} = n \cdot m_1 \pm \frac{Q_{CPMA}}{2\pi B_{n,min}^{n,max} L_{CPMA} r_c^2 \omega^2} = n \cdot m_1 \pm \frac{m_1}{R_m} \cdot \frac{B_1}{B_{n,min}^{n,max}}, \quad (16)$$

where  $m_{n,min}^{n,max}$  and  $B_{n,min}^{n,max}$  are the maximum and minimum mass and corresponding mobility of particles bearing number of elementary charges of  $n$  that the CPMA can select, respectively.”

and this sentence has been changed to “Combining Eq. (16), equation (26) gives instructions in actual operation to eliminate multiply charged particles. When selecting particles of certain  $d_m$  and  $m$ , by decreasing  $Q_{CPMA}$ , or increasing  $\omega$  and  $\beta_{DMA}$ , i.e., by increasing the resolution of the measurement, the potential of multiply charged particles is reduced”.

## 54. Comments and suggestions:

**Line 244:** “and the slope of PP0 derived from the actual condition”

This is unclear as written. How is the slope of PP<sub>0</sub> derived from actual conditions? Weren't the transfer functions from which PP<sub>0</sub> is determined theoretically calculated?

### Responses and Revisions:

We have revised it to “the key to evaluating whether there is a multiple charging effect lies in the particle morphology ( $D_{fm}$ ) and the slope of PP<sub>0</sub> calculated from Eq. (26) theoretically”.

## 55. Comments and suggestions:

**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<sub>0</sub>, so it is unclear where this value is coming from. What was the slope of PP<sub>0</sub> 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 ( $D_{fm} = 3$ ) 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.

### Responses and Revisions:

Thank you for the advice. We have removed it.

**56. Comments and suggestions:**

**Line 251:** “ $R_m = 8$ ”

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

**Responses and Revisions:**

Thank you for the comment. The description of  $R_m$  has been added which can be found in the reply to comment 53.

**57. Comments and suggestions:**

**Line 254:** “the slope” the critical slope

**Responses and Revisions:**

Revised.

**58. Comments and suggestions:**

**Line 251:** “contour lines in Fig. 3”

How were these contours calculated? From Eq. 26? If so, how was  $dm_2$  determined?

**Responses and Revisions:**

Yes, they were calculated from Eq. 26. We have revised this sentence to “Here, we simulate the critical slope of  $PP_0$  when selecting different  $d_m$  and  $m$  under the common selecting conditions ( $\beta_{DMA} = 0.1$ ,  $Q_{CPMA} = 0.3 \text{ L min}^{-1}$ ,  $R_m = 8$ ) using Eq. 26, which is represented as contour lines in Fig. 3”. The values of  $d_{m2,min}$  were calculated from Eq. 1 and  $(1 \pm \beta_{DMA}) \cdot Z_p^*$ , and we have added the description in Line 108 and Line 109 : “The limiting electrical mobilities that DMA can select are  $(1 \pm \beta_{DMA}) \cdot Z_p^*$ . The maximum and minimum values of  $d_m$  for particles with  $n$  charges can be derived combining  $(1 \pm \beta_{DMA}) \cdot Z_p^*$  and Eq. 1, and denote as  $d_{mn,max}$  and  $d_{mn,min}$ , respectively”.

**59. Comments and suggestions:**

**Line 259:** “mobility diameters larger than 200 nm, while it fails to eliminate ...”

As shown by the circles and squares in Figure 3.

**Responses and Revisions:**

Revised.

**60. Comments and suggestions:**

**Line 263:** “to charge aerosol particles”

To Boltzmann distribution or a known charge state?

**Responses and Revisions:**

Thank you for the comment. We have revised it to “The advantage of the AAC versus the CPMA is that there is no need for a neutralizer to charge aerosol particles to a known charge state”.

**61. Comments and suggestions:**

**Line 264:** “AAC cannot constrain the properties of aspherical particles as monodisperse as DMA or CPMA classification”

*Unclear as written.*

#### **Responses and Revisions:**

It has been revised to “However, aspherical particles with different mass can be selected by the AAC as having identical aerodynamic diameter”.

#### **62. Comments and suggestions:**

*Line 267: “selecting the same representative particles” The same as what? Please give values.*

#### **Responses and Revisions:**

The same particles as particles used in DMA-CPMA calculation, which is mentioned in Line 204 and Line 205. This sentence and the Line 273 are repeated, so we deleted this sentence and revised the Line 273 to “To simulate the transfer function of the DMA-AAC, the same particles ( $d_m = 100$  nm,  $m = 0.33$  fg,  $D_{fm} = 2.28$ ) as those used in the calculations of the DMA-CPMA were selected”.

#### **63. Comments and suggestions:**

*Line 268: “aspherical particles can be expressed as follows”*

*The  $\log(D_{ae})$  is expressed on the next line. Not aspherical particles.*

#### **Responses and Revisions:**

Thank you for the comment. We have revised it to “the relationship of  $d_{ae}$  and  $d_m$  of aspherical particles can be expressed as follows”.

#### **64. Comments and suggestions:**

*Line 273: “the same particles” Please give values as a reminder.*

#### **Responses and Revisions:**

Thank you for the comment. It has been revised to “To simulate the transfer function of the DMA-AAC, the same particles ( $d_m = 100$  nm,  $m = 0.33$  fg,  $D_{fm} = 2.28$ ) as those used in the calculations of the DMA-CPMA were selected. The corresponding  $d_{ae}$  was numerically solved using the known mass–mobility relationship. The transfer function of the DMA-AAC is shown in  $\log(d_{ae})$ - $\log(d_m)$  (Fig. 4a).”.

#### **65. Comments and suggestions:**

*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  $D_m$ ?*

#### **Responses and Revisions:**

Thank you for the comment. We have revised it to “In the transfer function of DMA-CPMA, the classification regions of singly charged particles and doubly charged particles are on the diagonal. The oblique line of particles population is more likely to go through the region of doubly charged particles in the transfer function of DMA-CPMA. The transfer functions of singly charged and doubly charged particles are in parallel for the DMA-AAC, suggesting that the particles population is less likely to overlap with the region of multiply charged particles”.

#### **66. Comments and suggestions:**

**Line 277:** “the example setups” What example setups?

**Responses and Revisions:**

It was mentioned in Line 206 and Line 207. We have revised it to “Using the example setups ( $d_m = 100$  nm,  $Q_{DMA} = 0.3$  L min<sup>-1</sup>,  $\beta_{DMA} = 0.1$ ,  $d_{ae} = 68.3$  nm,  $Q_{AAC} = 0.3$  L min<sup>-1</sup>,  $\beta_{AAC} = 0.1$ .) of the DMA-AAC, truly monodispersed particles are selected for spherical particles and typical soot particles.”.

**67. Comments and suggestions:**

**Line 290:** “which is the case for most atmospheric aerosol particles.” What is the case? This  $D_{fm}$  is smaller than for most aerosols.

**Responses and Revisions:**

Thank you for the comment. We have revised it to “This  $D_{fm}$  is smaller than that for most aerosols”.

**68. Comments and suggestions:**

**Line 292:** “is required” “may be required”

**Responses and Revisions:**

Revised.

**69. Comments and suggestions:**

**Line 295:** “When increasing  $\beta_{AAC}$  to 0.3”

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

**Responses and Revisions:**

Thank you for the comment. We have revised it to “When increasing  $\beta_{AAC}$  to 0.3 (decreasing the resolution of AAC) and leaving  $\beta_{DMA}$  unchanged, the transfer function becomes broader”.

**70. Comments and suggestions:**

**Line 302:** “the corresponding  $d_{ae}$  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  $\rho_{eff}$  were determined.

**Responses and Revisions:**

Thank you for the comment. It has been revised to “For each mobility-selected particles, 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  $\rho_{eff}$  were determined”.

**71. Comments and suggestions:**

**Line 304:** “measured spectral density” Measured distributions?

**Responses and Revisions:**

Thank you for the comment. It has been changed to “Representative plots for the measured distributions of  $m$  and  $d_{ae}$  of particles with  $d_m$  of 150 nm and 250 nm are shown in Fig. S2”.

## **72. Comments and suggestions:**

**Line 305:** “The results are summarized in Table 2.”

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

### **Responses and Revisions:**

Thank you for the comment. We have added the description of the uncertainties: “The uncertainties of  $d_{ae,c}$  and  $m_c$  were standard deviation of multiple measurements”.

## **73. Comments and suggestions:**

**Line 306:**  $k_f$  has units of mass.

### **Responses and Revisions:**

Thank you for the comment. The unit of “fg” has been added.

## **74. Comments and suggestions:**

**Line 308:** “two methods” Which two methods?

### **Responses and Revisions:**

Thank you for the comment. We have revised it to “The effective densities of generated soot particles vary from  $>500 \text{ kg m}^{-3}$  at  $d_m = 80 \text{ nm}$  to  $<300 \text{ kg m}^{-3}$  at  $d_m$  of  $250 \text{ nm}$  determined by DMA-CPMA and DMA-AAC”.

## **75. Comments and suggestions:**

**Line 308:** “the deviation” What deviation?

### **Responses and Revisions:**

Thank you for the comment. We have revised it to “the deviation of values of  $\rho_{\text{eff}}$  measured by DMA-CPMA and DMA-AAC monotonically decreases with increasing particle size”.

## **76. Comments and suggestions:**

**Line 316:** “the corresponding transfer function” DMA-CPMA transfer function?

### **Responses and Revisions:**

Thank you for the comment. It has been revised to “the corresponding DMA-CPMA transfer function is shown in Fig. 5a”.

## **77. Comments and suggestions:**

**Line 317:** “The particle population” Shown by the red  $D_{fm} = 2.28$  line?

### **Responses and Revisions:**

Thank you for the comment. It has been revised to “The particle population (red line) overlaps the transfer function region of doubly charged particles, suggesting the potential interferences of doubly charged particles in DMA-CPMA selection”.

## **78. Comments and suggestions:**

**Line 320:** “particles number aerodynamic size distribution”

Should be “particles number density aerodynamic size distribution”

**Responses and Revisions:**

Revised.

**79. Comments and suggestions:**

**Line 324:** “The mean  $d_{ae}$  values” For particles with  $d_m = 80$  nm?

**Responses and Revisions:**

Yes, it is. We have revised it to “For particles with  $d_m = 80$  nm, the mean  $d_{ae}$  values were 53.9 nm, 60.6 nm and 70.9 nm, and the corresponding  $d_{ae}$  values were calculated as 51.5 nm, 62.0 nm and 70.7 nm using Eq. (1) and Eq. (16)”.

**80. Comments and suggestions:**

**Line 327:** “In contrast, ...”

In contrast to what?

**Responses and Revisions:**

Thank you for the comment. It has been deleted.

**81. Comments and suggestions:**

**Line 332:** “PP0”

Subscript 0.

**Responses and Revisions:**

Revised.

**82. Comments and suggestions:**

**Line 363:** “ $26.7 \pm 3.0\%$ ”

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

**Responses and Revisions:**

Thank you for the comment. We have revised all the units.

**83. Comments and suggestions:**

**Line 387:** “Under the same setups” Same as what?

**Responses and Revisions:**

In this sentence, we tried to discuss the influence of particles  $D_{fm}$  and selected size on the multiple charging effect when the setups of DMA-CPMA is unchanged. We are sorry for the confusing expression. We have deleted it and this sentence has been revised to “This tandem system is more sensitive to multiple charging effect with decreasing  $D_{fm}$  and decreasing nominal size of particles.”.

**84. Comments and suggestions:**



**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.

### Responses and Revisions:

Thank you for the comment. It has been changed.

### 85. Comments and suggestions:

**Table 2:** “ $M(fg)$ ”

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

### Responses and Revisions:

It has been revised to “ $m_c$ ”.

### 86. Comments and suggestions:

**Table 3:**  $f_N$  and  $f_{abs}$  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?

### Responses and Revisions:

Thank you for the comment. We have added the description of  $f_N$ ,  $f_{abs}$  and overestimation of MAC:

“The fractional number concentration of particles with different charging state is expressed as follows,

$$f_{N,n} = \frac{\int_{d_{ae,min}}^{d_{ae,max}} \frac{dN_n}{d\log(d_{ae})} d\log(d_{ae})}{\sum_{i=1}^3 \int_{d_{ae,min}}^{d_{ae,max}} \frac{dN_n}{d\log(d_{ae})} d\log(d_{ae})}, \quad (29)$$

where  $f_{N,n}$  and  $N_n$  are the fractional number concentration and number concentration of particles bearing  $n$  charges.  $d_{ae,min}$  and  $d_{ae,max}$  denote the minimum and maximum value of  $d_{ae}$  scanned by AAC, respectively. The uncertainties are standard deviations of multiple measurements”.

“The fractional absorption coefficient for particles with different charging state was calculated as follows,

$$f_{abs,n} = \frac{\int_{d_{ve,low,n}}^{d_{ve,high,n}} \frac{d\alpha_{abs,n}}{d\log(d_{ve})} d\log(d_{ve})}{\sum_{i=1}^3 \int_{d_{ve,low,n}}^{d_{ve,high,n}} \frac{dN_n}{d\log(d_{ve})} d\log(d_{ve})}, \quad (30)$$

where  $f_{abs,n}$  and  $\alpha_{abs,n}$  are the fractional absorption coefficient and absorption coefficient of particles bearing  $n$  charges, respectively.  $d_{ve,low,n}$  and  $d_{ve,high,n}$  denote the minimum and maximum value of  $d_{ve}$  of particles with  $n$  charges, which are converted from  $d_{ae,low}$  and  $d_{ae,high}$  scanned by AAC, respectively.

The overestimation of mass absorption cross-section (MAC) was calculated by

$$\frac{\Delta MAC}{MAC} = \frac{\frac{\alpha_{abs,tot}}{m_p N_{tot}} \frac{f_{abs,1} \alpha_{abs,tot}}{m_p f_{N,1} N_{tot}}}{\frac{f_{abs,1} \alpha_{abs,tot}}{m_p f_{N,1} N_{tot}}} = \frac{f_{N,1}}{f_{abs,1}} - 1, \quad (31)$$

where  $\alpha_{abs,tot}$  and  $N_{tot}$  is the total absorption coefficient and number concentration of particles selected by DMA-CPMA, respectively.  $m_p$  is the actual mass of singly charged particles selected by DMA-CPMA. The uncertainties were calculated from propagation of errors”

### 87. Comments and suggestions:

**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.

### Responses and Revisions:

Thank you for the comment. We have revised it to “Example of the DMA-CPMA transfer function of flame-generated soot particles (Pei et al., 2018) in  $\log(m)$ - $\log(d_m)$ ”.

## 88. Comments and suggestions:

**Line 533:** “DMA and CPMA.”

DMA and CPMA, respectively.

## Responses and Revisions:

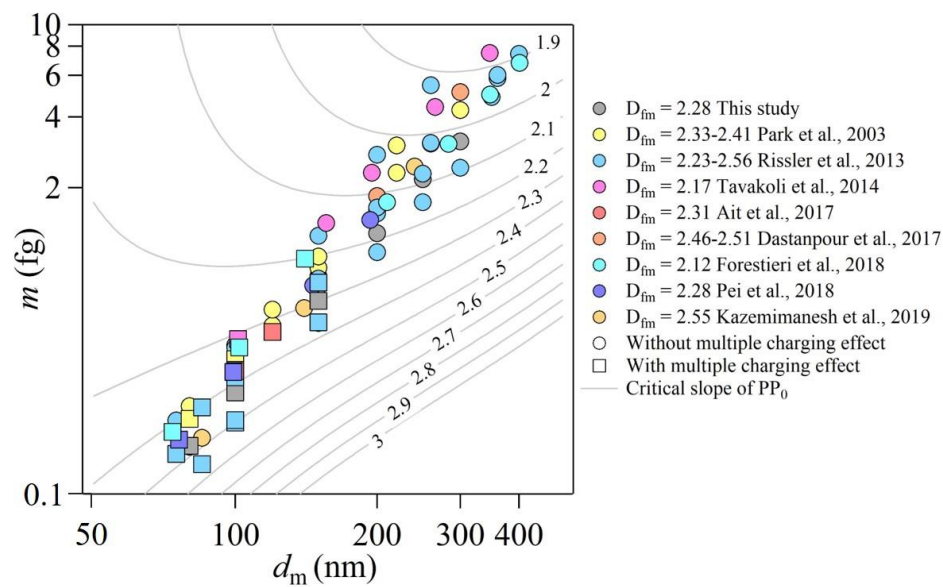
Revised.

## 89. Comments and suggestions:

**Figure 3:** What are the minimum values on the  $d_m$  and  $m$  axis?

## Responses and Revisions:

Thank you for the comment. The minimum values on the  $d_m$  and  $m$  axis has been added.



## 90. Comments and suggestions:

**Line 537:** “The contour lines denote the slope” Critical slope?

## Responses and Revisions:

Revised.