



1 **Characterization of tandem aerosol classifiers for selecting**  
2 **particles: implication for eliminating multiple charging**  
3 **effect**

4 Yao Song<sup>1</sup>, Xiangyu Pei<sup>1</sup>, Huichao Liu<sup>1</sup>, Jiajia Zhou<sup>1</sup>, Zhibin Wang<sup>1,2,3,4\*</sup>

5 <sup>1</sup>College of Environmental and Resource Sciences, Zhejiang University, Hangzhou, 310058, China

6 <sup>2</sup>Hangzhou Global Scientific and Technological Innovation Center, Zhejiang University, Hangzhou, 311200,  
7 China

8 <sup>3</sup>Zhejiang Provincial Key Laboratory of Organic Pollution Process and Control, Hangzhou 310058, China.

9 <sup>4</sup>Key Laboratory of Environment Remediation and Ecological Health, Ministry of Education, Zhejiang  
10 University, 310058 Hangzhou, China

11

12 *correspondence to:* Zhibin Wang (wangzhibin@zju.edu.cn)

13 **Abstract.** Accurate particle classification plays a vital role in aerosol studies. Differential mobility analyzer  
14 (DMA), centrifugal particle mass analyzer (CPMA) and aerodynamic aerosol classifier (AAC) are commonly  
15 used to select particles with a specific size or mass. However, multiple charging effect cannot be entirely  
16 avoided either using individual technique or using tandem system such as DMA-CPMA, especially when  
17 selecting soot particles with fractal structures. In this study, we demonstrate the transfer functions of DMA-  
18 CPMA and DMA-AAC systems, as well as the potential multiple charging effect. Our results show that the  
19 ability to remove multiply charged particles mainly depends on particles morphology and instruments setups  
20 of DMA-CPMA system. Using measurements from soot experiments and literature data, a general trend in  
21 the appearance of multiple charging effect with decreasing size when selecting aspherical particles was  
22 observed. Otherwise, our results indicated that the ability of DMA-AAC to resolve particles with multiple  
23 charges is mainly related to the resolutions of classifiers. In most cases, DMA-AAC can eliminate multiple  
24 charging effect regardless of the particle morphology, while particles with multiple charges can be selected  
25 when decreasing resolutions of DMA and AAC. We propose that the multiple charging effect should be  
26 reconsidered when using DMA-CPMA or DMA-AAC system in estimating size and mass resolved optical  
27 properties in the field and lab experiments.

28 **1 Introduction**

29 Atmospheric aerosol particles span a wide size range from 1 nm to > 100  $\mu\text{m}$ . Significant size dependence of  
30 aerosol physicochemical properties has been widely reported. Particle size can strongly alters the hygroscopic  
31 behavior (Biskos et al., 2006), phase state (Cheng et al., 2015) and cloud-nucleating ability (Dusek et al.,  
32 2006) of aerosol nanoparticles, indicating its importance when assessing the climate effect. Hence, accurate  
33 particle classification is essential to investigate the size dependence behavior of aerosol particles.



34 At present, particles are generally classified by either size or mass in atmospheric aerosol studies. Differential  
35 mobility analyzer (DMA) is the most commonly used size classifier, which selects particles based on the  
36 electrical mobility (Knutson and Whitby, 1975). Particle mass analyzer (PMA) includes the aerosol particle  
37 mass analyzer (APM) and the centrifugal particle mass analyzer (CPMA), both of which classify particles  
38 based on their mass-to-charge ratio (Ehara et al., 1996; Olfert and Collings, 2005). However, particles are  
39 required to be pre-charged when classified by the DMA or PMA, resulting in that particles with higher-order  
40 charges and identical apparent mobility or mass-to-charge ratio will be selected simultaneously, which are  
41 referred to as the multiple charging effect. This may introduce uncertainty in the subsequent characterization.  
42 Radney et al. (2013) demonstrated that although the single-charged particles account for the highest number  
43 fraction (46.3%) of the DMA-classified particles (200 nm), while their contributions to the total mass  
44 concentration and extinction are insignificant (10.8% and 7.96%, respectively). Thus, the reported extinction  
45 of particles with a certain diameter was greatly overestimated due to the multiple charging effect.  
46 Previous studies (Shiraiwa et al., 2010; Rissler et al., 2013; Johnson et al., 2014; Johnson et al., 2021) tried to  
47 utilize the combination of size and mass classifiers, like DMA-APM or DMA-CPMA system, to obtain singly  
48 charged particles. Theoretically, the ability of DMA-APM to eliminate multiply charged particles is governed  
49 by the particles morphology and setups of DMA-APM (Kuwata, 2015). This conclusion implies that it can  
50 hardly be achieved that all the multiply charged particles are effectively excluded for aspherical particles,  
51 especially for soot particles. Radney and Zangmeister (2016) conducted the limitation of DMA-APM with  
52 three types of particles (polystyrene Latex spheres (PSL), ammonia sulfate (AS) and soot particles), the  
53 results demonstrated that DMA-APM can resolve multiply charged particles for spherical particles (PSL and  
54 AS particles), but it failed for aspherical soot particles. Multiply charged soot particles led to over 110%  
55 errors in retrieving mass specific extinction cross section.  
56 In contrast to DMA and PMA, aerodynamic aerosol classifier (AAC) is a novel instrument which selects  
57 aerodynamic equivalent diameter of aerosol particles based on their relaxation time. The advantage of  
58 utilizing AAC is that no charging process is needed in particle classification compared with the  
59 aforementioned classifiers, hence the multiple charging effects can be avoided (Tavakoli and Olfert, 2013).  
60 Morphology information, such as effective density ( $\rho_{\text{eff}}$ ), mass-mobility exponent ( $D_{\text{fm}}$ ) and dynamic shape  
61 factor ( $\chi$ ), can be inferred using the tandem systems of DMA-PMA (Park et al., 2003; Zhang et al.,  
62 2008; Rissler et al., 2013; Pei et al., 2018; Zangmeister et al., 2018), DMA-AAC (Tavakoli and Olfert, 2014)  
63 and AAC-CPMA (Johnson et al., 2018), respectively.  
64 The theoretical transfer functions of individual classifier (DMA, CPMA and AAC) and DMA-APM system  
65 have been previously discussed (Knutson and Whitby, 1975; Ehara et al., 1996; Olfert and Collings, 2005;  
66 Stolzenburg and McMurry, 2008; Tavakoli and Olfert, 2013). In this study, we calculated the transfer  
67 functions of DMA-AAC and DMA-CPMA systematically. Combined with soot experiments, we  
68 demonstrated that multiple charging effects may still exist after DMA-CPMA classification when selecting  
69 aspherical particles, and evaluated the light absorption of selected particles with different charging states  
70 using Mie theory. Furthermore, we proposed the operating condition for DMA-CPMA to eliminate the



71 multiply charged particles in the future studies. Our results suggest that the size- and mass-resolved optical  
 72 properties may be overestimated for small soot particles when using DMA-CPMA system, which will lower  
 73 the accuracy of predicting soot climate effect.

## 74 2 Theory and experiment

### 75 2.1 Transfer function for individual aerosol classifier

#### 76 DMA

77 The DMA, consisting of two coaxial electrodes, classifies particle based upon electrical mobility  $Z_p$  (Knutson  
 78 and Whitby, 1975), which can be calculated as follows:

$$79 Z_p = qB = \frac{neCc(d_p)}{3\pi\mu d_p}, \quad (1)$$

80 where  $q$  is the particle charge,  $n$  is the number of particle charges,  $B$  is the mobility of particle,  $e$  is the  
 81 elemental charge,  $\mu$  is the viscosity of the air,  $Cc(d_p)$  is the Cunningham slip correction factor. When aerosol  
 82 inlet flow rate equals to aerosol sampling outlet flow rate, the  $Z_p^*$  selected by the DMA is defined as

$$83 Z_p^* = \frac{Q_{sh}}{2\pi V_{DMA} L_{DMA}} \ln\left(\frac{r_{2\_DMA}}{r_{1\_DMA}}\right), \quad (2)$$

84 where  $Q_{sh}$  is the sheath flow rate,  $V_{DMA}$  is the voltage between the two electrodes,  $L_{DMA}$  is the length of DMA,  
 85  $r_{1\_DMA}$  and  $r_{2\_DMA}$  are the inner and outer radii of DMA, respectively. The transfer function of DMA can be  
 86 expressed as follows when particle diffusion is negligible (Knutson and Whitby, 1975; Stolzenburg and  
 87 McMurry, 2008),

$$88 \Omega(\tilde{Z}_p, \beta_{DMA}) = \frac{1}{2\beta_{DMA}} [|\tilde{Z}_p - (1 + \beta_{DMA})| + |\tilde{Z}_p - (1 - \beta_{DMA})| - 2|\tilde{Z}_p - 1|], \quad (3)$$

89 where the  $\tilde{Z}_p = Z_p/Z_p^*$ ,  $\beta_{DMA} = Q_a/Q_{sh}$ , and  $Q_a$  is the sample flow rate. The transfer function is an isosceles  
 90 triangle with value of 1 at  $Z_p^*$  and going to 0 at  $(1 \pm \beta_{DMA}) \cdot Z_p^*$ .

#### 91 CPMA

92 The construction of CPMA is similar to the APM, but its inner cylinder rotates faster than outer one to create  
 93 a stable system of forces (Olfert and Collings, 2005). In the CPMA, the equation of particles motion is  
 94 expressed as

$$95 \frac{m}{\tau} \frac{dr}{dt} = \frac{mv_\theta(r)^2}{r} - \frac{qV_{CPMA}}{r \ln\left(\frac{r_{2\_CPMA}}{r_{1\_CPMA}}\right)}, \quad (4)$$

96 and the trajectory equation is

$$97 \frac{dr}{dz} = \frac{dr}{dt} \left(\frac{dz}{dt}\right)^{-1} = \frac{c_r}{v_z}, \quad (5)$$

98 where  $\tau$  is the relaxation time,  $m$  is the mass of the particle,  $t$  is time,  $V$  is the voltage difference between the  
 99 two electrodes,  $r_{1\_CPMA}$  and  $r_{2\_CPMA}$  are the radii of inner and outer electrodes, respectively.  $c_r$  is particle  
 100 migration velocity,  $v_z$  is the axial flow distribution and  $v_\theta$  is the velocity profile in the angular direction,

$$101 v_\theta = \omega_1 \frac{\hat{r}^2 - \hat{\omega}}{\hat{r}^2 - 1} r + \omega_1 r_{1\_CPMA}^2 \frac{\hat{\omega} - 1}{\hat{r}^2 - 1} \frac{1}{r} = \alpha r + \frac{\beta}{r}, \quad (6)$$



102 where  $\hat{\omega} = \omega_2/\omega_1$  is the ratio of the rotational speed of the outer electrode to the inner electrode and  $\omega_1$  and  
 103  $\omega_2$  are the rotational speed of the inner and outer electrode, respectively.  $\hat{r}$  is the ratio of the inner and outer  
 104 radius.

105 Sipkens et al. (2019) gave the methods to calculate the transfer function of CPMA. They proposed that the  
 106 Taylor series expansion at the center of the gap ( $r_c=(r_{2\_CPMA}+r_{1\_CPMA})/2$ ) is much simpler and more robust. In  
 107 this case, the particle migration velocity in the radial direction is

$$108 \quad c_r \approx C_3 + C_4(r - r_c), \quad (7)$$

109 where

$$110 \quad C_3 = \tau \left( \alpha^2 r_c + \frac{2\alpha\beta}{r_c} + \frac{\beta^2}{r_c^2} - \frac{C_0}{mr_c} \right), \quad (8)$$

$$111 \quad C_4 = \tau \left( \alpha^2 - \frac{2\alpha\beta}{r_c} - \frac{3\beta^2}{r_c^2} + \frac{C_0}{mr_c^2} \right), \quad (9)$$

$$112 \quad C_0 = \frac{qV_{CPMA}}{\ln(r_{2\_CPMA}/r_{1\_CPMA})}, \quad (10)$$

113 Assuming a plug flow, the transfer function should be

$$114 \quad \Omega = \frac{r_b - r_a}{2\delta}, \quad (11)$$

115 where  $\delta=(r_{2\_CPMA}-r_{1\_CPMA})/2$  is the half width of the gap between the two electrodes, and

$$116 \quad r_a = \min \left\{ r_{2\_CPMA}, \max \{ r_{1\_CPMA}, G_0(r_{1\_CPMA}) \} \right\}, \quad (12)$$

$$117 \quad r_b = \min \left\{ r_{2\_CPMA}, \max \{ r_{1\_CPMA}, G_0(r_{2\_CPMA}) \} \right\}, \quad (13)$$

$$118 \quad G_0(r_L) = r_c + \left( r_L - r_c + \frac{C_3}{C_4} \right) \exp(-C_4 L \bar{v}) - \frac{C_3}{C_4}, \quad (14)$$

119 where  $G_0(r)$  is the operator to map the final radial position of the particle to its position at the inlet,  $\bar{v}$  is  
 120 average flow velocity.

121 Reavell et al. (2011) calculated the resolution of CPMA assuming that the gap between two electrodes is  
 122 narrow enough that variation of force in the gap can be ignored. The limiting mass can be calculated by

$$123 \quad m_{1,min}^{1,max} \omega^2 r - qE = \pm \frac{Q_{CPMA}}{2\pi B_{1,min}^{1,max} L_{CPMA} r_c^2 \omega^2}, \quad (15)$$

124 where  $\omega$  is the equivalent rotational speed calculated by  $\omega = \alpha + \frac{\beta}{r_c}$ ,  $m_{1,min}^{1,max}$  and  $B_{1,min}^{1,max}$  are the maximum  
 125 and minimum mass and corresponding mobility with single charge that CPMA can select, respectively.  
 126 Further details can be found in Reavell et al. (2011) and Sipkens et al. (2019).

## 127 AAC

128 The AAC classifies particle based on relaxation time, which is defined by

$$129 \quad \tau = Bm = \frac{Cc(d_{ae})\rho_0 d_{ae}^2}{18\mu}, \quad (16)$$

130 where  $\mu$  is the viscosity of air.  $Cc(d_{ae})$  is the slip correction factor.  $\rho_0$  is the standard density with value of 1  
 131 g/cm<sup>3</sup> (Johnson et al. 2018). When aerosol inlet flow rate equals to aerosol sampling outlet flow rate, it can  
 132 be expressed as (Tavakoli and Olfert, 2013)

$$133 \quad \Omega = \frac{1}{2\beta_{AAC}} [|\bar{\tau} - (1 - \beta_{AAC})| + |\bar{\tau} - (1 + \beta_{AAC})| - 2|\bar{\tau} - 1|], \quad (17)$$



134  $\tau^*$  is the nominated relaxation time which is classified by the AAC,

$$135 \quad \tau^* = \frac{2Q_{sh}}{\pi\omega^2(r_{1\_AAC}+r_{2\_AAC})^2L}, \quad (18)$$

136 where  $\beta_{AAC} = \frac{Q_a}{Q_{sh}}$ ,  $\tilde{\tau} = \frac{\tau}{\tau^*}$ ,  $r_{1\_AAC}$  and  $r_{2\_AAC}$  are the inner and outer radii of AAC, respectively.

## 137 2.2 Experimental setup

138 A schematic of the experimental setup is illustrated in Fig. 1. Soot particles were generated by a miniature  
 139 inverted soot generator (Argonaut Scientific Ltd., Canada). Detailed aerosol generation methods can be found  
 140 in Moallemi et al. (2019). The poly-dispersed aerosols were dried to a relative humidity of <20% by a silica  
 141 dryer, and then were passed through a soft X-ray neutralizer (Model 3088, TSI, Inc., USA). Five mobility  
 142 diameters (80, 100, 150, 200 and 250 nm) of soot particles were selected with DMA (Model 3081, TSI Inc.,  
 143 USA,  $Q_{sh}/Q_a = 10$ ). For the soot characterization, the monodisperse aerosol flow was switched between two  
 144 parallel lines and fed into CPMA (Cambustion Ltd., UK) and AAC (Cambustion, Ltd., UK,  $Q_{sh}/Q_a = 10$ ),  
 145 respectively. The particles mass ( $m$ ) and aerodynamic diameter ( $d_{ae}$ ) were determined by stepping mode of  
 146 CPMA and AAC while the condensation particle counter (CPC, Model 3756, TSI, Inc., USA) recorded their  
 147 corresponding concentration at each setpoint, respectively. The  $m$  and  $d_{ae}$  distributions were measured and  
 148 fitted to log-normal distribution, thus the mode  $m$  and  $d_{ae}$  for the mobility-selected particles were determined.  
 149 CPMA and AAC were calibrated with certified PSL spheres (Thermo, USA) with sizes of 70, 150 and 303  
 150 nm before the measurement. The measured  $m$  and  $d_{ae}$  were compared to  $m_{PSL}$  and  $d_{ae, PSL}$  which were  
 151 calculated with the nominal diameter and density of PSL. The deviations between measured  $m$  and  $m_{PSL}$  or  
 152 measured  $d_{ae}$  and  $d_{ae, PSL}$  were 2.75% and 5.14%, respectively. In order to quantify the multiple charging  
 153 effect of particles selected by DMA-CPMA system, the  $d_{ae}$  distribution of twice classified particles was  
 154 obtained by stepping the AAC rotation speed of the cylinder with simultaneous measurement of the particle  
 155 concentration at the AAC outlet using a CPC (Fig. 1b).

## 156 3 Results and discussion

### 157 3.1 Transfer function of the tandem system

158 The DMA, PMA and AAC select particles based on the electrical diameter, mass and aerodynamic diameter,  
 159 respectively. These properties can be connected as follows (Decarlo et al. 2004)

$$160 \quad \frac{cc(d_{ae})\rho_0 d_{ae}^2}{6} = \frac{cc(d_m)\rho_{eff} d_m^2}{6} = m \frac{cc(d_m)}{\pi d_m}, \quad (19)$$

161 The transfer function of DMA-APM has been well documented, which can be found in Kuwata (2015). The  
 162 convolution of transfer functions of DMA-CPMA and DMA-AAC were calculated by the following  
 163 equations.

$$164 \quad \Phi_{DMA-CPMA} = \Omega_{CPMA}\Omega_{DMA}, \quad (20)$$

$$165 \quad \Phi_{DMA-AAC} = \Omega_{DMA}\Omega_{AAC}, \quad (21)$$



166 where  $\Phi$  and  $\Omega$  are the transfer functions of each classification system expressed by subscript. In the  
 167 following discussion, we explain the transfer functions of DMA-CPMA and DMA-AAC utilizing the  
 168 literature data of soot particles (Pei et al., 2018). The  $d_m$  and  $m$  of the representative particles are 100 nm and  
 169 0.33 fg, respectively, and the corresponding  $d_{ac}$  is 68.3 nm according to Eq. (19). The dimension of individual  
 170 classifier is summarized in Table 1.

### 171 DMA-CPMA

172 DMA-CPMA transfer function was calculated in the  $\log(d_m)$ - $\log(m)$  space, as shown in Fig. 2. In the  $\log(d_m)$ -  
 173  $\log(m)$  space, the mass-mobility relationship is

$$174 \quad m = \rho_f d_m^{D_{fm}}, \quad (22)$$

$$175 \quad \log(m) = D_{fm} \log(d_m) + \log(\rho_f), \quad (23)$$

176 In theory,  $D_{fm}$  equals to 3 for spherical particles and smaller than 3 for aspherical particles. In the  $\log(d_m)$ -  
 177  $\log(m)$  space, the relationship of  $m$  and  $d_m$  is linear with the slope expressed as mass-mobility exponent ( $D_{fm}$ )  
 178 and the intercept representing pre-exponential factor ( $\rho_f$ ). Under this specific operation condition, no overlap  
 179 was observed between spherical particles population (black line) and the classification region for doubly  
 180 charged particles, implying only the singly charged particles were selected. However, for aspherical particles  
 181 with  $D_{fm} < 3$ , such as soot particles with aggregate structure, the particles population may overlap the doubly  
 182 charged region when the slope ( $D_{fm}$ ) is small enough, however, the combination of DMA and CPMA is  
 183 generally used to avoid the multiple charge effect in soot studies. The reported  $D_{fm}$  values are typically in the  
 184 range of 2.2-2.4 for fresh soot particles (Rissler et al., 2013) and diesel soot particles (Park et al., 2003). In  
 185 the exemplary case, the derived  $D_{fm}$  of premixed flame generated soot particles was 2.28, resulting in the  
 186 particles population always goes through the transfer area of doubly charged particles. This implies that the  
 187 performance of DMA-CPMA to eliminate multiple-charged particles to a certain extent depends on the  
 188 particle morphology.

189 The DMA-CPMA system can eliminate the multiply charged particles only if the  $D_{fm}$  of particles is larger  
 190 than the slope of a line connecting  $(d_m, m) = (d_{m2, \min}, m_{2, \max})(d_{m1}, m_1)$  (as  $PP_0$  shown in Fig. 2). Since CPMA  
 191 is used downstream of the DMA, the value of mass limit can be expressed as follows according to Eq. (15)  
 192 assuming that all the classified particles have the same mobility.

$$193 \quad m_{n, \min}^{n, \max} = n \cdot m_1 \pm \frac{Q_{CPMA}}{2\pi B_{n, \min}^{n, \max} L_{CPMA} r_c^2 \omega^2}, \quad (24)$$

194 where  $m_{n, \min}^{n, \max}$  and  $B_{n, \min}^{n, \max}$  are the maximum and minimum particle mass and mechanical mobility which  
 195 would be selected by CPMA and DMA, respectively. The subscript  $n$  is charge quantity. Accordingly, the  
 196 ideal condition to completely eliminate the multiply charged particles is

$$197 \quad D_{fm} > PP_0 = \frac{\log(m_{2, \max}/m_1)}{\log(d_{m2, \min}/d_{m1})} = \frac{\log\left(2 + \frac{3Q_{CPMA}\mu d_{m1}}{(1-\beta_{DMA})L_{CPMA}r_c^2\omega^2 m_1 Cc(d_{m1})}\right)}{\log\left(\frac{2}{(1-\beta_{DMA})} \frac{Cc(d_{m2, \min})}{Cc(d_{m1})}\right)}, \quad (25)$$

198 The ability of DMA-CPMA to eliminate multiply charged particles depends on the selected  $d_m$ ,  $m$  and  
 199 resolutions of both DMA and CPMA. The Eq. (25) gives instructions in actual operation to eliminate  
 200 multiple-charged particles. When selecting particles of certain  $d_m$  and  $m$ , the smaller  $Q_{CPMA}$ , as well as larger



201  $\omega$  and  $\beta_{DMA}$  are necessary to reduce the potential of multiply charged particles. Thus, the key to evaluate  
202 whether there is multiple charging effect lies on particle morphology ( $D_{fm}$ ) and the slope of  $PP_0$  derived from  
203 the actual condition. Compared with DMA-CPMA, the selection of DMA-APM is more susceptible to  
204 multiple charging effect. According to the theoretical calculation described in Kuwata (2015), the slope of  
205  $PP_0$  of 3.55 was derived when DMA-APM selects the same example soot particles ( $d_m$  of 100 nm and  $m$  of  
206 0.33 fg) with  $D_{fm}$  of 2.28, indicating that the DMA-APM is more subjected to the multiple charging effect.  
207 Besides the instruments setups, particles morphology is also crucial for DMA-CPMA. Here we simulate the  
208 critical slope of  $PP_0$  when selecting different  $d_m$  and  $m$  under the common selecting conditions ( $\beta_{DMA} = 0.1$ ,  
209  $Q_{CPMA} = 0.3 \text{ L min}^{-1}$ ,  $R_m = 8$ ), which is shown in Fig. 3. Under this selecting conditions, DMA-CPMA can  
210 select monodispersed particles when the  $D_{fm}$  of particles is larger than the slope of  $PP_0$  which is represented  
211 as background color. When selecting small aspherical particles or particles with extremely low density, the  
212 slope of  $PP_0$  is relatively higher and DMA-CPMA classification is sensitive to multiple charging effect. As  
213 shown in Fig. 3, the  $d_m$ ,  $m$  and corresponding  $D_{fm}$  were taken from literature (Park et al., 2003; Rissler et al.,  
214 2013; Ait Ali Yahia et al., 2017; Dastanpour et al., 2017; Kazemimanesh et al., 2019). Generally, multiple  
215 charging effect can be avoided for DMA-CPMA to select soot particles with diameter larger than 200 nm.,  
216 while fails to eliminate multiply charged particles when selecting small soot particles. The potential  
217 uncertainties will be discussed in details with flame generated soot particles in Sect. 3.2.

#### 218 DMA-AAC

219 The advantage of AAC versus CPMA is there is no need for a neutralizer to charge aerosol particles.  
220 Therefore, the multiple charge effect could be avoided theoretically. The transfer function of DMA-AAC  
221 selecting the same representing particles was calculated and shown in  $\log(d_{ae}) - \log(d_m)$  (Fig. 4a). Moreover,  
222 according to Eqn. 19 and Eqn. 22, aspherical particles can be expressed as follows,

$$223 \log d_{ae} = \frac{1}{2}(D_{fm} - 1)\log d_m + \frac{1}{2}\log\left(\frac{6}{\pi} \frac{Cc(d_m)\rho_f}{Cc(d_{ae})\rho_0} \cdot 10^{9D_{fm}-18}\right), \quad (26)$$

224 which indicates that the relationship between  $d_{ae}$  and  $d_m$  is non-linear since the  $Cc(d_m)$  and  $Cc(d_{ae})$  varies with  
225  $d_m$  and  $d_{ae}$ , respectively. Particles morphology can be derived from the relationship between  $d_m$  and  $d_{ae}$   
226 measured by DMA and AAC, respectively. In order to simulate the transfer function of DMA-AAC selecting  
227 the same particles as that used in calculations of DMA-CPMA. The corresponding  $d_{ae}$  was numerically solved  
228 using known mass-size relationship. Unlike the DMA-CPMA system, the transfer functions of singly charged  
229 and doubly charged particles is in parallel for DMA-AAC, suggesting that particle population is less likely  
230 to overlap with the region of multiply charged particles. Using the example setups of DMA-AAC, truly  
231 monodispersed particles are selected for spherical particles and typical soot particles.

232 Similar to the DMA-CPMA system, to eliminate multiply charged particles requires the  $d_{ae,max}$  of the AAC  
233 at  $d_{m2,min}$  must be smaller than the  $d_{ae}$  of particles of interest which can be derived from  $d_{m2,min}$  and  $D_{fm}$  (Eqn.  
234 26),

$$235 d_{ae}(d_{m2,min}, D_{fm}) > d_{ae,max}(d_{m2,min}),$$



$$\Rightarrow D_{fm} > \frac{\log(2 \frac{1+\beta_{AAC}}{1+\beta_{DMA}})}{\log[\frac{2}{1+\beta_{DMA}} \frac{Cc(d_{m2,min})}{Cc(d_{m1})}]}, \quad (27)$$

237 This equation describes the minimum value of  $D_{fm}$  to eliminate multiple charging effect. It is clearly shown  
238 that the mobility resolution of DMA and the relaxation time resolution of AAC determine the limiting  
239 condition, and resolution of AAC is more important compared with resolution of DMA. The limiting  
240 condition is also related to selected  $d_m$  of DMA but independent of selected  $d_{ac}$  of AAC (Fig. S1). Setting the  
241 same resolutions of DMA and AAC, particle selection is more susceptible to multiple charging effect when  
242 selecting small sizes. In Fig. 4a, the values of  $\beta_{DMA}$  and  $\beta_{AAC}$  are 0.1, resulting in the minimum  $D_{fm}$  of 1.41,  
243 which is the case for most atmospheric aerosol particles. Hence, the selected particles of DMA-AAC are truly  
244 monodisperse regardless of particles morphology. However, in actual operations, larger sample flow rate is  
245 required to satisfy the apparatus downstream, while the maximum sheath flow rate of classifier is restricted  
246 by the instrument design (e.g., 30 L min<sup>-1</sup> for DMA and 15 L min<sup>-1</sup> for AAC). When increasing  $\beta_{AAC}$  to 0.3  
247 and remaining  $\beta_{DMA}$  unchanged, the transfer function becomes broader (Fig. 4b). The minimum  $D_{fm}$  is 2.44,  
248 which indicates that the multiple charging effect exists for typical soot particle with  $D_{fm}$  of 2.2-2.4. The line  
249 representing soot particles overlaps with the region of doubly charged particles. Thus, reducing resolutions  
250 of DMA or AAC is not suggested in actual operations.

### 251 3.2 Evaluation of the multiple charging effect

252 To quantify the possible uncertainties of multiple charging effect in DMA-CPMA system, we conducted the  
253 soot experiment as demonstrated in Fig. 1. For each mobility-selected particle, the corresponding  $d_{ac}$  and  $m$   
254 were determined using scan mode of AAC and CPMA, from which the effective densities were derived,  
255 respectively. The results are summarized in table 2. The fitted value of  $D_{fm}$  was 2.28, indicating a fractal  
256 structure, which is the same as the previous studies (Pei et al., 2018). The effective densities of generated  
257 soot particles vary from > 500 kg m<sup>-3</sup> at  $d_m = 80$  nm to <300 kg m<sup>-3</sup> at  $d_m$  of 250 nm for two methods. In  
258 general, the deviation monotonically decreases along with increasing particle size. The deviation is 7.65%  
259 for particles of 80 nm, whereas it decreased to <1% of particles larger than 200 nm. The results reveal a strict  
260 agreement between two methods for retrieving particle effective density.

261 According to Fig. 3, the critical slopes of PP<sub>0</sub> for soot particles with  $d_m$  of 80, 100, 150, 200 and 250 nm are  
262 2.46, 2.40, 2.29, 2.17 and 2.07, respectively. The measured  $D_{fm}$  of 2.28 is smaller than the calculated PP<sub>0</sub>  
263 for particles with  $d_m$  smaller than 150 nm, which suggested that multiply charged particles are still classified in  
264 this circumstance.

265 When selecting particles with  $d_m$  of 80 nm and  $m$  of 0.16 fg, the corresponding transfer function is shown in  
266 Fig. 5a. The particle population overlaps the transfer function region of doubly charged particles, suggesting  
267 the potential interferences of doubly charged particles in DMA-CPMA selection. Since the classification of  
268 AAC is insusceptible to particle charging states, the multiply charged particles can be resolved in  
269 aerodynamic size distribution. Fig. 5b shows the particles number aerodynamic size distribution (PNSD<sub>ac</sub>)  
270 scanned by AAC. PNSD<sub>ac</sub> was fitted using log-normal distributions and three peaks which correspond to





271 singly, doubly and triply charged particles were identified. The mean  $d_{ac}$  were 53.8, 58.2 and 69.1 nm, and  
272 the corresponding  $d_{ac}$  were calculated as 51.1, 61.2 and 69.4 nm using Eq. (1) and Eq. (30), respectively. The  
273 experimental results are consistent with the theoretical results with deviation within 5.3%.

274 On the contrary, when selecting particles with  $d_m$  of 200 nm and  $m$  of 1.28 fg, the transfer function is shown  
275 in Fig. 6a. The slope of  $PP_0$  of 2.07 is smaller than  $D_{fm}$  of 2.28, the generated particles population does not  
276 overlap with the block of doubly charged particles, thus DMA-CPMA classified particles were truly mono-  
277 dispersed.  $PNSD_{ac}$  measured by AAC is unimodal, implying that the classified particles were singly charged.  
278 The results of other experiments are shown in Fig. S2. Although the transfer function of DMA-CPMA  
279 showed that no multiply charged particles would be selected when classifying 150 nm particles (Fig. S2a),  
280 doubly charged particles were resolved by AAC (Fig. S2b). These doubly charged particles were selected  
281 probably owing to particles diffusion. The non-diffusion models were used to calculate the transfer function  
282 but actually the transfer function can be broader because of diffusion. In summary, for a type of particles  
283 (with the same mass-mobility relationship), the possibility of multiple charging increases for small particles  
284 when selected by DMA-CPMA system, which is consistent with the theoretical calculation in Sect. 3.1.

### 285 3.3 Atmospheric implication

286 The DMA-APM and DMA-CPMA system are usually adopted to eliminate multiply charged particles in soot  
287 aerosol studies. As previously discussed, although they might fail to select monodispersed particles,  
288 downstream measurements by instrument such as single particle soot photometer (SP2), will not be interfered  
289 which characterize the distinct information of single particle. Nevertheless, for techniques measuring  
290 properties of entire aerosol population, e.g., scattering coefficient by nephelometer or absorption coefficient  
291 by photoacoustic spectrometer, multiply charged particles can induce significant bias. Pervious study  
292 (Radney and Zangmeister, 2016) pointed out that DMA-APM failed to resolve multiply charged particles for  
293 soot particles when selecting 150 nm flame-generated, which caused 110% error in extinction measurement.  
294 In order to investigate the multiple charging effect for DMA-CPMA classification, the optical absorption  
295 coefficient of particles with different charging states after DMA-CPMA classification was calculated from  
296  $PNSD_{ac}$ . Mie theory was used to calculate the theoretical absorption coefficient at the wavelength of 550 nm.  
297 The refractive index used in the Mie code was  $1.95+0.79i$  (Bond and Bergstrom, 2006). The  $PNSD_{ac}$  for  
298 different charging state particles were converted to volume equivalent diameter size distributions ( $PNSD_{ve}$ )  
299 which were used in Mie theory to determine the absorption coefficient for particles with single, double and  
300 triple charging, respectively. The method to calculate  $PNSD_{ve}$  is described in S1. Then absorption cross-  
301 section was derived using the absorption coefficient and integral concentration for particles with different  
302 charging states, which denoted as  $\sigma_{abs}$ . For soot particles with diameter of 80 nm, the contributions of particles  
303 with different charging states are shown in Fig. 5c. Doubly charged particles only account for 29.6% of the  
304 total number concentration but provide the largest fractional contribution in the total absorption (53.1%).  
305 Also, small fraction (0.7%) of triply charged particles account for 1.9% of the absorption. As a result, the



306 mass absorption cross-section (MAC) was overestimated by 54.8% and the directive radiative force (DRF)  
307 was overestimated by 54.8%. DRF was calculated using previous global climate models (Bond et al., 2016).  
308 Huge amount of 70-90 nm soot particles was emitted from diesel engine (Wierzbicka et al., 2014), neglecting  
309 of the multiple charging effect on this size range will result in significant bias in estimation of radiative  
310 forcing of automobile emitted soot particles, which may lead to huge error in climate model.  
311 For soot particles with diameter  $< 200$  nm, the optical absorption contributions of particles with different  
312 charging state and the MAC overestimation are summarized in Table 3. The number fraction of doubly  
313 charged particles declines with the size of nominated particles, i.e. 53.1% and 34.8% for 80 and 100 nm  
314 particle, respectively, but only 9.2% for 150 nm particles. Accordingly, the MAC was largely overestimated  
315 for 80 and 100 nm particles (54.8% and 27.1%, respectively) but moderately overestimated for 150 nm  
316 particles (0.69%). To summarize, our results indicated that the combination of tandem classifiers is not  
317 sufficient to completely eliminate multiply charged particles when selecting small size flame-generated soot  
318 particle, which introduced severe bias for absorption measurement and led to overestimation of MAC, as  
319 a result, the DRF of soot particles was also overestimated.

#### 320 **4 Conclusion**

321 In this study, we demonstrate the transfer functions of DMA-CPMA and DMA-AAC and discuss their  
322 limitations to eliminate multiply charged particles. For aspherical particles, there is no guarantee that  
323 multiple-charging effect can be avoided in DMA-CPMA or DMA-AAC systems. Usually, DMA-AAC can  
324 select truly monodisperse particles but it can be suffered of particles with multiple charges when decreasing  
325 the resolutions of DMA and AAC. The ability of DMA-CPMA to eliminate multiple charging effect mainly  
326 depends on the particles morphology and the instrument resolutions. Under the same setups of DMA-CPMA,  
327 this tandem system is more sensitive to multiple charging effect with decreasing  $D_{\text{fm}}$  and decreasing  
328 nominated size of particles. DMA-CPMA failed to eliminate multiply charged particles when selecting soot  
329 particles with diameter  $< 150$  nm. Although doubly charged particles accounted for a small fraction of number  
330 concentration, they contributed most significantly to light absorption, which indicated that multiply charged  
331 particles can induce obvious contribution on light absorption and lead to overestimation of DRF for flame-  
332 generated soot particles.

333

334 *Code/Data availability.* Code/Data is available upon request.

335 *Author contributions.* ZW determined the main goal of this study. YS and XP designed the methods. YS  
336 carried them out and prepared the paper with contributions from all coauthors. YS, HL and JZ analyzed the  
337 optical data.

338 *Competing interests.* The authors declare that they have no conflict of interest.

339 *Acknowledgements.* We specially acknowledge useful comments and suggestions on MATLAB script of  
340 CPMA transfer function from Timothy A. Sipkens. The study was supported by the National Natural Science  
341 Foundation of China (grant No. 91844301 and 41805100).



342 **Appendix A**

343 **Table A1. Symbol used in this study**

$\mu$	Air viscosity
$\beta$	The ratio of flow rates of aerosol flow and sheath flow, $Q_a/Q_{sh}$
$\tau$	Relaxation time
$\omega_1$	Rotational speed of the inner electrode
$\omega_2$	Rotational speed of the outer electrode
$\hat{\omega}$	$\omega_1/\omega_2$
$\delta$	Half width of the gap between the two electrodes
$\Omega$	Transfer function
$\rho_0$	Standard density, which equals to $1\text{kg/m}^3$
$\tau$	Relaxation time
$\tau^*$	$\tau$ at the maximum of the transfer function
$\tilde{\tau}$	Dimensionless particle relaxation time, $\tilde{\tau} = \tau/\tau^*$
$\rho_{\text{eff}}$	Effective density
$\rho_f$	Mass-mobility pre-exponential factor
$\sigma_{\text{cal}}$	Absorption cross-section calculated with Mie theory
$\sigma_{\text{CAPS-ALB}}$	Absorption cross-section measured by CAPS-ALB
$B$	Mechanical mobility
$C_c(d_p)$	Cunningham slip correction factor
$c_r$	Particle migration velocity
$D_{\text{fm}}$	Mass-mobility exponent
$d_{\text{ae}}$	Aerodynamic equivalent diameter
$d_m$	Mobility equivalent diameter
$d_{\text{ve}}$	Volume-equivalence size
$e$	Elementary charge
$L$	Length of DMA, CPMA or AAC
$m$	Particle mass
$n$	Number of elementary charges on the particle
PNSD	Particle number size distribution
PNSD <sub>ae</sub>	Particle number aerodynamic size distribution
PNSD <sub>ve</sub>	Particle number volume-equivalence size distribution
$Q_a$	Sample flow rate
$Q_{sh}$	Sheath flow rate
$q$	Electrical charge on the particle
$R_m$	Mass resolution of CPMA



$r_a$	Lower initial radial position that passes through the classifier
$r_b$	Upper initial radial position that passes through the classifier
$r_1$	Inner radius
$r_2$	Outer radius
$\hat{r}$	$r_1 / r_2$
$t$	Time
$V$	Voltage between the two electrodes of DMA or CPMA
$\bar{v}$	Average flow velocity
$v_z$	Axial flow distribution
$v_\theta$	Velocity profile in the angular direction
$Z_p^*$	$Z_p$ at the maximum transfer function of DMA
$Z_p$	Electrical mobility
$\bar{Z}_p$	$Z_p / Z_p^*$

---

#### 344 **References**

- 345 Ait Ali Yahia, L., Gehin, E., and Sagot, B.: Application of the Thermophoretic Annular Precipitator (TRAP)  
346 for the study of soot aggregates morphological influence on their thermophoretic behaviour, *J. Aerosol Sci.*,  
347 113, 40-51, <https://doi.org/10.1016/j.jaerosci.2017.07.018>, 2017.
- 348 Biskos, G., Malinowski, A., Russell, L. M., Buseck, P. R., and Martin, S. T.: Nanosize Effect on the  
349 Deliquescence and the Efflorescence of Sodium Chloride Particles, *Aerosol Sci. Technol.*, 40, 97-106,  
350 <https://doi.org/10.1080/02786820500484396>, 2006.
- 351 Bond, T. C., and Bergstrom, R. W.: Light Absorption by Carbonaceous Particles: An Investigative Review,  
352 *Aerosol Sci. Technol.*, 40, 27-67, <https://doi.org/10.1080/02786820500421521>, 2006.
- 353 Bond, T. C., Doherty, S. J., Fahey, D. W., Forster, P. M., Berntsen, T., DeAngelo, B. J., Flanner, M. G., Ghan,  
354 S., Kärcher, B., Koch, D., Kinne, S., Kondo, Y., Quinn, P. K., Sarofim, M. C., Schultz, M. G., Schulz, M.,  
355 Venkataraman, C., Zhang, H., Zhang, S., Bellouin, N., Guttikunda, S. K., Hopke, P. K., Jacobson, M. Z.,  
356 Kaiser, J. W., Klimont, Z., Lohmann, U., Schwarz, J. P., Shindell, D., Storelvmo, T., Warren, S. G., and  
357 Zender, C. S.: Bounding the role of black carbon in the climate system: A scientific assessment, *J. Geophys.*  
358 *Res.: Atmos.: Atmospheres*, 118, 5380-5552, <https://doi.org/10.1002/jgrd.50171>, 2013.
- 359 Cheng, Y., Su, H., Koop, T., Mikhailov, E., and Pöschl, U.: Size dependence of phase transitions in aerosol  
360 nanoparticles, *Nat. Commun.*, 6, <https://doi.org/10.1038/ncomms6923>, 2015.
- 361 Dastanpour, R., Momenimovahed, A., Thomson, K., Olfert, J., and Rogak, S.: Variation of the optical  
362 properties of soot as a function of particle mass, *Carbon*, 124, 201-211,  
363 <https://doi.org/10.1016/j.carbon.2017.07.005>, 2017.



- 364 Dusek, U., Frank, G. P., Hildebrandt, L., Curtius, J., Schneider, J., Walter, S., Chand, D., Drewnick, F., Hings,  
365 S., Jung, D., Borrmann, S., and Andreae, M. O.: Size Matters More Than Chemistry for Cloud-Nucleating  
366 Ability of Aerosol Particles, *Science*, 312, 1375-1378, <https://doi.org/10.1126/science.1125261>, 2006.
- 367 Ehara, K., Hagwood, C., and Coakley, K. J.: Novel method to classify aerosol particles according to their  
368 mass-to-charge ratio—Aerosol particle mass analyser, *J. Aerosol Sci*, 27, 217-234,  
369 [https://doi.org/10.1016/0021-8502\(95\)00562-5](https://doi.org/10.1016/0021-8502(95)00562-5), 1996.
- 370 Johnson, T. J., Nishida, R., Irwin, M., Symonds, J. P. R., Olfert, J. S., Boies, A.: Agreement Between Different  
371 Aerosol Classifiers Using Spherical Particles, <https://doi.org/10.13140/RG.2.2.30999.27043>, 2018.
- 372 Johnson, T. J.; Nishida, R. T.; Zhang, X.; Symonds, J. P. R.; Olfert, J. S.; Boies, A. M., Generating an aerosol  
373 of homogeneous, non-spherical particles and measuring their bipolar charge distribution. *J. Aerosol Sci*, 153.  
374 <https://doi.org/10.1016/j.jaerosci.2020.105705>, 2021.
- 375 Johnson, T. J.; Olfert, J. S.; Cabot, R.; Treacy, C.; Yurteri, C. U.; Dickens, C.; McAughey, J.; Symonds, J. P.  
376 R., Steady-state measurement of the effective particle density of cigarette smoke. *J. Aerosol Sci*, 75, 9-16,  
377 <https://doi.org/10.1016/j.jaerosci.2014.04.006>, 2014.
- 378 Kazemimanesh, M., Dastanpour, R., Baldelli, A., Moallemi, A., Thomson, K. A., Jefferson, M. A., Johnson,  
379 M. R., Rogak, S. N., and Olfert, J. S.: Size, effective density, morphology, and nano-structure of soot particles  
380 generated from buoyant turbulent diffusion flames, *J. Aerosol Sci.*, 132, 22-31,  
381 <https://doi.org/10.1016/j.jaerosci.2019.03.005>, 2019.
- 382 Knutson, E. O., and Whitby, K. T.: Aerosol classification by electric mobility: apparatus, theory, and  
383 applications, *J. Aerosol Sci.*, 6, 443-451, [https://doi.org/10.1016/0021-8502\(75\)90060-9](https://doi.org/10.1016/0021-8502(75)90060-9), 1975.
- 384 Kuwata, M.: Particle Classification by the Tandem Differential Mobility Analyzer–Particle Mass Analyzer  
385 System, *Aerosol Sci. Technol.*, 49, 508-520, <https://doi.org/10.1080/02786826.2015.1045058>, 2015.
- 386 Moallemi, A.; Kazemimanesh, M.; Corbin, J. C.; Thomson, K.; Smallwood, G.; Olfert, J. S.; Lobo, P.,  
387 Characterization of black carbon particles generated by a propane-fueled miniature inverted soot generator.  
388 *J. Aerosol Sci.*, 135, 46-57, <https://doi.org/10.1016/j.jaerosci.2019.05.004>, 2019
- 389 Olfert, J. S., and Collings, N.: New method for particle mass classification—the Couette centrifugal particle  
390 mass analyzer, *J. Aerosol Sci.*, 36, 1338-1352, <https://doi.org/10.1016/j.jaerosci.2005.03.006>, 2005.
- 391 Park, K., Cao, F., And, D. B. K., and McMurry, P. H.: Relationship between Particle Mass and Mobility for  
392 Diesel Exhaust Particles, *Environ. Sci. & Technol.*, 37, 577-583, <https://doi.org/10.1021/es025960v>, 2003.
- 393 Pei, X., Hallquist, M., Eriksson, A. C., Pagels, J., Donahue, N. M., Mentel, T., Svenningsson, B., Brune, W.,  
394 and Pathak, R. K.: Morphological transformation of soot: investigation of microphysical processes during  
395 the condensation of sulfuric acid and limonene ozonolysis product vapors, *Atmos. Chem. Phys.*, 18, 9845-  
396 9860, <https://doi.org/10.5194/acp-18-9845-2018>, 2018.
- 397 Radney, J. G., Ma, X., Gillis, K. A., Zachariah, M. R., Hodges, J. T., and Zangmeister, C. D.: Direct  
398 Measurements of Mass-Specific Optical Cross Sections of Single-Component Aerosol Mixtures, *Anal.*  
399 *Chem.*, 85, 8319-8325, <https://doi.org/10.1021/ac401645y>, 2013.



- 400 Radney, J. G., and Zangmeister, C. D.: Practical limitations of aerosol separation by a tandem differential  
401 mobility analyzer–aerosol particle mass analyzer, *Aerosol Sci. Technol.*, 50, 160-172,  
402 <https://doi.org/10.1080/02786826.2015.1136733>, 2016.
- 403 Rissler, J., Messing, M. E., Malik, A. I., Nilsson, P. T., Nordin, E. Z., Bohgard, M., Sanati, M., and Pagels,  
404 J. H.: Effective Density Characterization of Soot Agglomerates from Various Sources and Comparison to  
405 Aggregation Theory, *Aerosol Sci. Technol.*, 47, 792-805, [10.1080/02786826.2013.791381](https://doi.org/10.1080/02786826.2013.791381), 2013.
- 406 Shiraiwa, M., Kondo, Y., Iwamoto, T., and Kita, K.: Amplification of Light Absorption of Black Carbon by  
407 Organic Coating, *Aerosol Sci. & Technol.*, 44, 46-54, <https://doi.org/10.1080/02786820903357686>, 2010.
- 408 Sipkens, T. A., Olfert, J. S., and Rogak, S. N.: New approaches to calculate the transfer function of particle  
409 mass analyzers, *Aerosol Sci. Technol.*, 54, 111-127, <https://doi.org/10.1080/02786826.2019.1680794>, 2019.
- 410 Stolzenburg, M. R., and McMurry, P. H.: Equations Governing Single and Tandem DMA Configurations  
411 and a New Lognormal Approximation to the Transfer Function, *Aerosol Sci. Technol.*, 42, 421-432,  
412 <https://doi.org/10.1080/02786820802157823>, 2008.
- 413 Tavakoli, F., and Olfert, J. S.: An Instrument for the Classification of Aerosols by Particle Relaxation Time:  
414 Theoretical Models of the Aerodynamic Aerosol Classifier, *Aerosol Sci. Technol.*, 47, 916-926,  
415 <https://doi.org/10.1080/02786826.2013.802761>, 2013.
- 416 Tavakoli, F., and Olfert, J. S.: Determination of particle mass, effective density, mass–mobility exponent,  
417 and dynamic shape factor using an aerodynamic aerosol classifier and a differential mobility analyzer in  
418 tandem, *J. Aerosol Sci.*, 75, 35-42, <https://doi.org/10.1016/j.jaerosci.2014.04.010>, 2014.
- 419 Zangmeister, C. D., You, R., Lunny, E. M., Jacobson, A. E., Okumura, M., Zachariah, M. R., and Radney, J.  
420 G.: Measured in-situ mass absorption spectra for nine forms of highly-absorbing carbonaceous aerosol,  
421 *Carbon*, 136, 85-93, <https://doi.org/10.1016/j.carbon.2018.04.057>, 2018.
- 422 Zhang, R., Khalizov, A. F., Pagels, J., Zhang, D., Xue, H., and McMurry, P. H.: Variability in morphology,  
423 hygroscopicity, and optical properties of soot aerosols during atmospheric processing, *Proc. Natl. Acad. Sci.*,  
424 105, 10291, <https://doi.org/10.1073/pnas.0804860105>, 2008.
- 425



426

**Table 1 Dimensions of the three classifiers used for transfer function calculation**

Parameter	DMA	CPMA	AAC
$r_1$ (mm)	9.37	100	43
$r_2$ (mm)	19.61	103	45
L (mm)	44.369	200	210
$\omega_2/\omega_1$	—	0.945	—

427

428

**Table 2. Mobility diameter, mass, aerodynamic diameter, effective densities calculated by DMA-AAC and DMA-CPMA, and the deviation between them for fresh soot particles in the size range of 80-250 nm.**

429

$d_m$ (nm)	$m$ (fg)	$d_{ac}$ (nm)	$\rho_{DMA-AAC}$ (kg m <sup>-3</sup> )	$\rho_{DMA-CPMA}$ (kg m <sup>-3</sup> )	Deviation
80	0.16	48	551.2	596.8	7.65%
100	0.27	55	488.0	515.7	5.38%
150	0.66	67	359.1	373.5	3.86%
200	1.28	82	303.2	305.6	0.77%
250	2.17	96	262.8	265.2	0.90%

430

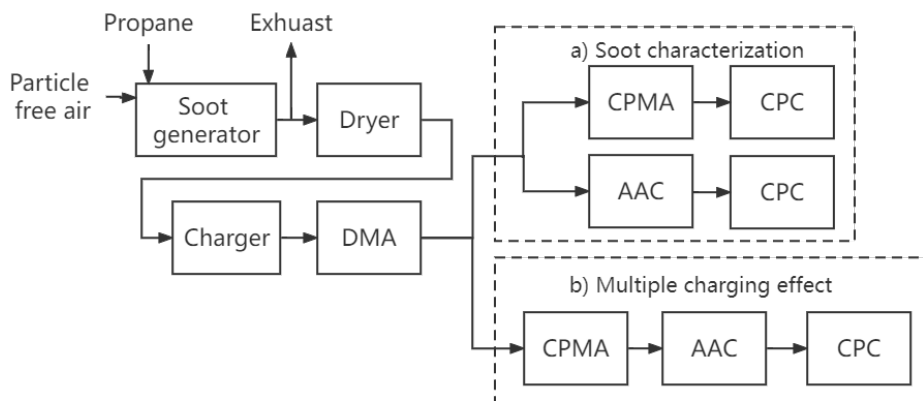
431

**Table 3. Number concentration fractions and absorption contributions for different size fresh soot particles with single, double or triple charges and the overestimation of MAC accordingly.**

432

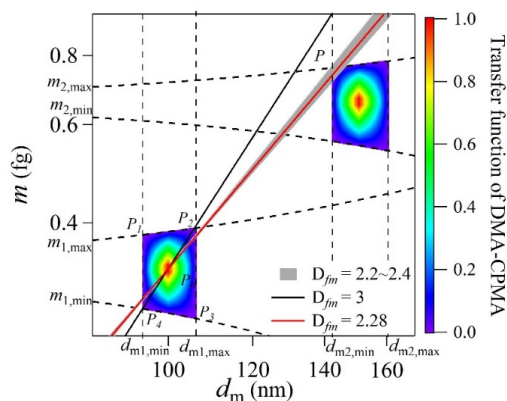
$d_m$ (nm)	singly charged particles		doubly charged particles		triply charged particles		MAC overestimation
	$f_N$	$f_{abs}$	$f_N$	$f_{abs}$	$f_N$	$f_{abs}$	
80	69.7%	45.0%	29.6%	53.1%	0.7%	1.9%	54.8 %
100	82.9%	65.2%	17.1%	34.8%	-	-	27.1 %
150	97.0%	90.8%	3.0%	9.2%	-	-	0.69 %

433



434  
 435  
 436

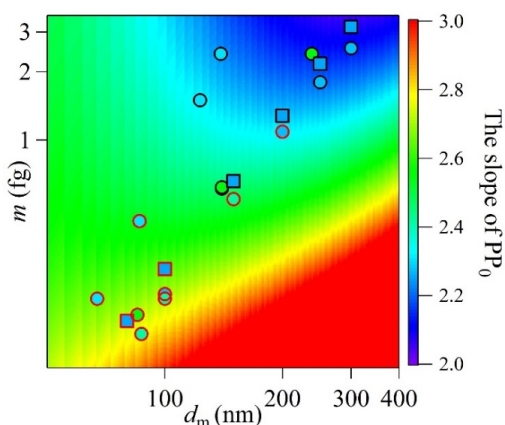
**Figure 1: Schematic of the experimental setup: (a) soot characterization and (b) evaluation of multiple charging effects.**



437  
 438  
 439  
 440  
 441  
 442  
 443  
 444

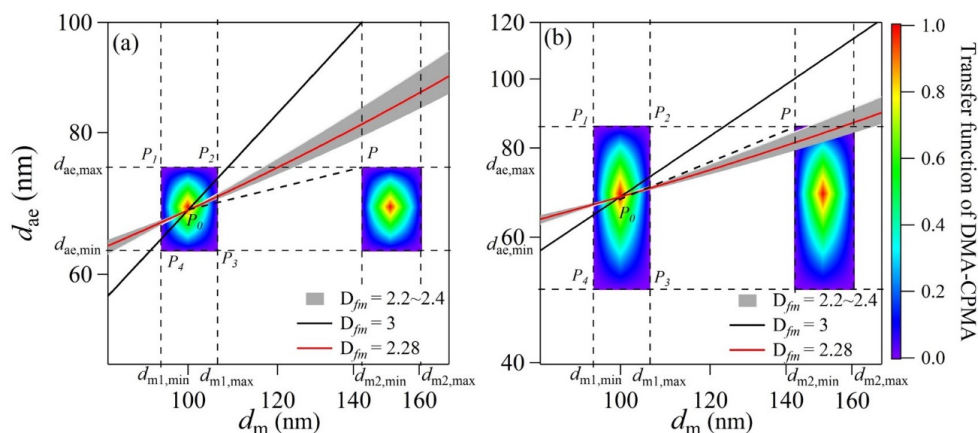
**Figure 2: Example of DMA-CPMA transfer function. The following parameter set was employed for the calculations:  $d_m = 100$  nm,  $\beta_{DMA} = 0.1$ ,  $m = 0.33$  fg,  $Q_{CPMA} = 0.3$  L min<sup>-1</sup>,  $R_m = 8$ . The color blocks are the transfer function of DMA-CPMA with the rainbow color representing the transfer function for singly charged (lower left block) and doubly charged (upper right block) particles. The black and red solid lines are particles population with  $D_{fm}$  of 3 and 2.28, respectively. The grey region is particle population with  $D_{fm}$  of 2.2-2.4, which is typical for soot aerosols. The dashed lines are the limits of  $d_m$  and  $m$  of DMA and CPMA. The DMA-CPMA transfer function for +2 particles does not overlap with the line for spherical particles with single charge ( $D_{fm}=3$ ).**





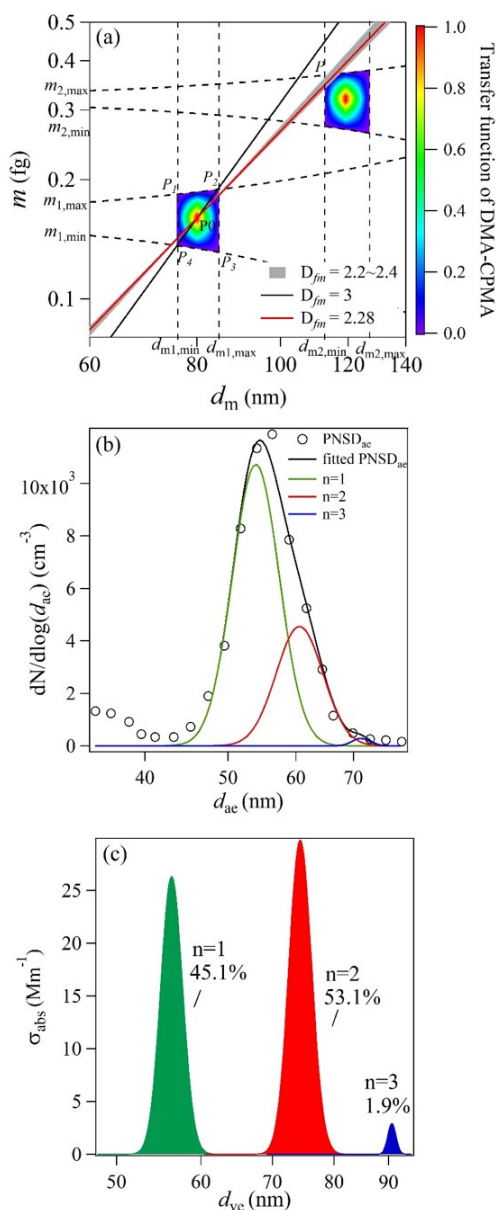
445

446 **Figure 3:** Variations of the slope of  $PP_0$  as a function of classified  $d_m$  and  $m$ . The following parameter set was  
 447 employed for the calculations:  $\beta_{DMA} = 0.1$ ,  $Q_{CPMA} = 0.3 \text{ L min}^{-1}$ ,  $R_m = 8$ . The background color coding denotes the  
 448 slope of  $PP_0$  with red represents the slope of  $PP_0 \geq 3$ . The circles and squares represent the reported  $D_{fm}$  values  
 449 from literatures (Park et al., 2003; Rissler et al., 2013; Ait Ali Yahia et al., 2017; Dastanpour et al., 2017;  
 450 Kazemimanesh et al., 2019) and measured soot particles in this study (See details in section 3.2), respectively.  
 451 Symbol colors indicate the particle  $D_{fm}$ . Symbols with red border correspond to the cases that potential multiple  
 452 charging effect may exist.



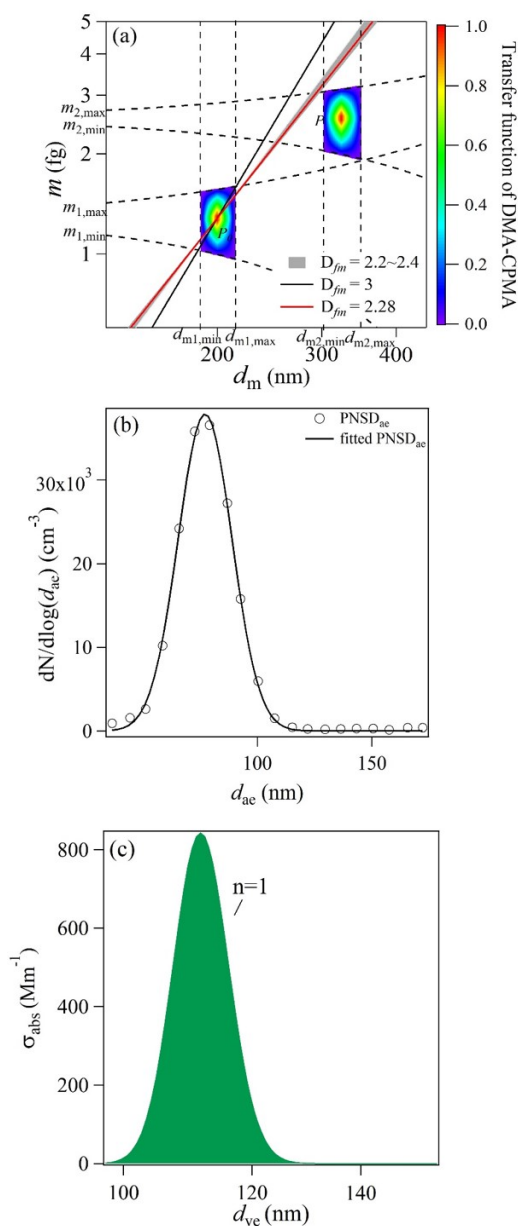
453

454 **Figure 4:** Example of transfer function calculation of DMA-AAC. The following parameter set was employed for  
 455 the calculations:  $Q_a = 0.3 \text{ L min}^{-1}$ ,  $d_{m1} = 100 \text{ nm}$ ,  $d_{ae} = 68.3 \text{ nm}$ , (a)  $\beta_{DMA} = 0.1$ ,  $\beta_{AAC} = 0.1$ , (b)  $\beta_{DMA} = 0.1$ ,  $\beta_{AAC} = 0.3$ .  
 456 The color blocks are the transfer functions of DMA-AAC. The black and red solid lines are particles population  
 457 with  $D_{fm}$  of 3 and 2.28, respectively. The grey region is particle population with  $D_{fm}$  of 2.2-2.4, which is typical for  
 458 soot aerosols. The black dashed lines are the limiting  $d_m$  and  $d_{ae}$  of DMA and AAC.



459

460 **Figure 5: (a) The transfer functions of DMA-CPMA when selecting 80 nm particles. The following parameter set**  
 461 **was employed for the calculations:  $d_{m1} = 80$  nm,  $\beta_{DMA} = 0.1$ ,  $m_1 = 0.16$  fg,  $Q_{CPMA} = 0.3$  L min<sup>-1</sup>,  $R_m = 8$ . The red solid**  
 462 **line is the generated soot particle population. (b) The aerodynamic size distribution of particles classified by DMA-**  
 463 **CPMA. The circles are data measured by AAC-CPC and the black, green red and blue lines are log-normal fitted**  
 464 **distributions of bulk, singly charged, doubly charged and triply charged particle population. (c) The contributions**  
 465 **to light absorption of particles with single, double and triple charges calculated with Mie theory.**



466

467 **Figure 6:** (a) The transfer functions of DMA-CPMA when selecting 200 nm particles. The following parameter set  
 468 was employed for the calculations:  $d_{m1} = 200$  nm,  $\beta_{DMA} = 0.1$ ,  $m_1 = 1.28$  fg,  $Q_{CPMA} = 0.3$  L min<sup>-1</sup>,  $R_m = 8$ . The red  
 469 solid line is the generated soot particle population. (b) the aerodynamic size distribution of particles classified by  
 470 DMA-CPMA. The circles are data measured by AAC-CPC and the solid line is log-normal fitted distribution.  
 471 (c) the contributions to light absorption of particles with single charge calculated with Mie theory.

472