

Response to reviewer#1

Thanks for the reviewer's helpful suggestions! The point-by-point responses are listed below.

*Comment: This study uses combined two most common instruments to achieve the characterization of particle size for BC-containing particles, in addition addressing the importance of evaluating the radiative forcing impacts of BC by introducing the particle size. This study is well structured but needs to consider the following points before publication.*

**Reply:** We thank the reviewer's comments.

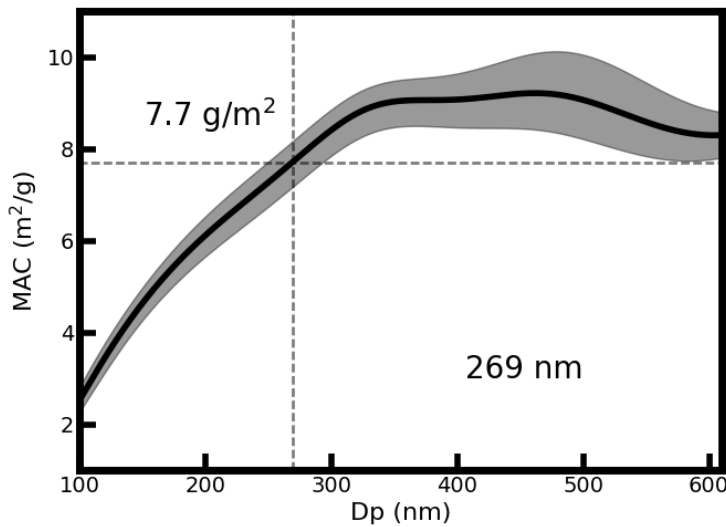
*Comment: 1. The observed two modes of the BCMSD in this study was the key conclusion, however the author should consider if the absorption measured by AE51 could be amplified due to the coating on BC particles at large size mode. As the aethalometer can only measure the absorption of the aerosol, it will bear large differences to convert the absorption of BC to BC mass at different size modes. The measured BCMSD will be also biased.*

**Reply:** We agree with the reviewer's valuable suggestion. Some revisions of deriving the BCMSD were made in the manuscript. In the manuscript, we first derived the BC absorption coefficient size distribution (BCASD), instead of the BCMSD. Then the BCASD were used to calculate the BCMSD using size-dependent mass absorption cross-section (MAC) value of BC.

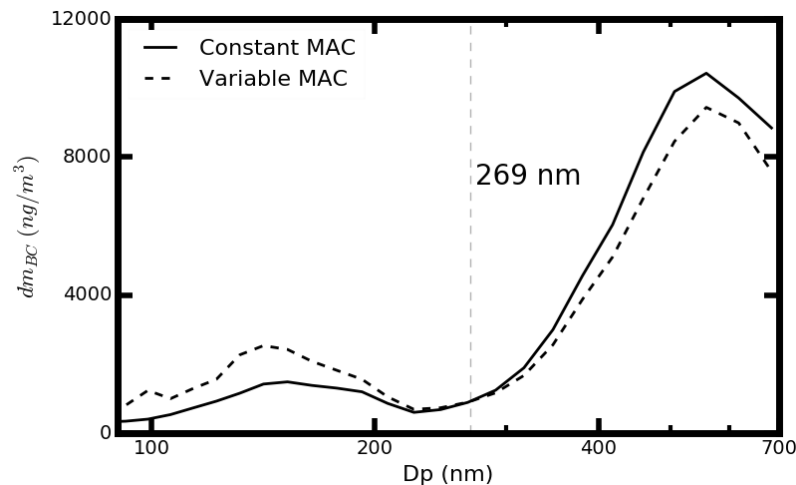
The size-resolved MAC was calculated using the Mie scattering model (Bohren and Huffman, 2007). Based on the Mie scattering theory, the MAC varies for different aerosol core diameter and different aerosol diameter. Results from SP2 measurement show that the size distribution of the BC core diameter peaked at around 120 nm in Beijing (Zhang et al., 2017). For each aerosol diameter in our study, the MAC value with core diameter of 120 nm was used to transform the BCASD into the BCMSD. MAC values with core diameter at  $120\pm 15$  nm were calculated and shown in Fig. R1. It varied significantly between 3.6 and 9.2 m<sup>2</sup>/g.

Fig. R2 gave one example of the measured BCMSD using a constant MAC and a size-resolved MAC respectively. From fig. R2, the BCMSD was larger when the diameter was lower than 269 nm and smaller when the diameter was larger than 269 nm respectively when compared with the BCMSD derived using a constant MAC.

Some revisions were made in the manuscript.



**Figure R1.** Calculated mass absorption coefficient of different aerosol diameter with assumption that the core diameter is  $120\pm 15$  nm



**Figure R2.** The retrieved mean BCMSD using a constant MAC of  $7.7 \text{ g/m}^2$  (solid line) and a variable MAC (dashed line) respectively.

*Comment: 2. The radiative forcing impacts due to introducing the BC size information is rather vague. As there is no direct measurement of vertical profile, how could SSA be so low in the upper level, which means there is a large fraction of BC, as shown in Fig. S6? It has not been demonstrated in the main text that how and why the BC size could influence the DARF results, due to the influence of asymmetry parameter? The DARF section needs a thorough revision I suggest.*

**Reply:** We thank the anonymous reviewer's comments and suggestions.

We added the multiple scattering correction of the measured light absorption coefficient in the revised manuscript. After the multiple scattering correction, the SSA was much larger than the previous value.

The BC size distribution influence the DARF results significantly. When the BCMSD was different for the same total BC mass concentration, the aerosol optical properties varied correspondingly. As noted in fig. 6 in the manuscript, the asymmetry factor, the scattering coefficient and absorption coefficient changed significantly. Therefore, the corresponding DARF were different due to different aerosol optical properties.

We added some discussions in the text. More descriptions of estimating the DARF were input in the manuscript.

*Comment: Other comments 1. p. 1, line 23: the author should consider the BC mixing state on influencing the absorption at large mode.*

**Reply:** Thanks for the comment. We have revised the text correspondingly.

*Comment: 2. p. 2, line 57: "and the BCMSD properties under different polluted conditions are not known yet", it was not correct since BCMSD have been measured using SP2 for years.*

**Reply:** We agree with the comment and deleted this sentence.

*Comment: 3. p. 2, line 58: BCMSD is not correct, the author should mention that the diameter measured by MOUDI, SP2 and DMA is different. The diameter for the MOUDI was aerodynamic diameter, and the DMA was the mobility diameter. BC diameter of the SP2 was the diameter of BC core. The value could change due to different density and shape factor. I strongly recommend the authors to rewrite this paragraph.*

**Reply:** Thanks for the comments. We rewrote this paragraph and added some descriptions in the text.

*Comment: 4. p. 3, line 72: there are some mistakes with the reference “(Xiaofeng Huang et al., 2006)”.*

**Reply:** Thanks for the comment. We have revised the reference.

*Comment: 5. p. 4, line 111: what does the “the ambient aerosol BCMSD” mean?*

**Reply:** Thanks for the comment. We rephrased this sentence into ‘ambient dry aerosol BCMSD corresponding to aerosol mobility diameter’.

*Comment: 6. p. 5, Sec 3.1.1: The authors just mention how to correct the “loading effect” of the aethalometer, but not mentioning the multiple scattering correction of the filter which might overestimate the mBC value.*

**Reply:** Thanks for the comments. We have revised the text correspondingly. As for the multiple scattering corrections, Zhang et al. (2018) compared the measured  $\sigma_{abs}$  measured by AE33 and by Multi-Angle Absorption Photometer (MAAP) at Tsinghua University, which was about 2 km away from our measurement site. They recommended a compensation factor of 2.6. We used the same factor for multiple scattering correction in our study.

*Comment: 7 p. 5, line 145: If the measurement of AE33 haven't been corrected for the multiple scattering effect, it should not be treated as a reference.*

**Reply:** Thanks for the comment. We performed the multiple scattering effect corrections of AE33 in our manuscript.

Bohren, C.F., Huffman, D.R., (2007) Absorption and Scattering by a Sphere, Absorption and Scattering of Light by Small Particles. Wiley-VCH Verlag GmbH, pp. 82-129.

Zhang, Y., Su, H., Kecorius, S., Wang, Z., Hu, M., Zhu, T., He, K., Wiedensohler, A., Zhang, Q., Cheng, Y. (2017) Mixing State of Refractory Black Carbon of the North China Plain

Regional Aerosol Combining a Single Particle Soot Photometer and a Volatility Tandem Differential Mobility Analyzer. Atmospheric Chemistry and Physics Discussions, 1-27.

Zhang, Y., Zhang, Q., Cheng, Y., Su, H., Li, H., Li, M., Zhang, X., Ding, A., He, K. (2018) Amplification of light absorption of black carbon associated with air pollution. Atmospheric Chemistry and Physics 18, 9879-9896.

# 1 **Role of black carbons mass size distribution in the direct aerosol radiative forcing**

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## 7 **Abstract**

8 Large uncertainties exist when estimating radiative effects of ambient black carbon (BC) aerosol.  
9 Previous studies about the BC aerosol radiative forcing mainly focus on the BC aerosols' mass  
10 concentrations and mixing states, while the effects of BC mass size distribution (BCMSD) were not  
11 well considered. In this paper, we developed a method by measuring the BCMSD by using a  
12 differential mobility analyzer in tandem with an aethalometer. A comprehensive method of multiple  
13 charging corrections was proposed and implemented in measuring the BCMSD. Good agreement  
14 was obtained between the BC mass concentration integrated from this system and that measured in  
15 bulk phase, demonstrating the reliability of our proposed method. Characteristics of the BCMSD and  
16 corresponding radiative effects were studied based on field measurements conducted in the North  
17 China Plain by using our own designed measurement system. Results showed that the BCMSD had  
18 two modes and the mean peak diameters of the two modes were 150 nm and 503 nm respectively.  
19 The BCMSD of coarser mode varied significantly under different pollution conditions with peak  
20 diameter varying between 430 nm and 580 nm, which gave rise to significant variation in aerosol  
21 buck optical properties. The aerosol direct aerosol radiative forcing was estimated to vary by  
22 ~~22.58.45~~% for different measured BCMSDs of coarser mode, which shared the same magnitude to  
23 the variation associated with assuming different aerosol mixing states (~~21.510.5~~%). Our study  
24 reveals that the BCMSD matters as well as their mixing state in estimating the direct aerosol  
25 radiative forcing. Knowledge of the BCMSD should be fully considered in climate models.

## 26 **1 Introduction**

27 Atmospheric black carbon (BC) is the second strongest absorbing components in atmosphere  
28 (Bond et al., 2013) but the magnitudes of the warming effects are poorly quantified. When emitted to  
29 the surrounding, BC particles transform the morphology from fractal to spherical and then grow as  
30 fully compact particles with other components depositing on the BC aerosol (Peng et al., 2016). The

31 variation in the shapes of BC aerosols, together with the variation in the mixing states, can lead to  
32 substantial change of aerosol optical properties (Liu et al., 2017;China et al., 2013;Wu et al.,  
33 2016a;Wu et al., 2018). BC aerosols also have significant influence on the climate by interacting  
34 with clouds (Koch and Del Genio, 2010;Roberts et al., 2008;Stevens and Feingold, 2009), ice and  
35 snow (Bond et al., 2013). Recent study shows that the solar absorption of BC can suppress the  
36 turbulence in the atmospheric boundary layer (Wilcox et al., 2016). It is found that BC emissions  
37 may be responsible for the incensement of droughts and floods in China and India (Menon et al.,  
38 2002). In addition, BC can pose a serve threat to human health through inhalation (Nichols et al.,  
39 2013;Janssen et al., 2011).

40 Comprehensive studies have been carried out to evaluate the climate effect of BC based on the  
41 measurement of BC mass concentrations ( $m_{BC}$ ) (Koch et al., 2009;Ramanathan and Carmichael,  
42 2008). The  $m_{BC}$  near the ground have been well characterized (Ramachandran and Rajesh,  
43 2007;Ran et al., 2016b;Reddington et al., 2013;Song et al., 2013), and the BC vertical distributions  
44 are widely measured and evaluated as well (Ran et al., 2016a;Babu et al., 2011;Ferrero et al., 2011).  
45 Despite these measurements, more insights into the BC microphysical properties can help to estimate  
46 the influence of BC aerosols on visibility (Zhang et al., 2008), climate (Jacobson, 2001) and human  
47 health (Lippmann and Albert, 1969). These microphysical properties include BC morphology (Zhang  
48 et al., 2016), density (Zhang et al., 2016), complex refractive index (Bond et al., 2013),  
49 hygroscopicity (Zhang et al., 2008;Peng et al., 2017),\_mixing states (Moffet et al., 2016;Raatikainen  
50 et al., 2017), and particularly, the mass size distribution (BCMSD) (Cheng et al., 2012;Cheng and  
51 Yang, 2016;Gong et al., 2016). Knowledge of BCMSD is not only helpful to study the mixing state  
52 of BC aerosols (Raatikainen et al., 2017), but also essential to model the role of BC in evaluating  
53 regional and global climate accurately-. BC radiative effects is highly sensitive to the emitted BC  
54 particle size distribution (Matsui et al., 2018). The health impacts of BC are significantly related to  
55 BCMSD (Turner et al., 2015). Furthermore, the information of BCMSD can help to study the source,  
56 the evolution and the mixing state of ambient BC aerosols (Yu et al., 2010).~~However, few studies~~  
57 ~~have focused on the characteristics of the BCMSD, and the BCMSD properties under different~~  
58 ~~polluted conditions are not known yet.~~

59 Many methods have been proposed to measure the BCMSD. For instance, the BCMSD was  
60 measured by sampling the aerosol in the size range from about 50 nm to several micrometers onto

61 quartz fiber filter substrates using a micro-orifice uniform deposit impactor (MOUDI)  
62 (Venkataraman and Friedlander, 1994;Guo, 2016). Cheng et al. (2014) developed a method to  
63 measure the BCMSD by employing two aethalometers in parallel, with one to measure total  $m_{BC}$   
64 and the other to measure  $m_{BC}$  below specific particle sizes using a size cut-off inlet. The above two  
65 methods measure the BCMSD corresponding to the aerodynamic diameter. The Single Particle Soot  
66 Photometer (SP2) is developed and widely used because it provides single particle information, hence  
67 the BCMSD and the mixing state of the atmospheric aerosols can be derived directly (Schwarz et al.,  
68 2006;Gao et al., 2007;Huang et al., 2012;Singh et al., 2016). -The BCMSD corresponding to the  
69 ambient aerosol mobility diameter can be measured by using a differential mobility analyzer (DMA)  
70 in tandem with SP2 (Raatikainen et al., 2017). However, the laser-induced incandescence method  
71 cannot provide reliable information about the particles beyond the range of 70 nm and 400 nm  
72 (Moteki and Kondo, 2010), which results in the lack of the knowledge of the BCMSD characteristics  
73 for these aerosols over 400 nm. The results from MOUDI find that a great amount of BC locates at  
74 the aerodynamic diameter range of larger-between than-370 and 1000 nm (Hu et al., 2012;Huang and  
75 Yu, 2008). However, the measurements of MOUDI cannot give detailed information of the BCMSD  
76 evolution due to the low temporal and diameter resolution (Hu et al., 2012;Huang and Yu, 2008). The  
77 characteristics of the BCMSD larger than 370 nm is not well studied due to the limitation of the  
78 instrument.

79 Recently, Ning et al. (2013) and Stabile et al. (2012) proposed a new method to measure the  
80 BCMSD by using differential mobility analyzer (DMA) in tandem with Aethalometer (AE). This  
81 method has the potential of measuring the BCMSD from 20 nm to 584 nm with high time resolution.  
82 We develop and validate the BCMSD measurement system based on the works of Ning et al. (2013).  
83 The developed measurement system was employed in a field campaign in the North China Plain. The  
84 characteristics of the measured BCMSD were studied based on the field measurement. Furthermore,  
85 the effects of BCMSD variations on the aerosol optical properties and corresponding direct aerosol  
86 radiative properties were evaluated. The aerosol optical properties were calculated by using the Mie  
87 scattering theory. The direct aerosol radiative forcing (DARF) were estimated by using the Santa  
88 Barbara DISORT (discrete ordinates radiative transfer) Atmospheric Radiative Transfer (SBDART)  
89 model.

90 The structure of this paper are organized as follows. Section 2 gives the information about the



91 instrument setup and field measurement. Section 3 gives the detailed method used in this study,  
92 which contains: 1, conducting multiple charging corrections when deriving the aerosol BCMSD and  
93 2, evaluating the aerosol optical and radiative properties for different BCMSD. Results and  
94 discussions are shown in section 4. The conclusion is drawn in the last part.

## 95 **2 Instrument Setup**

96 The measurement system setup was based on the works of Stabile et al. (2012) and Ning et al.  
97 (2013) as schematically shown in Fig.1. The ambient sample aerosol particles were firstly dried to  
98 below relative humidity of 30% through a Nafion drying tube before passing through to the DMA  
99 (Model 3081, TSI, USA). The DMA scanned aerosol particles with diameter ranges from 12.3 to 697  
100 nm over a period of 285 seconds and started another scanning after a pause of 15 seconds, so one  
101 complete cycle took 5 minutes. The sheath and sample flow rates of the DMA were 3 lpm and 0.5  
102 lpm, respectively. The quasi-monodisperse aerosols that passed through the DMA were further  
103 divided into two flows: with one lead to an aethalometer (AE51, Model 51, MicroAeth, USA) with a  
104 flow rate of 0.2 lpm to measure the  $\sigma_{abs}$  at 1 second time resolution; and  
105 the other one with flow rate of 0.3 lpm flow directed to a CPC (Model 3772, TSI, USA), which  
106 counted particle number concentrations at 0.1 second resolution. Clean air with a flow rate of 0.7  
107 lpm was used to compensate for the CPC inlet flow, which had default flow rate of 1 lpm. Overall,  
108 the combination system of DMA, CPC and AE51 could provide one PNSD and ~~BCMSD~~  
109 size-resolved  $\sigma_{abs}$  scan every 5 minutes. If the mass absorption coefficient (MAC) at a given  
110 diameter is known, the BCMSD can be derived correspondingly.

111 ~~At the same time, another~~ aethalometer (AE33, Model 33, Magee, USA) was used to measure  
112 the  $\sigma_{abs}$  or  $m_{BC}$  with a time resolution of 1 minute. The mass concentration of particles with  
113 diameter smaller than 2.5  $\mu\text{m}$  (PM<sub>2.5</sub>) was concurrently measured with time resolution of 1 minute  
114 during the filed observations by the Tapered Element Oscillating Microbalance (TEOM)  
115 Dichotomous Ambient Particulate Monitor (1405-DF), which was an indicator of the pollution  
116 conditions.

117 From 21 March to 9 April in 2017, an intensive field measurement was conducted to  
118 characterize of the ambient dry aerosol BCMSD corresponding to aerosol mobility diameter at the  
119 AERONET BEIJING\_PKU station (N39°59', E116°18'). This station was located on one roof of  
120 Peking University campus in the north west of Beijing, China. There were two main streets, Chengfu

121 Road to the south and Zhongguancun Street to the west that surrounding the station. The aerosol  
122 sampled at this station were mainly composed of urban roadside aerosols (Zhao et al., 2018).

### 123 3 Methodologies

#### 124 3.1 Retrieving the BCMSD

125 ~~Four~~ Five steps were involved to calculate the BCMSD using the raw data from the  
126 measurement system: 1), correcting the ‘loading effect’ and ‘multiple scattering effect’ of  $\sigma_{abs}m_{BC}$   
127 measured by AE51; 2), matching the instrument time between the AE51 and CPC; 3), matching the  
128 measured  $\sigma_{abs}m_{BC}$  and diameter to get the raw BCMSDsize-resolve  $\sigma_{abs}$  that is not involved in  
129 multiple charging corrections; 4), conducting the multiple charging corrections of the measured raw  
130 BCMSDsize-resolved  $\sigma_{abs}$ ; 5), transforming the size-resolved  $\sigma_{abs}$  into BCMSD.

##### 131 3.1.1 Obtaining the raw BCMSDsize-resolved $\sigma_{abs}$

132 The aethalometer (AE51 and AE33) is a well-developed and widely used instrument to measure  
133 the  $\sigma_{abs}m_{BC}$  (Drinovec et al., 2015; Hansen et al., 1984). When absorbing aerosols accumulates on  
134 the sample filter of the aethalometer continuously, the  $\sigma_{abs}m_{BC}$  can be determined by concurrently  
135 measuring the light intensities  $I$  after the fiber filter and the light intensities  $I_0$  transmitted through  
136 reference spot which is free of aerosol loading. The light attenuation (ATN) is defined as:

$$137 \quad \text{ATN} = 100 \cdot \ln\left(\frac{I_0}{I}\right). \quad (1)$$

138 The total  $\sigma_{abs}$  mass of BC of the loaded particle on the filter is given by:

$$139 \quad \sigma_{abs,tot}m_{BC,tot} = \frac{A \cdot \text{ATN}}{100 \cdot \sigma_{BC}}, \quad (2)$$

140 where A is the sample spot area on the filter and  $\sigma_{BC}$  is the mass attenuation cross-section of BC.

141 The instantaneous  $\sigma_{abs}$  can be calculated through The equivalent  $m_{BC}$  can be calculated through  
142 the increment of  $\sigma_{abs,tot}m_{BC,tot}$ :

$$143 \quad \sigma_{abs}m_{BC} = \frac{\sigma_{abs,tot}m_{BC,tot}}{\Delta t} = \frac{A \cdot \Delta \text{ATN}}{100 \cdot \sigma_{BC} \cdot F \cdot \Delta t}, \quad (3)$$

144 where F is the flow rate and  $\Delta \text{ATN}$  is the ATN variation during the time period of  $\Delta t$ . The  $\sigma_{abs,tot}$   
145 can be transformed to  $m_{BC}$  when the mass attenuation cross-section (MAC) of BC is known.  
146 Traditionally, a constant MAC at 7.7 g/m<sup>2</sup> was used to deduce the  $m_{BC}$  (Drinovec et al., 2015).

147 Corrections of the measured  $\sigma_{abs}$  are necessary because the systematic bias exists due to the  
148 prevalingly known ‘loading effect’ and multiple scattering effect (Drinovec et al., 2015; Virkkula et  
149 al., 2015; Virkkula et al., 2007). The AE33 can directly provide the corrected  $\sigma_{abs}m_{BC}$  values

150 through measuring two light intensities of two spots with different BC load efficiencies (Drinovec et  
151 al., 2015). For AE51, The correcting method in Virkkula et al. (2007) was adopted:

$$152 \quad \sigma_{abs, corrected} \overline{m_{BC, corrected}} = (1 + k \times ATN) \sigma_{abs, uncorrected} \overline{m_{BC, uncorrected}},$$

153 (4)

154 where k is the correction factor and a constant value of 0.004 is employed in this study to correct  
155 the  $\sigma_{abs} \overline{m_{BC}}$  from AE51. In the first part of the supplementary material, we showed that the loading  
156 effects corrections of  $\sigma_{abs} \overline{m_{BC}}$  from AE51 were essential and the value of  $\sigma_{abs} \overline{m_{BC}}$  from AE33  
157 could be used as a reference for the measured BCMSD. As for the multiple scattering corrections,  
158 Zhang et al. (2018) compared the measured  $\sigma_{abs}$  measured by AE33 and by Multi-Angle  
159 Absorption Photometer (MAAP) at Tsinghua University, which is about 2 km away from our  
160 measurement site. They recommended a compensation factor of 2.6 to be used and we adopted the  
161 same factor in our study.

162 Time correction was needed because time gaps between voltages implied on the DMA (particle  
163 size) and sample particles measured by different instruments were not the same. The time correction  
164 procedures were conducted every day during the field measurement to ensure that the time deviations  
165 of the CPC and the AE51 were constrained within 2 seconds.

166 Fig. S3 gave the time series diagram of scanned aerosol diameters by DMA, measured  $\sigma_{abs} \overline{m_{BC}}$   
167 from AE51, and the aerosol number concentrations counted by CPC. The aerosol PNSD (or  
168 BCMSD-size-resolved  $\sigma_{abs}$ ) could be calculated by matching the DMA diameter and the measured  
169 aerosol number concentrations (or measured  $\sigma_{abs} \overline{m_{BC}}$ ) by simply using the single particle charge  
170 ratio for each electrical mobility diameter. These measured PNSD and BCMSD-size-resolved  $\sigma_{abs}$   
171 did not consider the effect of multiple-charging corrections and are labeled as raw aerosol PNSD and  
172 raw aerosol size-resolved  $\sigma_{abs}$ , BCMSD-

### 173 **3.1.2 Multiple charging corrections of raw size-resolved $\sigma_{abs}$ , BCMSD**

174 In the work of Ning et al. (2013) study, lots of efforts were made to evaluate the performance of  
175 the instrument. They considered the diffusion corrections and particle charging corrections. However,  
176 the particle charging corrections were limited to single particle charge ratio as they mentioned that  
177 they simplified the particle charge correction by applying the peak electrical mobility for the  
178 calculation of representative particle size for each mobility bin and single particle charge ratio for

179 each primary mobility. They ignored the fact that the aerosol samples selected by the DMA were  
 180 quasi-monodisperse with different charges and different diameters.

181 We proposed a new algorithm for the multi-charge corrections of the BCMSDsize-resolved  $\sigma_{abs}$ .  
 182 Multi-charge corrections to the measured size distribution were prevailing when the DMA was used  
 183 to scan the aerosol sizes. When the DMA and CPC are used together to measure the aerosol particle  
 184 number size distribution (PNSD), the multi-charging corrected aerosol PNSD can be significantly  
 185 different from the raw measured one (Bau et al., 2014;He and Dhaniyala, 2013;He et al., 2015). As  
 186 shown in the results part of this study, the multi-charge corrections of the BCMSDsize-resolved  
 187  $\sigma_{abs}$  could cause differences in both the magnitude and shape of the BCMSDsize-resolved  $\sigma_{abs}$ .  
 188 Therefore, it is necessary to perform multi-charge corrections on the size-resolved  $\sigma_{abs}$  BCMSD.  
 189 This study developed a new algorithm to correct the size-resolved  $\sigma_{abs}$  from measured  
 190 value from measured  $m_{BC}$  based on the work of Hagen and Alofs (2007) and Deng et al. (2011).

191 When the DMA is charged with a negative voltage, those aerosols with a small range of  
 192 electrical mobility ( $Z_p$ ) can pass through the DMA:

$$193 \quad Z_p = \frac{q_{sh}}{2\pi VL} \ln\left(\frac{r_1}{r_2}\right), \quad (5)$$

194 where  $q_{sh}$  is the sheath air flow rate; V is the average voltage on the inner center rod;  $r_1$  and  $r_2$   
 195 are the outer and inner radius of annular space respectively. The  $Z_p$  is related with  $D_p$  by  
 196 elementary charge ( $e$ ), number of elementary charges on the particle ( $n$ ), and gas viscosity poise ( $\mu$ )  
 197 with:

$$198 \quad Z_p = \frac{neC(D_p)}{3\pi\mu D_p}, \quad (6)$$

199 where  $C(D_p)$  is Cunningham slip correction:

$$200 \quad C = 1 + \frac{2\tau}{D_p} \left(1.142 + 0.558e^{-\frac{0.999D_p}{2\tau}}\right), \quad (7)$$

201 where  $\tau$  is the gas mean free path. From equation 7, aerosol particles can have the same  $Z_p$  despite  
 202 that they have different  $n$  and  $D_p$ . At the same time, there exists a relatively larger portion of  
 203 multiple charged particles for those particles with diameters between 100 nm and 400 nm when the  
 204 ambient aerosols pass through the X-ray (Tigges et al., 2015;Wiedensohler and Fissan, 1988).  
 205 Through the above discussion, the selected aerosols by DMA at a given electrical mobility can have  
 206 different charges which will correspond to different diameters.

207 When the scan diameter is set as  $Dp_i$  for the singly charged particles and the respective voltage

208 of DMA is  $V_i$  ( $i=1, 2, \dots, I$ ), aerosol particles with electro-mobility of  $Z_{p,i}$  ( $i=1, 2, \dots, I$ ) can pass  
 209 through the DMA and the observed  $\sigma_{abs}$  by AE51 can be expressed as:

$$210 \quad R_i = \int_0^\infty G(i, x)A(x)n(x)dx, \quad (8)$$

211 where  $x$  is the scale parameter, with the definition of  $x = \log(Dp_i)$ ,  $A(x)$  is the average  
 212  $\sigma_{abs}$  of single particles for scale parameter  $x$ , and  $n(x) = dN/d\log Dp$  is aerosol  
 213 PNSD that is the multiple charging corrected results from the measured aerosol PNSD. We define the  
 214 kernel function  $G(i, x)$ , which is crucial to the algorithm, as:

$$215 \quad G(i, x) = \sum_{v=1}^\infty \phi(x, v)\Omega(x, v, i), \quad (9)$$

216 where  $\phi(x, v)$  is the probability of particles that are charged with  $v$  charges at the scale parameter  
 217 of  $x$  (Wiedensohler, 1988).  $\Omega(x, v, i)$  is the probability of particles that can pass through the DMA  
 218 with  $v$  charges at the scale parameter  $x$  (Knutson and Whitby, 1975). In this study, the maximum  
 219 value of  $v$  is 10.

220 The multiple charging corrections can be expressed as computing the  $A(x_i^*)$ , in which  $x_i^*$  is the  
 221 predetermined scale parameter from the DMA. To get the numerical integration results of equation 9,  
 222 the diameter interval that is 1/50 of the measured diameter is used. Thus, equation 9 can be written as

$$223 \quad R_i = \int_0^\infty G(i, x)A(x)n(x)dx = \Delta x_i \sum_{j=1}^{50} \beta_j G(i, x_{i,j})A(x_{i,j})n(x_{i,j}), \quad (10)$$

224 where  $\beta = \begin{cases} 0.5, & j = 1, J \\ 1, & \text{otherwise} \end{cases}$ ;  $x_{i,j}$  is the  $j^{\text{th}}$  ( $j=1, 2, \dots, 50$ ) parameter that locates at the parameter  $x_i$  and  
 225  $x_{i+1}$  and  $A(x_{i,j})$  ( $i=1, 2, \dots, I; j=1, 2, \dots, 50$ ), the BC mass ratio at scale parameter  $x_{i,j}$ , is expressed  
 226 as the linear interpolation of the values at the measured diameters.

$$227 \quad A(x_{i,j}) = A(x_i) + P_i(x_{i,j} - x_i), \quad (11)$$

228 where  $P_i$  is the slope of the linear interpolation result of

$$229 \quad A(x_k^*) = C + P_i \cdot x_k^*. \quad (12)$$

230  $x_k^*$  refers to these five diameters that are nearest to the predetermined scale parameter  $x_i$ .  $C$  is the  
 231 intercept of the linear interpolation result.

232 With  $H_{i,j} = \beta_j \Delta x_i G(i, x_{i,j})n(x_{i,j})$ , equation 11 can be written as

$$233 \quad R_i = \sum_{j=1}^J H_{ij} [A(x_i) + P_i(x_{i,j} - x_i)] = \sum_{j=1}^J H_{ij} A(x_i) + \sum_{j=1}^J H_{ij} P_i x_{i,j} - \sum_{j=1}^J H_{ij} P_i x_i$$

$$234 \quad = \sum_{k=1}^I (\sum_{j=1}^J H_{ij} \delta(i - k)) A(x_k^*) + \sum_{k=1}^I (\sum_{j=1}^J H_{ij} x_{i,j} \delta(i - k)) P_k - \sum_{k=1}^I (\sum_{j=1}^J H_{ij} \delta(i - k)) P_k x_k^*$$

235 
$$= \sum_{k=1}^I Q_{ik} A(x_k^*) + \sum_{k=1}^I T_{ik} P_k - \sum_{k=1}^I Q_{ik} P_k x_k^*, \quad (13)$$

236 where  $\delta(x) = \begin{cases} 0, & x \neq 0 \\ 1, & x = 0 \end{cases}$ ,

237 
$$Q_{ik} = \sum_{j=1}^J H_{ij} \delta(i - k), \quad (14)$$

238 and  $T_{ik} = \sum_{j=1}^J H_{ij} x_{i,j} \delta(i - k). \quad (15)$

239 By letting the

240 
$$S_i = R_i - \sum_{k=1}^I T_{ik} P_k + \sum_{k=1}^I Q_{ik} P_k x_k^*. \quad (16)$$

241 This equation is then expressed as

242 
$$S_i = \sum_{k=1}^I Q_{ik} A(x_k^*), \quad (17)$$

243 or

244 
$$S = QA, \quad (18)$$

245 where S and A are  $I \times 1$  vectors and Q is an  $I \times I$  matrix. This matrix can be solved by using the  
246 non-negative least square method.

247 Finally, the A(x) can be determined and the corresponding size-resolved  $\sigma_{abs}$  BCMSD that is  
248 multiple charging corrected can be calculated.

### 249 3.1.3 Transform the size-resolved $\sigma_{abs}$ into BCMSD

250 MAC of different size range is necessary when transform the size-resolved  $\sigma_{abs}$  into BCMSD.  
251 The MAC at different size should be different. When the size-resolved  $\sigma_{abs}$  is converted into  
252 BCMSD with a constant MAC, the derived BCMSD would be biased.

253 The size-resolved MAC was calculated using the Mie scattering model (Bohren and Huffman,  
254 2007). Based on the Mie scattering theory, MAC values vary for different aerosol core diameter and  
255 different total diameter. Results from SP2 measurement show that the size distribution of the BC  
256 core diameter peaked at around 120 nm in Beijing (Zhang et al., 2017). For each aerosol diameter,  
257 the MAC value with core diameter of 120 nm was used to transform the BCASD into the BCMSD.  
258 MAC values with core diameter at 120±15 nm were calculated and shown in Fig. 2. From Fig. 2, the  
259 MAC varied significantly between 3.6 and 9.2 m<sup>2</sup>/g. The constant MAC values 7.7 m<sup>2</sup>/g  
260 corresponded to the aerosol diameter of 269 nm. The calculated mean MAC values in Fig. 2 under  
261 different diameter were used in this study.

### 262 3.1.3-4 Validation of the multiple charging corrections

263 An example of the multiple charging corrections of the size-resolved  $\sigma_{abs}$  was shown in Fig. 23.

264 The corrections of aerosol PNSD were based on the work of Hagen and Alofs (2007). As shown in  
 265 Fig. 2(a), the corrected aerosol PNSD was significantly different from the original uncorrected one.  
 266 There were about half of the measured particles have multiple elementary charge in the size range  
 267 between 100 and 200 nm. The raw uncorrected aerosol PNSD had a peak value of  $10920 \text{ cm}^{-3}$  at 98  
 268 nm while the corrected aerosol PNSD reached its peak value of  $8450 \text{ cm}^{-3}$  at 98 nm. The peak  
 269 positions of the raw aerosol particle mass size distribution (PMSD,  $\text{dm}/\text{dlogDp}$ ) peaked at ~~371nm~~  
 270 322 nm with a peak value of ~~56-86.3~~  $\mu\text{g}/\text{m}^3$  and the corrected aerosol PMSD had a peak value of 53  
 271  $\mu\text{g}/\text{m}^3$  at ~~445-461~~ nm. The peak position of the aerosol PMSD shifted a lot before and after the  
 272 multiple charging corrections. The similar case for the ~~BCMSD-size-resolved  $\sigma_{abs}$~~  was shown in  
 273 Fig. 2(b). The shape of ~~size-resolved  $\sigma_{abs}$ BCMSD~~ had changed substantially due to the multiple  
 274 charging corrections. The measured raw BCMSD had a peak diameter near ~~32000~~ nm and the  
 275 magnitude of ~~size-resolved  $\sigma_{abs}$ BCMSD~~ plateau reached ~~6000  $\text{ng}/\text{m}^3$  at 283nm~~ 34.3  $\text{Mm}^{-1}$ , which  
 276 was in accordance with the results of Ning et al. (2013), where the multiple charging corrections  
 277 were not involved. However, the corrected ~~size-resolved  $\sigma_{abs}$ BCMSD~~ peaks near ~~400nm~~ 410 nm,  
 278 with a peak value of about ~~5500  $\text{ng}/\text{m}^3$  at 407nm~~ 29.5  $\text{Mm}^{-1}$ . According to the result, a small amount  
 279 of  ~~$\sigma_{abs}$ BC~~ remained in particles with diameter between 100nm and 200nm. The measured  
 280 ~~size-resolved  $\sigma_{abs}$ BCMSD~~ changed a lot when multiple charging corrections were implemented,  
 281 which highlighted the necessity of implementation of appropriate multiple charging corrections

282 The  ~~$\sigma_{abs}m_{BC}$~~  integrated from measured ~~size-resolved  $\sigma_{abs}$ BCPMSD~~ changed after multiple  
 283 charging corrections. Fig. S4 showed the comparison results of the  ~~$\sigma_{abs}m_{BC}$~~  measured by AE33 and  
 284 the  ~~$\sigma_{abs}m_{BC}$~~  integrated from AE51 measurements. The  ~~$\sigma_{abs}m_{BC}$~~  integrated from uncorrected and  
 285 corrected ~~size-resolved  $\sigma_{abs}$ BCMSD~~ versus  ~~$\sigma_{abs}m_{BC}$~~  measured by AE33 were shown in Fig.S4(a)  
 286 and Fig.S4(b), respectively. Before multiple charging corrections, the  ~~$\sigma_{abs}m_{BC}$~~  from uncorrected  
 287 ~~size-resolved  $\sigma_{abs}$ BCPMSD~~ increased linearly with the  ~~$\sigma_{abs}m_{BC}$~~  from AE33, with  $R^2$  equaling  
 288 0.87, but it was 2.37 times that of AE33 in average. As a comparison, overall magnitude of  
 289  ~~$\sigma_{abs}m_{BC}$~~  integrated from corrected ~~size-resolved  $\sigma_{abs}$ BCPMSD~~ agreed better with that from AE33  
 290 with a slope of 1.2. With the discussion above, multiple charging corrections were essential for  
 291 size-resolved  $\sigma_{abs}$  and BCMSD measurements.

### 292 3.2 Fitting the BCMSD by using two log-normal models

293 Based on the measurement results, the BCMSD had two modes for most of the conditions. The

294 BCMSD are assumed to be of two log-normal distributions as:

$$295 \quad m_{fit,Dp} = \sum_{i=1,2} \frac{m_i}{\sqrt{2\pi \log(GSD_i)}} \cdot \exp\left(-\frac{[\log(D_p) - \log(D_{m,i})]^2}{2 \log^2(GSD_i)}\right), \quad (19)$$

296 Where  $D_p$  is the diameter of the aerosols;  $m_i$  is the mass of mode  $i$  ( $i=1,2$ );  $GSD_i$  is the geometric  
297 standard deviation at mode  $i$  ( $i=1,2$ ), and  $D_{m,i}$  is the geometric mean diameter of the mode  $i$  ( $i=1,2$ ).  
298 The  $GSD_i$  and  $D_{m,i}$  can be determined by using the least square method with the objective function  
299 as :

$$300 \quad J = \sum_{i=1,n} (m_{Dp_i} - m_{fit,Dp_i}(D_{m1}, GSD_1, D_{m2}, GSD_2))^2, \quad (20)$$

301 Where  $m_{Dp_i}$  is the measured mass distribution at  $Dp_i$ , while  $m_{fit,Dp_i}$  is the fit mass distribution at  
302  $Dp_i$ .

### 303 3.3 Estimating aerosol optical properties with different BCMSD

304 The Mie scattering model was used to study the influence of the BCMSD variation on the  
305 aerosol optical properties. When running the Mie model, aerosol PNSD and BC were necessary. ~~The~~  
306 ~~aerosol PNSD and  $m_{BC}$  used here is the mean result of aerosol PNSD and  $m_{BC}$  over the whole~~  
307 ~~field measurement respectively. The amount of BC particle adopted in this study is the mean value of~~  
308 ~~the  $m_{BC}$  measured by AE33.~~ In this study, The BCMSD was assumed to be log-normal distributed.  
309  $D_m$  of the BCMSD was set to vary from 100 nm to 600 nm. Geometric standard deviation (GSD) of  
310 the BCMSD was set to be in the range between 1.3 and 1.8. BC was treated as partially externally  
311 mixed and the remaining aerosols was treated as core-shell mixed. The ratio of externally mixed  
312  $m_{BC}$  to core-shell  $m_{BC}$  was determined by the method introduced in Ma et al. (2012) and a mean  
313 ratio of 0.51 was used. The density and refractive index of BC were set as 1.5 g/cm<sup>3</sup> and 1.8+0.54i  
314 (Kuang et al., 2015), respectively. The complex refractive index of non-absorbing aerosols was  
315 1.53+10<sup>-7</sup>i (Wex et al., 2002) at the wavelength of 525 nm. More details of calculating the aerosol  
316 optical properties by using the aerosol PNSD and BCMSD, can refer to Kuang et al. (2016).

317 The aerosol PNSD and  $m_{BC}$  used here is the mean result of aerosol PNSD and  $m_{BC}$  over the  
318 whole field measurement respectively. The amount of BC particle adopted in this study is the mean  
319 value of the  $m_{BC}$  measured by AE33. For each BCMSD, extinction coefficient ( $\sigma_{ext}$ ), the scattering  
320 coefficient ( $\sigma_{sca}$ ), the single scattering albedo (SSA), and the asymmetry factor ( $g$ ) could be obtained  
321 from the output of Mie scattering model.

### 322 3.4 Evaluating the DARF with different BCMSD



323 In this study, the SBDART model (Ricchiuzzi et al., 1998) was employed to estimate the DARF.  
324 In our study, the instantaneous DARF for cloud free conditions at the top of atmosphere was  
325 calculated for irradiance wavelength range from 0.25 to 4  $\mu\text{m}$ . Input of the model required the  
326 profiles of aerosol  $\sigma_{\text{ext}}$ , SSA, g. These ~~values-profiles~~ were calculated ~~by-from the parameterized~~  
327 ~~aerosol PNSD, BCMSD-profiles~~ parameterization of the aerosol vertical distributions. Details of  
328 calculating the  $\sigma_{\text{ext}}$ , SSA and g profiles can refer to part 4 in the supplementary material. ~~The~~  
329 ~~corresponding DARF for different BCMSD could be estimated.~~ In brief, the aerosol  $\sigma_{\text{ext}}$ , SSA  
330 and g profiles were calculated based on the given aerosol PNSD and BCMSD. The DARF can be  
331 estimated using the above aerosol optical profiles.

332 The aerosol optical properties and the corresponding aerosol optical profiles vary with different  
333 BCMSD. Then the DARF should be different for different BCMSD. By estimating the DARF using  
334 different aerosol BCMSD, the influence of BCMSD on the aerosol radiative properties can be  
335 studied. More details of estimating the DARF could refer to part 4 and 5 in the supplementary  
336 ~~material. The corresponding DARF for different BCMSD could be estimated. The DARF was~~  
337 ~~estimated for the measured mean aerosol PNSD and  $m_{\text{gc}}$  under different BCMSD conditions to~~  
338 ~~study the effects of BCMSD variations on the aerosol DARF.~~

## 339 **4 Results and Discussions**

### 340 **4.1 Measurement results of the BCMSD**

341 The time series of measured PM<sub>2.5</sub>, aerosol PNSD and BCMSD were shown in Fig. 3. During  
342 the observation period, the PM<sub>2.5</sub> varied from 0.06 to 220  $\mu\text{g}/\text{m}^3$ , with a mean value of  $71.5 \pm 52.56$   
343  $\mu\text{g}/\text{m}^3$ . Three periods of heavy PM<sub>2.5</sub> loading were observed: (1) PM<sub>2.5</sub> increased from around 100  
344  $\mu\text{g}/\text{m}^3$  to 200  $\mu\text{g}/\text{m}^3$  and decreased slowly to 1  $\mu\text{g}/\text{m}^3$  in the period 21-26, March; (2) the PM<sub>2.5</sub>  
345 accumulated slowly from 28 to 30, March and dissipated quickly from 30, March to 1, April; (3) the  
346 rapid accumulation and dissipation of PM<sub>2.5</sub> happened during 2 to 5, April. During the last five days,  
347 PM<sub>2.5</sub> fluctuated between 20 and 120  $\mu\text{g}/\text{m}^3$ . For each pollution condition, both the aerosol total  
348 number concentrations and the aerosol median diameter increased. The aerosol median diameter  
349 varied between 31 nm and 169 nm with a mean value of  $78 \pm 31$  nm.

350 ~~As for the BCMSD, a distribution with two modes could be detected. Our measurements shew~~  
351 ~~that the BCMSD had two modes with the coarser mode ranging between 430 nm and 580 nm in~~

352 mobility diameter. Many field measurements had revealed that most of the BC mass locates in the  
353 aerodynamic diameter range of 320 nm and 560 nm using the MOUDI (Hu et al., 2012;Huang and  
354 Yu, 2008). When the aerodynamic diameter was transformed into mobility diameter with assumption  
355 a aerosol effective density of 1.3, the measured BC aerodynamic diameter range corresponded to  
356 mobility diameter range of 280 nm and 491 nm. Therefore, the measured size range for coarser mode  
357 of BCMSD agreed well with the previous measurement.

358 The measured aerosol in the field site was representative of the urban aerosol. The BC particles  
359 emitted by vehicles contributed significantly to the total aerosol BC mass. These BC particles were  
360 rarely coated or thinly coated, and the BC core diameter peaked around 120 nm (Zhang et al., 2017).  
361 Therefore, the BCMSD of the smaller mode measured in our study corresponded to these uncoated  
362 of thinly coated particles. The presence of the first mode in the size range between 100 and 200 nm  
363 provided a verification of previous field measurements that the BC concentrated in the particle  
364 diameter range from 100 to 200 nm. (Huang et al., 2012;Ohata et al., 2011;Wu et al., 2016b). The  
365 peak diameter of second mode ranged from 300 nm to 600 nm, which agrees well with the measured  
366 BCMSD by MOUDI (Klaus Willeke and Baron, 1996;Yu and Yu, 2009;Huang and Yu, 2008). The  
367 main BC mass loading was in the coarser mode for the sampled particles when comparing the BC  
368 mass concentrations at two modes.

369 The total  $m_{BC}$  measured by AE33 ranged from 0.1 to 14  $\mu\text{g}/\text{m}^3$  with an average of  $5.04 \pm 2.64$   
370  $\mu\text{g}/\text{m}^3$ . Good consistence was achieved between  $m_{BC}$  measured by AE33 and  $m_{BC}$  calculated from  
371 measured BCMSD as shown in Fig. 34(ed).

## 372 **4.2 Evolution of the BCMSD under different polluted conditions**

373 Log-normal distribution was used to fit each mode of the BCMSD by using the least square  
374 method as introduced in section 3.2. For each mode, the geometric mean diameter ( $D_m$ ) and the  
375 geometric standard deviation (GSD) of the BCMSD were studied.

376 During the measurement period, both  $D_m$  and GSD of the two modes had changed significantly  
377 as shown in Fig 4S7. The  $D_m$  of first and second mode varied from ~~128-139~~ to ~~162-161~~ nm and  
378 from ~~430-420~~ to ~~580-597~~ nm, respectively. The corresponding mean  $D_m$  was ~~150-151~~ and ~~503-520~~  
379 nm. The  $D_m$  of the two modes was found to be positive correlated in Fig. 4aS7(a). When the  
380 pollution was released from the beginning to 27, March, the  $D_m$  decreased from ~~590to-597~~ to 420

381 nm and from ~~155-160~~ to ~~130-140~~ nm for the coarser mode and the smaller mode respectively. The  
382 BC containing aerosols tended to be aged and grew larger when the air surrounding get polluted.

383 GSD for the coarser mode and the smaller mode showed very different properties as shown in  
384 Fig. ~~4bS7(b)~~. For the second mode, GSD varied from around 1.49 to 1.68 with a mean value of 1.57.  
385 The GSD get decreased with the pollution condition, which indicated that BC containing aerosols  
386 tend to accumulate to a small range of diameters during the aging processing. This phenomenon was  
387 consistent with the fact that larger particles grew relative slower in diameter because the growth ratio  
388 of small aerosol particle is proportion to the negative power of it's diameter. For the first mode, GSD  
389 ranged from ~~1.5-41~~ to around ~~1.85-86~~ with a mean value of ~~1.6263~~. However, GSD of the smaller  
390 mode tend to be larger when the surrounding air get cleaner, which might be related to the complex  
391 sources of the BC emission. A small amount of fresh emitted BC particles can have substantial  
392 influence on the mass size distribution of the smaller mode because the BC concentrations of the  
393 smaller mode were small, especially under clean conditions. In general, the GSD of coarser mode  
394 was a good indicator of the BC aging process and that of the smaller mode could partially reflect the  
395 complex sources of the BC fine particles.

396 The relationship between the  $D_m$  and the GSD for coarser mode was further analyzed by  
397 analyzing the distribution of the  $D_m$  and GSD. The GSD and  $D_m$  had opposite trends as shown in  
398 Fig 5. With the increment of the  $D_m$  from 420 to 540 nm, the mean value of GSD decreased from  
399 around ~~1.6085~~ to ~~1.548-528~~ while the  $m_{BC}$  increased with the  $D_m$ . The statistical relationship  
400 between  $D_m$  and GSD offered a reasonable representation of the BCMSD under different polluted  
401 conditions. In the following work, mean values of the GSD at different  $D_m$  were used to for further  
402 discussion. The  $m_{BC}$  and GSD is positively correlated. The  $m_{BC}$  increased from 2.4 to 8.3  
403  $\mu\text{g}/\text{m}^3$  when the  $D_m$  increased from 420 to 540 nm.

404 Note that the GSD get slightly increased with the increment of  $D_m$  when  $D_m$  was larger than  
405 520 nm. This might be caused by the limit diameter range of BCMSD measuring system which was  
406 from 20 to 680 nm. The multiple charge corrections applied to the BCMSD could influence the  
407 BCMSD when  $D_m$  of the BCMSD was near the end of the scanned diameter and may lead to  
408 significant uncertainties to the BCMSD. The measurement results indicated that cases of measured  
409  $D_m$  of BCMSD larger than 520 nm were few, demonstrating that this multiple correction effect  
410 influenced little on shape of measured BCMSD in most cases.

### 4.3 Influence of BCMSD variation on the aerosol optical properties

The aerosol optical parameters using the measured mean aerosol PNSD and mean  $m_{BC}$  corresponding to different GSD and  $D_m$  values were shown in Fig. 6. In Fig. 6(a), the aerosol  $g$  varied from 0.617-603 to 0.649-627 (variation of 5.84%). Recent work by Zhao et al, 2017 showed that the aerosol  $g$  value in the NCP may vary at a range of 10% due to the change of aerosol PNSD. Aerosol  $g$  was more sensitive to  $D_m$  when the geometric mean diameter of the BCMSD was lower than 400 nm. However, when the  $D_m$  was larger than 400 nm, the  $g$  become sensitive to both the  $D_m$  and the GSD of BCMSD. Overall, the  $g$  varied a little bit (0.617-02 to 0.624609) under the representative conditions during the measurement period. For the aerosol SSA, it was sensitive to the  $D_m$  over the whole range as shown in Fig. 6(b). SSA varied between 0.86-90 and 0.88-94 under the representative measurement conditions. The  $\sigma_{sca}$  had large changes from 264-325.6  $Mm^{-1}$  to 313-364.4  $Mm^{-1}$ . The  $\sigma_{sca}$  was quite sensitive to variations in BCMSD when the  $D_m$  was lower-larger than 400-450 nm as shown in Fig.6c, ~~which varied substantially from 264  $Mm^{-1}$  to 313  $Mm^{-1}$ .~~ In addition, variations in  $\sigma_{sca}$  relied more on the variations in  $D_m$  when  $D_m$  was larger-lower than 400 nm. Within the measurement conditions of BCMSD, the  $\sigma_{sca}$  varied from 265-328  $Mm^{-1}$  to 280-345  $Mm^{-1}$ . The measured GSD under different  $D_m$  went along with the gradient direction of the  $\sigma_{sca}$ , which mean that the evolution of BCMSD in the atmosphere influenced substantially on  $\sigma_{sca}$ . As for the  $\sigma_{abs}$ , it changed from 21.944.06  $Mm^{-1}$  to 44.1237.27  $Mm^{-1}$  and the corresponding mass absorption cross section (MAC) was estimated to be in the range of 4.755.44 to 9.568.08  $m^2/g$ , suggesting that MAC of the BC aerosols should be carefully studied under different BCMSD conditions.

### 4.4 Influence of BCMSD on the direct aerosol radiative forcing

The estimated DARF values for different GSD and  $D_m$  conditions were estimated. When estimating the DARF, the measured mean aerosol PNSD and mean BC mass concentration were used-. The results of estimated DARF were shown in Fig. 7(a). DARF at the surface varied from -4.90-4.3  $w/m^2$  to -2.02-3.59  $w/m^2$  for different BCMSD. Within the measured BCMSD range, the DARF varied from -2.04w-3.97w/m<sup>2</sup> to -2.53.67w/m<sup>2</sup>, which corresponding to 22.58.45% of variation. The heating rate within the mixed layer was a powerful indicator of the BC particles' absorbing effects, which may help evaluate the development of the boundary layer. The calculated mean heating rate within the mixed layer changed from 3.252.16 K/day to 3.892.65 K/day for different  $D_m$  and GSD, as shown in Fig. 7(b). The heating rate with the measured BCMSD range

could change from ~~3.562.24~~ to ~~3.752.50~~ with a variation of ~~5.2311.6~~%.

Mixing states of BC play significant roles in calculations of aerosol optical properties and estimations of DARF (Jacobson, 2001). As a comparison, we estimated the DARF under different conditions of BC mixing state: (1) internally mixed, (2) externally mixed and (3) core-shell mixed. Table 1 gave the estimated DARF and mean heating rate within the mixed layer under different mixing state conditions. Results showed that the DARF under different BC mixing states conditions may change by ~~21.510.50~~%, which shared the same magnitude with ~~22.58.45~~% variation of DARF caused by BCMSD variations. In addition, the heating rate was estimated to vary by ~~6.059.71~~%. These results highlighted that the BCMSD plays significant roles in variations of aerosol optical properties and estimations of DARF as well as the air heating rate caused by the existence of BC particles. It was recommended that a real time measured BCMSD be used when estimating the aerosol DARF, instead of a constant one. The BCMSD was as important as that of the BC mixing states.

## 5 Conclusions

Knowledge of the BC microphysical properties especially the size-dependent information can help reduce the uncertainties when estimating the aerosol radiative effects. BCMSD is an important quantity in its own right, being directly and indirectly applicable to determination the sources, aging processes and mixing states of BC aerosols. In this study, the characteristics of BCMSD were studied from the field measurement results by using our own developed measurement algorithm.

The BCMSD measurement system was developed and validated based on the works of Ning et al. (2013) by using differential mobility analyzer (DMA) in tandem with Aethalometer (AE). When deriving the BCMSD, a comprehensive multiple charging correction algorithm was proposed and implied. This algorithm was validated by closure study between the measured total  $m_{BC}$  from AE33 and the  $m_{BC}$  integrated from the measured BCPMSD using the datasets from field measurements. Results showed that the multiple charging corrections could significantly change the shapes and magnitudes of the raw measured BCPMSD. The accurate BCPMSD characteristics could be obtained by our proposed method in this paper.

The developed measurement system was employed in a field campaign in the North China Plain from 21 March to 9 April in 2017. The BCMSD was found to have two quasi-lognormal modes with peaks at around 150 nm and 500 nm, respectively. These two modes were consistent with the

471 previous measurement results by MOUDI (Wang et al., 2015;Hu et al., 2012). The amount of the BC  
472 mass concentrations for the coarser mode peaks were about twice to that of the ~~fine-smaller~~ mode.

473 The characteristic of the BCMSD was studied by fitting the shape of BCMSD with a bi-normal  
474 distribution. The relationships between the fitted  $D_m$  and GSD were statistically studied. During the  
475 aging processing, the opposite trends for the  $D_m$  and GSD were found for coarser mode. This is the  
476 first time that the coarser mode of the BCMSD were synthetically studied. The BCMSD of coarser  
477 mode varied significantly under different pollution conditions with peak diameter changed between  
478 430 and 580 nm. However, the relationship between the  $D_m$  and GSD for smaller mode BC aerosols  
479 were more complex due to the complex sources.

480 When the BCMSD were changed with the polluted condition, the corresponding aerosol optical  
481 properties changes significantly. Sensitivity studies found that the aerosol  $g$  varies from ~~0.617-603~~ to  
482 ~~0.649-627~~ due to the variations in BCMSD. Aerosol  $g$  was more sensitive to  $D_m$  when the  
483 geometric mean diameter of the BCMSD is in the range of 300 nm and 370 nm. The SSA can  
484 changed from ~~0.9086~~ to ~~0.9394~~. The  $\sigma_{sca}$  experienced significant change with the variation of  
485 BCMSD from ~~264-325.6~~  $Mm^{-1}$  to ~~313-364.4~~  $Mm^{-1}$  and the  $\sigma_{abs}$  changed in the range between  
486 ~~21.94.064~~  $Mm^{-1}$  and ~~44.1237.27~~  $Mm^{-1}$ . The corresponding BC MAC was calculated to be in the  
487 range between ~~4.755.44~~ and ~~9.568.08~~  $m^2/g$ .

488 The variations in DARF were estimated due to the variations of the BCMSD by using the  
489 SBDART model. Results showed that the DARF can varies by about ~~22.58.45~~% for different  
490 BCMSD and the heating rate for different measured BCMSD conditions could change from ~~3.562.24~~  
491 to ~~3.752.50~~, corresponding to a variation of ~~5.2311.6~~%. At the same time, the variations in DARF  
492 due to the variations in the BC mixing state was estimated to be ~~21.510.5~~% and that of the heating  
493 rate is ~~6.058.45~~%. Thus, the variations of the BCMSD may had significant influence on the aerosol  
494 radiative budget and an accurate measurement of BCMSD was very necessary.

495  
496 **Competing interests.** The authors declare that they have no conflict of interest.

497 **Data availability.** The data used in this study is available when requesting the authors.

498 **Author contributions.** GZ, CZ, JT and YK designed and conducted the experiments; CS, YY, CZ and GZ  
499 discussed the results.

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502

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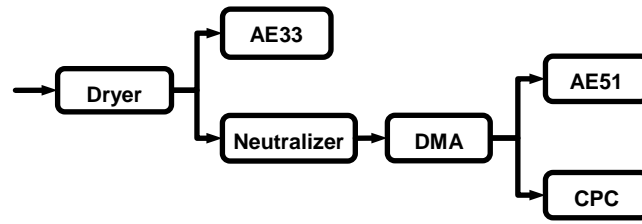
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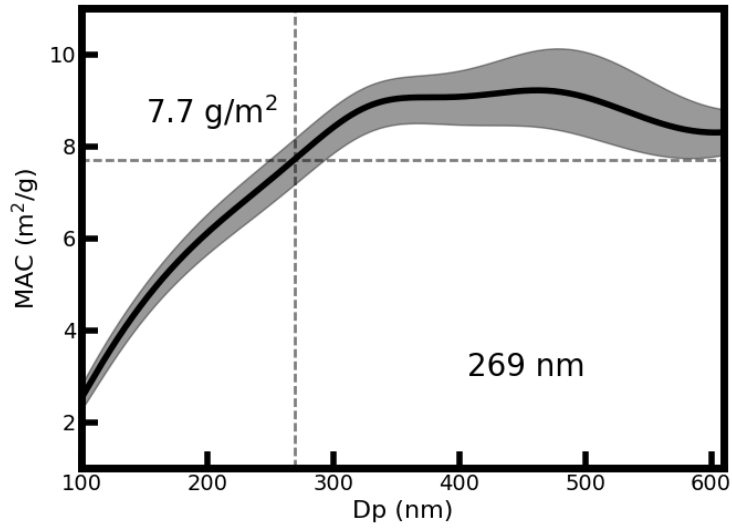


**Figure 1.** The schematic diagram of the instrument setup.

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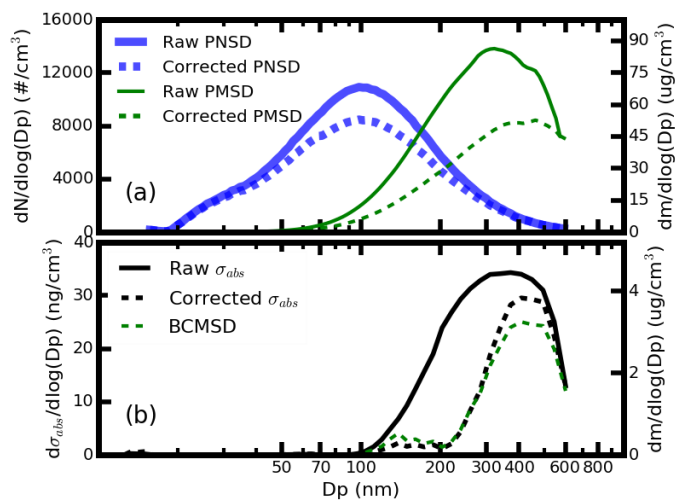


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759 **Figure 2.** Calculated mass absorption coefficient of different aerosol.

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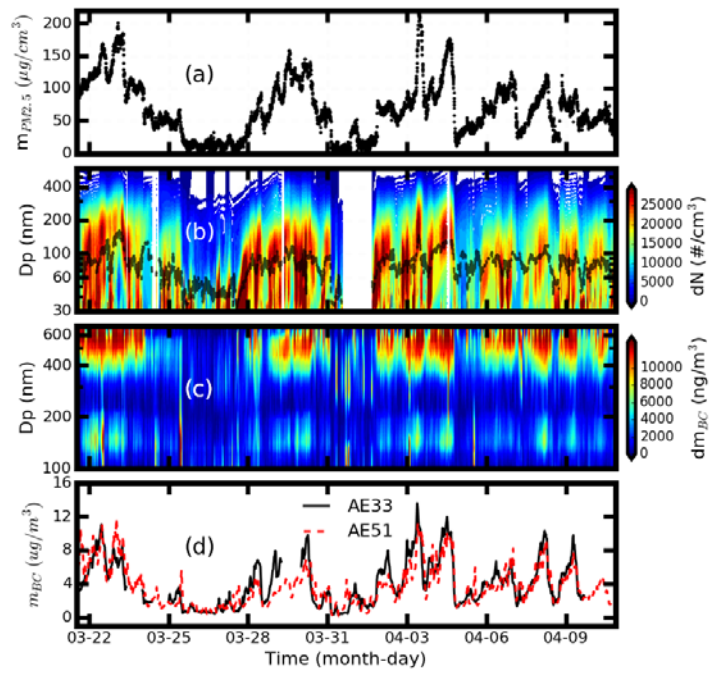


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763 **Figure 23.** Case of multiple charging correction processing. (a) the multiple charging correction of  
 764 the aerosol PNSD and aerosol PMSD, (b) the multiple charging correction of the size-resolved  
 765  $\sigma_{abs}$  ~~the BCPMSD~~. The solid line is the measured results without multiple charging corrections and  
 766 the dotted line is the multiple charging corrections results.

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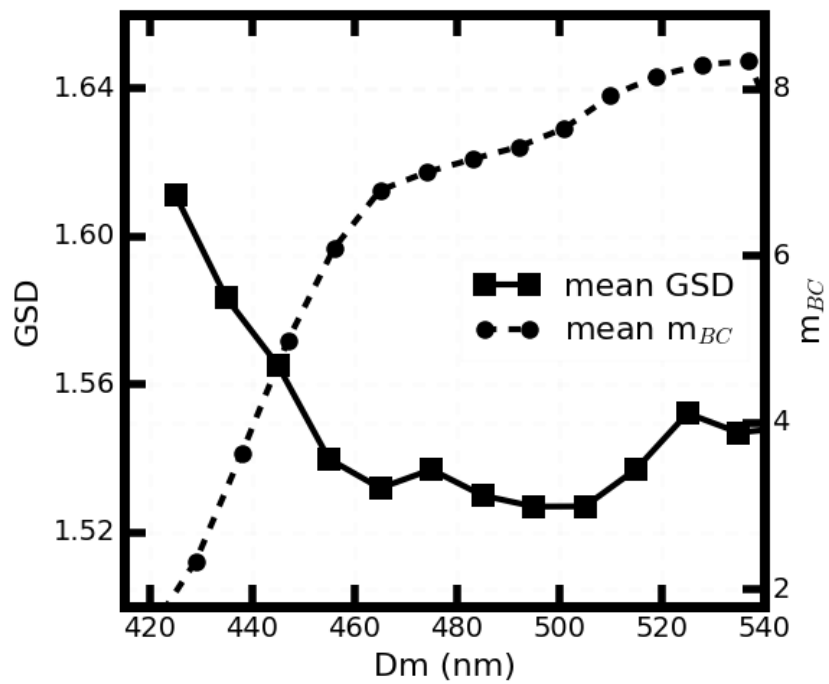
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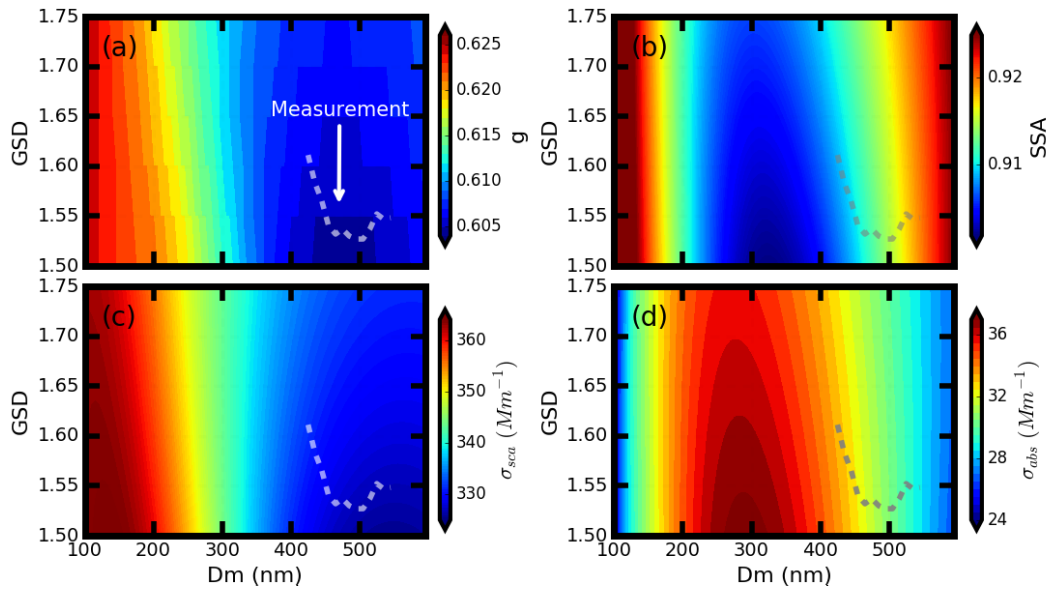
**Figure 34.** The measured time series of mass concentrations for (a) the PM<sub>2.5</sub>; (b) the aerosol PNSD in filled color, the geometric median diameter in dotted line; ~~and~~ (c) the BCMSD and; (d) the  $m_{BC}$  by AE33 (black) and  $m_{BC}$  from integrated BCMSD from AE51 (red).



776

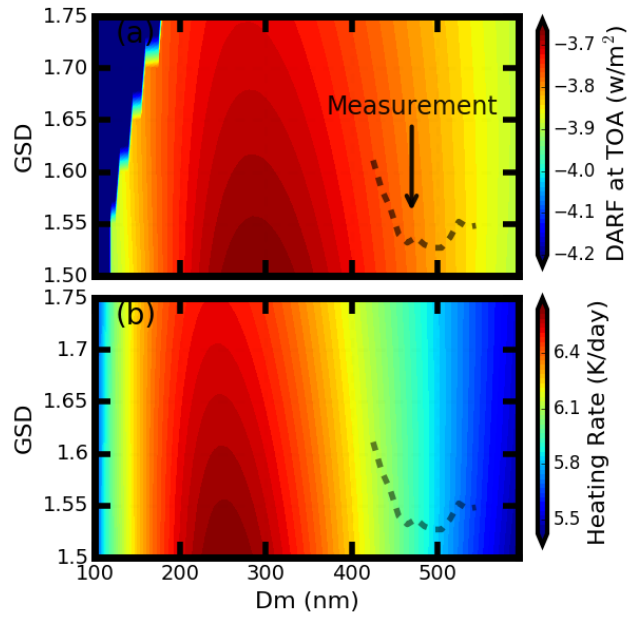
777 **Figure 5.** The relationship between the Dm and the GSD. The black dots show the real measured Dm  
 778 and GSD. The black line shows the mean results of the GSD for different Dm. The black line marked  
 779 with square shows the variation of mean  $m_{BC}$  with the Dm.

780



781 **Figure 6.** Variations of aerosol optics properties using the measured mean aerosol PNSD and  $m_{BC}$   
 782 under different BCMSD conditions, which are represented by different Dm and GSD values: (a)  
 783 aerosol asymmetry factor, (b) single scatter albedo, (c) scattering coefficient and (d) extinction  
 784 coefficient . The grey dotted line in the figure shows the evolution path of the BCMSD according to  
 785 results of field measurements.

786



787

788 **Figure 7.** Variations of (a) DARF and (b) heating rate under different BCMSD conditions, which are  
 789 represented by different Dm and GSD values. The black dotted line in the figure shows the evolution  
 790 path of the BCMSD according to results of field measurements.

791

792 **Table 1.** Comparison of the DARF and heating rate values under different BC mixing states and  
 793 different BCMSD conditions.

		Mixing State			BCMSD	
		Internal	External	Core-Shell	Minimum	Maximum
<b>DARF</b>	<b>Value(w/m<sup>2</sup>)</b>	<b>-3.45</b>	<b>-3.56</b>	<b>-3.81</b>	<b>-3.97</b>	<b>-3.67</b>
	<b>Variation</b>	<b>10.5%</b>			<b>8.45%</b>	
<b>Heat Rate</b>	<b>Value(K/day)</b>	<b>2.51</b>	<b>2.32</b>	<b>2.53</b>	<b>2.24</b>	<b>2.50</b>
	<b>Variation</b>	<b>9.71%</b>			<b>11.6%</b>	

794

1 Supplement for

2 **Role of black carbons mass size distribution in the direct aerosol radiative forcing**

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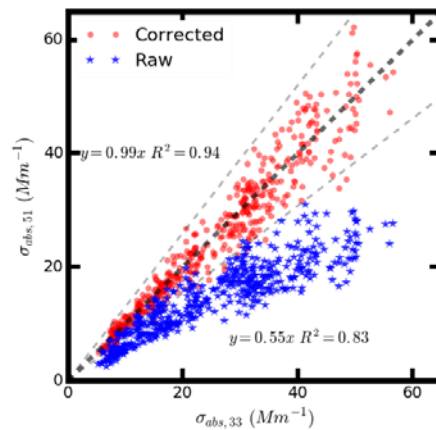
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7 **1. Correcting the AE51**

8 Fig. S1 showed the results of the loading effect corrections. At the beginning of the field  
9 experiment, parallel measurement of  $\sigma_{abs}$  by AE51 and AE33 was conducted. Before corrections, the  
10 measured  $\sigma_{abs}$  by AE51 and AE33 showed significant discrepancy with each other with slope and  $R^2$   
11 equaling 0.55 and 0.83. However, the  $\sigma_{abs}$  measured by AE33 and by AE51 with loading effects  
12 corrections showed good consistency in trends and magnitudes with slope and  $R^2$  of 0.98 and 0.94  
13 respectively. These results demonstrated that the loading effects corrections of  $\sigma_{abs}$  from AE51 were  
14 essential and the value of  $\sigma_{abs}$  from AE33 can be used as a reference for the measured size-resolved  
15  $\sigma_{abs}$ .

16

17



18

19 **Figure S1.** Comparison between the  $\sigma_{abs}$  measured by AE51 and AE33. The blue stars and the red  
20 dots represents uncorrected and corrected  $\sigma_{abs}$  of AE51 respectively.

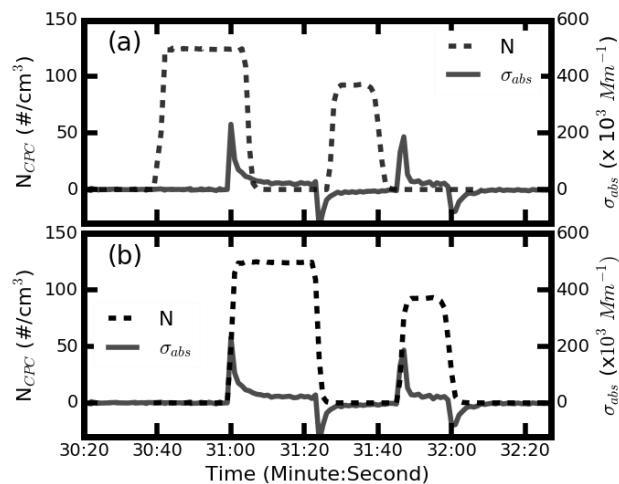
21

22 **2. Time correction**



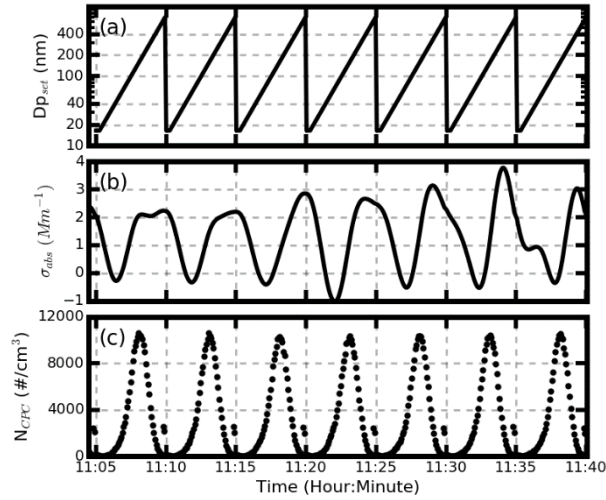
23 There were two reasons that can lead to this difference: firstly, the time of the AE51 system and  
 24 the computer that controls the CPC cannot be synchronized all the time; secondly, there existed a  
 25 difference in the plumbing delay time, which was the time required for particles to flow through the  
 26 tubing interconnecting the DMA and CPC or AE51, and arrive at the detector. To sum up, the  
 27 synchronization of the time reported by CPC and AE51 was necessary.

28 Time synchronization was conducted by measuring the time lag of the signal pulses from the  
 29 DMA to CPC and AE51. The signal pulses resulted from the sudden change of the aerosol diameter  
 30 scanned by DMA. Details of the method were shown below. In fig. S2, the black solid line gave the  
 31 time series of the measured  $\sigma_{abs}$  by AE51. The dotted lines gave the time series of the aerosol number  
 32 counted by CPC of (a) unsynchronized and (b) synchronized. In the beginning, the scan diameter of  
 33 the DMA was set to be less than 13nm and the values measured by AE51 and CPC are nearly zero. The  
 34 values get a step jump and a step drop when changing the scan diameter up to about 200nm and down  
 35 back to less than 13nm. About 15s later, these procedures were conducted once again. From fig. S2(a)  
 36 and fig. S2(b), the lag time of the AE51 and CPC were determined to be 20s by matching the pulse  
 37 signals.



38  
 39 **Figure S2.** An example of time synchronization processing, (a) for unsynchronized and (b) for  
 40 synchronized. The dotted line is the aerosol number concentration time series counted by CPC. The  
 41 black solid line is the  $\sigma_{abs}$  measured by AE51.

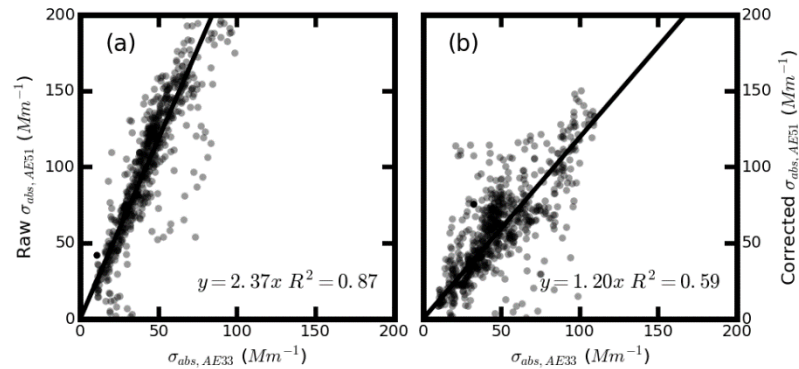
42 **3. Time series diagram of scanned aerosol diameters, measured  $m_{BC}$  and the aerosol number**



44

45 **Figure S3** (a) the diameters of the aerosols that pass through the DMA (b) The  $\sigma_{abs}$  values measured  
46 by AE51, (c) the aerosol number concentrations measured by CPC.

#### 47 4 Validation of the multiple charging corrections



48

49 **Figure S4.**  $\sigma_{abs}$  measured by AE33 versus  $\sigma_{abs}$  integrated from AE51 of (a) uncorrected  
50 size-resolved  $\sigma_{abs}$ , (b) multiple-charging corrected size-resolved  $\sigma_{abs}$ .

51

#### 52 5 Estimate the DARF

53 DARF is defined as the difference between radiative flux at the TOA under present aerosol  
54 conditions and aerosol-free conditions:

$$55 \quad \text{DARF} = (f_a \downarrow - f_a \uparrow) - (f_m \downarrow - f_m \uparrow) , \quad (21)$$

56 Where  $f_a \downarrow$  is the downward radiative irradiance and  $f_a \uparrow$  is the outward radiative irradiance under  
57 given aerosol distributions;  $(f_a \downarrow - f_a \uparrow)$  is the downward radiative irradiance flux with given aerosol  
58 distributions and  $(f_m \downarrow - f_m \uparrow)$  is the radiative irradiance flux under aerosol free conditions.

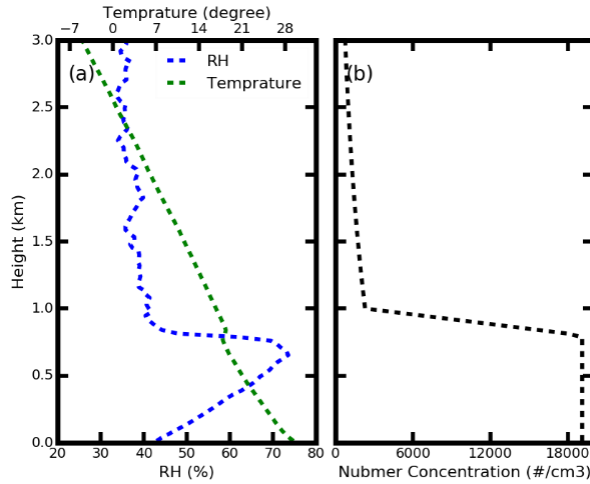
59 Input data for the SBDART are listed below. Vertical profiles of the aerosol optical properties,  
60 which include the aerosol extinction coefficient ( $\sigma_{\text{ext}}$ ), aerosol single scattering albedo (SSA) and  $g$   
61 with a height resolution of 50 m, come from the parameterization of aerosol vertical distributions (as  
62 shown in fig. S4 and the next paragraph) and the results of the Mie model. Atmospheric gas and  
63 meteorological parameter profiles come from the mean results of the radiosonde observations at the  
64 Meteorological Bureau of Beijing (39°48' N, 116°28' E), which include profiles for water vapor,  
65 pressure and temperature during the spring. Surface albedo values are obtained from the Moderate  
66 Resolution Imaging Spectroradiometer (MODIS) V005 Climate Modeling Grid (CMG) Albedo  
67 Product (MCD43C3) during March, 2017 of Beijing, where the field campaign is conducted. The  
68 remaining input data for the SBDART are set to their default values.

### 69 **5.1 Parameterization of the aerosol vertical distribution**

70 Liu et al. (2009) studied vertical profiles of aerosol total number concentration ( $N_a$ ) with aircraft  
71 measurements, and derived a parameterized vertical distribution. In this scheme,  $N_a$  is constant in the  
72 mixed layer, with a transition layer where it linearly decreases and an exponential decrease of  $N_a$   
73 above the transition layer. The same parameterized scheme proposed by Liu et al. (2009) is adopted by  
74 this study as shown in fig. S4 (b). Both the study of Liu et al. (2009) and Ferrero et al. (2010) manifest  
75 that the dry aerosol PNSD in the mixed layer varies little. The shape of the dry aerosol PNSD is  
76 assumed constant with height, which means that aerosol PNSD at different heights divided by  $N_a$  give  
77 the same normalized PNSD.

78 As for the BC vertical distribution, Ferrero et al. (2011) and Ran et al. (2016) demonstrate that BC  
79 mass concentration in the mixed layer remains relatively constant and decreases sharply above the  
80 mixed layer. According to this, the parameterization scheme of BC vertical distribution is assumed to  
81 be the same as that of aerosol. The shape of the size-resolved BC mass concentration distribution is  
82 also assumed to be the same as that at the surface.

83



84

85 **Figure S5.** The mean RH, temperature, and aerosol number concentration profiles.

86

87 **5.2 Calculate the aerosol optical profiles under the given RH profile**

88 With the vertical distribution of aerosol PNSD and BCMSD, the aerosol optical properties at a  
 89 given RH profile can be calculated by using the Mie scattering model and  $\kappa$ -Köhler theory (Petters and  
 90 Kreidenweis, 2007).

91 The aerosol hygroscopic growth is taken into consideration when calculate the aerosol optical  
 92 properties under the given RH. The  $\kappa$ -Köhler theory (Petters and Kreidenweis, 2007) is widely used to  
 93 describe the hygroscopic growth of aerosol particles by using a single aerosol hygroscopic growth  
 94 parameter ( $\kappa$ ) and the  $\kappa$ -Köhler equation, which is shown as

95 
$$\frac{RH}{100} = \frac{gf^3 - 1}{gf^3 - (1 - \kappa)} \cdot \exp\left(\frac{4\sigma_{s/a}M_{water}}{R \cdot T \cdot D_d \cdot gf \cdot \rho_w}\right), \quad (1)$$

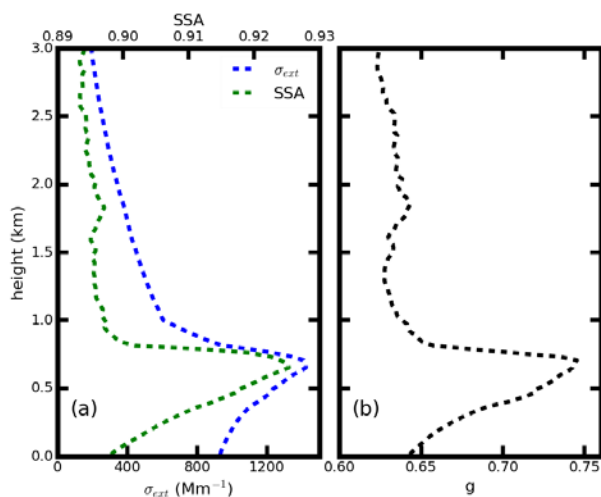
96 where  $D_d$  is the dry particle diameter;  $gf(RH)$  is the aerosol growth factor, which is defined as the  
 97 ratio of the aerosol diameter at a given RH and the dry aerosol diameter ( $D_{RH}/D_d$ );  $T$  is the  
 98 temperature;  $\sigma_{s/a}$  is the surface tension of the solution;  $R$  is the universal gas constant and  $\rho_w$  is the  
 99 density of water. The aerosol hygroscopic growth parameter  $\kappa$  can be further used to investigate the  
 100 influence of aerosol hygroscopic growth on aerosol optical properties (Tao et al., 2014; Kuang et al.,  
 101 2015; Zhao et al., 2017) and aerosol liquids water contents (Bian et al., 2014).

102 The  $\kappa$ -Köhler theory and the Mie scattering model are combined to calculate aerosol extinction  
 103 coefficient, aerosol single scattering albedo and aerosol asymmetry factor under different RH  
 104 conditions. The measured mean  $\kappa$ , which is derived from the humidified nephelometer system (Kuang

105 et al., 2017), is used to account for aerosol hygroscopic growth. For each RH value, the gf can be  
 106 calculated based on equation (1). The corresponding ambient aerosol PNSD at a given RH can be  
 107 determined. The refractive index ( $\tilde{m}$ ), which accounts for water content in the particle, is derived as a  
 108 volume mixture between the dry aerosol and water (Wex et al., 2002):

$$109 \quad \tilde{m} = f_{V,dry} \tilde{m}_{aero,dry} + (1 - f_{V,dry}) \tilde{m}_{water} \quad (2)$$

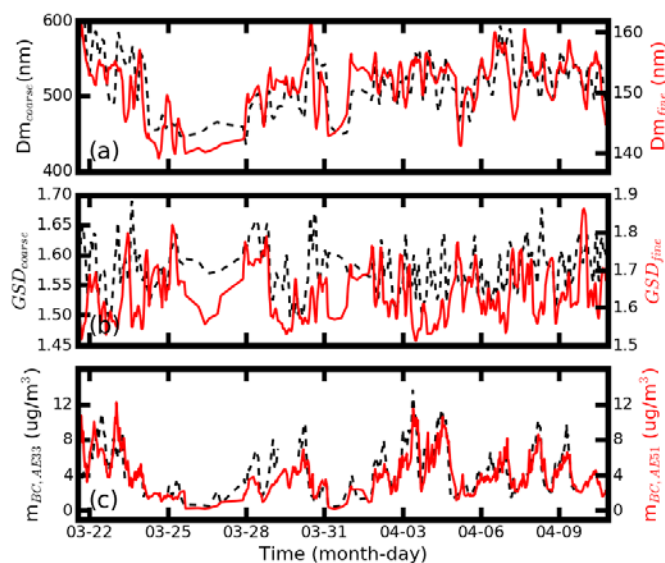
110 where  $f_{v,dry}$  is the ratio of the dry aerosol volume to the total aerosol volume under a given RH  
 111 condition;  $\tilde{m}_{aero,dry}$  is the refractive index for dry ambient aerosols and  $\tilde{m}_{water}$ , the refractive index  
 112 of water, is  $1.33+10^{-7}i$ . Then, the corresponding aerosol optical properties under the given RH and  
 113 PNSD can also be calculated. Finally, the aerosol optical profiles can be calculated. Fig. S6 shows one  
 114 of the calculated aerosol optical profiles.



115  
 116 **Figure S6.** The calculated profiles of the aerosol extinction coefficient, aerosol single scattering  
 117 albedo and the aerosol asymmetry factor.

118

119 **6 Relationship between the GSD, Dm and mBC**



120

121 **Figure S7.** The (a)  $D_m$  and (b)  $GSD$  of the BCMSD at coarse mode (black) and fine mode (red); (c)  
 122 measured  $m_{BC}$  by AE33 (black) and measured  $m_{BC}$  from integrated  $m_{BC}$  of the BCMSD from AE51.

123

124

125

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160

# 1 **Role of black carbons mass size distribution in the direct aerosol radiative forcing**

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## 7 **Abstract**

8 Large uncertainties exist when estimating radiative effects of ambient black carbon (BC) aerosol.  
9 Previous studies about the BC aerosol radiative forcing mainly focus on the BC aerosols' mass  
10 concentrations and mixing states, while the effects of BC mass size distribution (BCMSD) were not  
11 well considered. In this paper, we developed a method by measuring the BCMSD by using a  
12 differential mobility analyzer in tandem with an aethalometer. A comprehensive method of multiple  
13 charging corrections was proposed and implemented in measuring the BCMSD. Good agreement  
14 was obtained between the BC mass concentration integrated from this system and that measured in  
15 bulk phase, demonstrating the reliability of our proposed method. Characteristics of the BCMSD and  
16 corresponding radiative effects were studied based on field measurements conducted in the North  
17 China Plain by using our own designed measurement system. Results showed that the BCMSD had  
18 two modes and the mean peak diameters of the two modes were 150 nm and 503 nm respectively.  
19 The BCMSD of coarser mode varied significantly under different pollution conditions with peak  
20 diameter varying between 430 nm and 580 nm, which gave rise to significant variation in aerosol  
21 buck optical properties. The aerosol direct aerosol radiative forcing was estimated to vary by 8.45%  
22 for different measured BCMSDs of coarser mode, which shared the same magnitude to the variation  
23 associated with assuming different aerosol mixing states (10.5%). Our study reveals that the BCMSD  
24 matters as well as their mixing state in estimating the direct aerosol radiative forcing. Knowledge of  
25 the BCMSD should be fully considered in climate models.

## 26 **1 Introduction**

27 Atmospheric black carbon (BC) is the second strongest absorbing components in atmosphere  
28 (Bond et al., 2013) but the magnitudes of the warming effects are poorly quantified. When emitted to  
29 the surrounding, BC particles transform the morphology from fractal to spherical and then grow as  
30 fully compact particles with other components depositing on the BC aerosol (Peng et al., 2016). The



31 variation in the shapes of BC aerosols, together with the variation in the mixing states, can lead to  
32 substantial change of aerosol optical properties (Liu et al., 2017;China et al., 2013;Wu et al.,  
33 2016a;Wu et al., 2018). BC aerosols also have significant influence on the climate by interacting  
34 with clouds (Koch and Del Genio, 2010;Roberts et al., 2008;Stevens and Feingold, 2009), ice and  
35 snow (Bond et al., 2013). Recent study shows that the solar absorption of BC can suppress the  
36 turbulence in the atmospheric boundary layer (Wilcox et al., 2016). It is found that BC emissions  
37 may be responsible for the incensement of droughts and floods in China and India (Menon et al.,  
38 2002). In addition, BC can pose a serve threat to human health through inhalation (Nichols et al.,  
39 2013;Janssen et al., 2011).

40 Comprehensive studies have been carried out to evaluate the climate effect of BC based on the  
41 measurement of BC mass concentrations ( $m_{BC}$ ) (Koch et al., 2009;Ramanathan and Carmichael,  
42 2008). The  $m_{BC}$  near the ground have been well characterized (Ramachandran and Rajesh,  
43 2007;Ran et al., 2016b;Reddington et al., 2013;Song et al., 2013), and the BC vertical distributions  
44 are widely measured and evaluated as well (Ran et al., 2016a;Babu et al., 2011;Ferrero et al., 2011).  
45 Despite these measurements, more insights into the BC microphysical properties can help to estimate  
46 the influence of BC aerosols on visibility (Zhang et al., 2008), climate (Jacobson, 2001) and human  
47 health (Lippmann and Albert, 1969). These microphysical properties include BC morphology (Zhang  
48 et al., 2016), density (Zhang et al., 2016), complex refractive index (Bond et al., 2013),  
49 hygroscopicity (Zhang et al., 2008;Peng et al., 2017), mixing states (Moffet et al., 2016;Raatikainen  
50 et al., 2017), and particularly, the mass size distribution (BCMSD) (Cheng et al., 2012;Cheng and  
51 Yang, 2016;Gong et al., 2016). Knowledge of BCMSD is not only helpful to study the mixing state  
52 of BC aerosols (Raatikainen et al., 2017), but also essential to model the role of BC in evaluating  
53 regional and global climate accurately. BC radiative effects is highly sensitive to the emitted BC  
54 particle size distribution (Matsui et al., 2018). The health impacts of BC are significantly related to  
55 BCMSD (Turner et al., 2015). Furthermore, the information of BCMSD can help to study the source,  
56 the evolution and the mixing state of ambient BC aerosols (Yu et al., 2010).

57 Many methods have been proposed to measure the BCMSD. For instance, the BCMSD was  
58 measured by sampling the aerosol in the size range from about 50 nm to several micrometers onto  
59 quartz fiber filter substrates using a micro-orifice uniform deposit impactor (MOUDI)  
60 (Venkataraman and Friedlander, 1994;Guo, 2016). Cheng et al. (2014) developed a method to

61 measure the BCMSD by employing two aethalometers in parallel, with one to measure total  $m_{BC}$   
62 and the other to measure  $m_{BC}$  below specific particle sizes using a size cut-off inlet. The above two  
63 methods measure the BCMSD corresponding to the aerodynamic diameter. The Single Particle Soot  
64 Photometer (SP2) is developed and widely used because it provides single particle information, hence  
65 the BCMSD and the mixing state of the atmospheric aerosols can be derived directly (Schwarz et al.,  
66 2006;Gao et al., 2007;Huang et al., 2012;Singh et al., 2016). The BCMSD corresponding to the  
67 ambient aerosol mobility diameter can be measured by using a differential mobility analyzer (DMA)  
68 in tandem with SP2 (Raatikainen et al., 2017). However, the laser-induced incandescence method  
69 cannot provide reliable information about the particles beyond the range of 70 nm and 400 nm  
70 (Moteki and Kondo, 2010), which results in the lack of the knowledge of the BCMSD characteristics  
71 for these aerosols over 400 nm. The results from MOUDI find that a great amount of BC locates at  
72 the aerodynamic diameter range of between 370 and 1000 nm (Hu et al., 2012;Huang and Yu, 2008).  
73 However, the measurements of MOUDI cannot give detailed information of the BCMSD evolution  
74 due to the low temporal and diameter resolution (Hu et al., 2012;Huang and Yu, 2008). The  
75 characteristics of the BCMSD larger than 370 nm is not well studied due to the limitation of the  
76 instrument.

77 Recently, Ning et al. (2013) and Stabile et al. (2012) proposed a new method to measure the  
78 BCMSD by using differential mobility analyzer (DMA) in tandem with Aethalometer (AE). This  
79 method has the potential of measuring the BCMSD from 20 nm to 584 nm with high time resolution.  
80 We develop and validate the BCMSD measurement system based on the works of Ning et al. (2013).  
81 The developed measurement system was employed in a field campaign in the North China Plain. The  
82 characteristics of the measured BCMSD were studied based on the field measurement. Furthermore,  
83 the effects of BCMSD variations on the aerosol optical properties and corresponding direct aerosol  
84 radiative properties were evaluated. The aerosol optical properties were calculated by using the Mie  
85 scattering theory. The direct aerosol radiative forcing (DARF) were estimated by using the Santa  
86 Barbara DISORT (discrete ordinates radiative transfer) Atmospheric Radiative Transfer (SBDART)  
87 model.

88 The structure of this paper are organized as follows. Section 2 gives the information about the  
89 instrument setup and field measurement. Section 3 gives the detailed method used in this study,  
90 which contains: 1, conducting multiple charging corrections when deriving the aerosol BCMSD and

91 2, evaluating the aerosol optical and radiative properties for different BCMSD. Results and  
92 discussions are shown in section 4. The conclusion is drawn in the last part.

## 93 **2 Instrument Setup**

94 The measurement system setup was based on the works of Stabile et al. (2012) and Ning et al.  
95 (2013) as schematically shown in Fig.1. The ambient sample aerosol particles were firstly dried to  
96 below relative humidity of 30% through a Nafion drying tube before passing through to the DMA  
97 (Model 3081, TSI, USA). The DMA scanned aerosol particles with diameter ranges from 12.3 to 697  
98 nm over a period of 285 seconds and started another scanning after a pause of 15 seconds, so one  
99 complete cycle took 5 minutes. The sheath and sample flow rates of the DMA were 3 lpm and 0.5  
100 lpm, respectively. The quasi-monodisperse aerosols that passed through the DMA were further  
101 divided into two flows: with one lead to an aethalometer (AE51, Model 51, MicroAeth, USA) with a  
102 flow rate of 0.2 lpm to measure the absorption coefficient ( $\sigma_{abs}$ ) at 1 second time resolution; and the  
103 other one with flow rate of 0.3 lpm flow directed to a CPC (Model 3772, TSI, USA), which counted  
104 particle number concentrations at 0.1 second resolution. Clean air with a flow rate of 0.7 lpm was  
105 used to compensate for the CPC inlet flow, which had default flow rate of 1 lpm. Overall, the  
106 combination system of DMA, CPC and AE51 could provide one PNSD and size-resolved  $\sigma_{abs}$  scan  
107 every 5 minutes. If the mass absorption coefficient (MAC) at a given diameter is known, the  
108 BCMSD can be derived correspondingly.

109 An aethalometer (AE33, Model 33, Magee, USA) was used to measure the  $\sigma_{abs}$  or  $m_{BC}$  with a  
110 time resolution of 1 minute. The mass concentration of particles with diameter smaller than 2.5  $\mu\text{m}$   
111 (PM<sub>2.5</sub>) was concurrently measured with time resolution of 1 minute during the filed observations  
112 by the Tapered Element Oscillating Microbalance (TEOM) Dichotomous Ambient Particulate  
113 Monitor (1405-DF), which was an indicator of the pollution conditions.

114 From 21 March to 9 April in 2017, an intensive field measurement was conducted to  
115 characterize of the ambient dry aerosol BCMSD corresponding to aerosol mobility diameter at the  
116 AERONET BEIJING\_PKU station (N39°59', E116°18'). This station was located on one roof of  
117 Peking University campus in the north west of Beijing, China. There were two main streets, Chengfu  
118 Road to the south and Zhongguancun Street to the west that surrounding the station. The aerosol  
119 sampled at this station were mainly composed of urban roadside aerosols (Zhao et al., 2018).

## 120 **3 Methodologies**

### 121 3.1 Retrieving the BCMSD

122 Five steps were involved to calculate the BCMSD using the raw data from the measurement  
123 system: 1), correcting the ‘loading effect’ and ‘multiple scattering effect’ of  $\sigma_{abs}$  measured by AE51;  
124 2), matching the instrument time between the AE51 and CPC; 3), matching the measured  $\sigma_{abs}$  and  
125 diameter to get the raw size-resolve  $\sigma_{abs}$  that is not involved in multiple charging corrections; 4),  
126 conducting the multiple charging corrections of the measured raw size-resolved  $\sigma_{abs}$ ; 5),  
127 transforming the size-resolved  $\sigma_{abs}$  into BCMSD.

#### 128 3.1.1 Obtaining the raw size-resolved $\sigma_{abs}$

129 The aethalometer (AE51 and AE33) is a well-developed and widely used instrument to measure  
130 the  $\sigma_{abs}$  (Drinovec et al., 2015; Hansen et al., 1984). When absorbing aerosols accumulates on the  
131 sample filter of the aethalometer continuously, the  $\sigma_{abs}$  can be determined by concurrently  
132 measuring the light intensities  $I$  after the fiber filter and the light intensities  $I_0$  transmitted through  
133 reference spot which is free of aerosol loading. The light attenuation (ATN) is defined as:

$$134 \quad ATN = 100 \cdot \ln\left(\frac{I_0}{I}\right). \quad (1)$$

135 The total  $\sigma_{abs}$  of the loaded particle on the filter is given by:

$$136 \quad \sigma_{abs,tot} = \frac{A \cdot ATN}{100}, \quad (2)$$

137 where A is the sample spot area on the filter. The instantaneous  $\sigma_{abs}$  can be calculated through the  
138 increment of  $\sigma_{abs,tot}$ :

$$139 \quad \sigma_{abs} = \frac{\sigma_{abs,tot}}{\Delta t} = \frac{A \cdot \Delta ATN}{100 \cdot F \cdot \Delta t}, \quad (3)$$

140 where F is the flow rate and  $\Delta ATN$  is the ATN variation during the time period of  $\Delta t$ . The  $\sigma_{abs,tot}$   
141 can be transformed to  $m_{BC}$  when the mass attenuation cross-section (MAC) of BC is known.  
142 Traditionally, a constant MAC at 7.7 g/m<sup>2</sup> was used to deduce the  $m_{BC}$  (Drinovec et al., 2015).

143 Corrections of the measured  $\sigma_{abs}$  are necessary because the systematic bias exists due to the  
144 prevalingly known ‘loading effect’ and multiple scattering effect (Drinovec et al., 2015; Virkkula et  
145 al., 2015; Virkkula et al., 2007). The AE33 can directly provide the corrected  $\sigma_{abs}$  values through  
146 measuring two light intensities of two spots with different BC load efficiencies (Drinovec et al.,  
147 2015). For AE51, The correcting method in Virkkula et al. (2007) was adopted:

$$148 \quad \sigma_{abs, corrected} = (1 + k \times ATN) \sigma_{abs, uncorrected}, \quad (4)$$

149 where  $k$  is the correction factor and a constant value of 0.004 is employed in this study to correct  
150 the  $\sigma_{abs}$  from AE51. In the first part of the supplementary material, we showed that the loading  
151 effects corrections of  $\sigma_{abs}$  from AE51 were essential and the value of  $\sigma_{abs}$  from AE33 could be used  
152 as a reference for the measured BCMSD. As for the multiple scattering corrections, Zhang et al.  
153 (2018) compared the measured  $\sigma_{abs}$  measured by AE33 and by Multi-Angle Absorption Photometer  
154 (MAAP) at Tsinghua University, which is about 2 km away from our measurement site. They  
155 recommended a compensation factor of 2.6 to be used and we adopted the same factor in our study.

156 Time correction was needed because time gaps between voltages implied on the DMA (particle  
157 size) and sample particles measured by different instruments were not the same. The time correction  
158 procedures were conducted every day during the field measurement to ensure that the time deviations  
159 of the CPC and the AE51 were constrained within 2 seconds.

160 Fig. S3 gave the time series diagram of scanned aerosol diameters by DMA, measured  $\sigma_{abs}$   
161 from AE51, and the aerosol number concentrations counted by CPC. The aerosol PNSD (or  
162 size-resolved  $\sigma_{abs}$ ) could be calculated by matching the DMA diameter and the measured aerosol  
163 number concentrations (or measured  $\sigma_{abs}$  by simply using the single particle charge ratio for each  
164 electrical mobility diameter. These measured PNSD and size-resolved  $\sigma_{abs}$  did not consider the  
165 effect of multiple-charging corrections and are labeled as raw aerosol PNSD and raw aerosol  
166 size-resolved  $\sigma_{abs}$ .

### 167 **3.1.2 Multiple charging corrections of raw size-resolved $\sigma_{abs}$**

168 In the work of Ning et al. (2013) study, lots of efforts were made to evaluate the performance of  
169 the instrument. They considered the diffusion corrections and particle charging corrections. However,  
170 the particle charging corrections were limited to single particle charge ratio as they mentioned that  
171 they simplified the particle charge correction by applying the peak electrical mobility for the  
172 calculation of representative particle size for each mobility bin and single particle charge ratio for  
173 each primary mobility. They ignored the fact that the aerosol samples selected by the DMA were  
174 quasi-monodisperse with different charges and different diameters.

175 We proposed a new algorithm for the multi-charge corrections of the size-resolved  $\sigma_{abs}$ .  
176 Multi-charge corrections to the measured size distribution were prevailing when the DMA was used  
177 to scan the aerosol sizes. When the DMA and CPC are used together to measure the aerosol particle  
178 number size distribution (PNSD), the multi-charging corrected aerosol PNSD can be significantly

179 different from the raw measured one (Bau et al., 2014;He and Dhaniyala, 2013;He et al., 2015). As  
 180 shown in the results part of this study, the multi-charge corrections of the size-resolved  $\sigma_{abs}$  could  
 181 cause differences in both the magnitude and shape of the size-resolved  $\sigma_{abs}$ . Therefore, it is  
 182 necessary to perform multi-charge corrections on the size-resolved  $\sigma_{abs}$ . This study developed a new  
 183 algorithm to correct the size-resolved  $\sigma_{abs}$  from measured value based on the work of Hagen and  
 184 Alofs (2007) and Deng et al. (2011).

185 When the DMA is charged with a negative voltage, those aerosols with a small range of  
 186 electrical mobility ( $Z_p$ ) can pass through the DMA:

$$187 \quad Z_p = \frac{q_{sh}}{2\pi VL} \ln\left(\frac{r_1}{r_2}\right), \quad (5)$$

188 where  $q_{sh}$  is the sheath air flow rate; V is the average voltage on the inner center rod;  $r_1$  and  $r_2$   
 189 are the outer and inner radius of annular space respectively. The  $Z_p$  is related with  $D_p$  by  
 190 elementary charge ( $e$ ), number of elementary charges on the particle ( $n$ ), and gas viscosity poise ( $\mu$ )  
 191 with:

$$192 \quad Z_p = \frac{neC(D_p)}{3\pi\mu D_p}, \quad (6)$$

193 where  $C(D_p)$  is Cunningham slip correction:

$$194 \quad C = 1 + \frac{2\tau}{D_p} (1.142 + 0.558e^{-\frac{0.999D_p}{2\tau}}), \quad (7)$$

195 where  $\tau$  is the gas mean free path. From equation 7, aerosol particles can have the same  $Z_p$  despite  
 196 that they have different  $n$  and  $D_p$ . At the same time, there exists a relatively larger portion of  
 197 multiple charged particles for those particles with diameters between 100 nm and 400 nm when the  
 198 ambient aerosols pass through the X-ray (Tigges et al., 2015;Wiedensohler and Fissan, 1988).  
 199 Through the above discussion, the selected aerosols by DMA at a given electrical mobility can have  
 200 different charges which will correspond to different diameters.

201 When the scan diameter is set as  $D_{p_i}$  for the singly charged particles and the respective voltage  
 202 of DMA is  $V_i$  ( $i=1, 2, \dots, I$ ), aerosol particles with electro-mobility of  $Z_{p,i}$  ( $i=1, 2, \dots, I$ ) can pass  
 203 through the DMA and the observed  $\sigma_{abs}$  by AE51 can be expressed as:

$$204 \quad R_i = \int_0^\infty G(i, x)A(x)n(x)dx, \quad (8)$$

205 where  $x$  is the scale parameter, with the definition of  $x = \log(D_{p_i})$ ,  $A(x)$  is the average  $\sigma_{abs}$  of  
 206 single particle for scale parameter  $x$ , and  $n(x) = dN/d\log D_p$  is aerosol PNSD that is the multiple

207 charging corrected results from the measured aerosol PNSD. We define the kernel function  $G(i, x)$ ,  
 208 which is crucial to the algorithm, as:

$$209 \quad G(i, x) = \sum_{v=1}^{\infty} \emptyset(x, v) \Omega(x, v, i), \quad (9)$$

210 where  $\emptyset(x, v)$  is the probability of particles that are charged with  $v$  charges at the scale parameter  
 211 of  $x$  (Wiedensohler, 1988).  $\Omega(x, v, i)$  is the probability of particles that can pass through the DMA  
 212 with  $v$  charges at the scale parameter  $x$  (Knutson and Whitby, 1975). In this study, the maximum  
 213 value of  $v$  is 10.

214 The multiple charging corrections can be expressed as computing the  $A(x_i^*)$ , in which  $x_i^*$  is the  
 215 predetermined scale parameter from the DMA. To get the numerical integration results of equation 9,  
 216 the diameter interval that is 1/50 of the measured diameter is used. Thus, equation 9 can be written as

$$217 \quad R_i = \int_0^{\infty} G(i, x) A(x) n(x) dx = \Delta x_i \sum_{j=1}^{50} \beta_j G(i, x_{i,j}) A(x_{i,j}) n(x_{i,j}), \quad (10)$$

218 where  $\beta = \begin{cases} 0.5, & j = 1, J \\ 1, & \text{otherwise} \end{cases}$ ;  $x_{i,j}$  is the  $j^{\text{th}}$  ( $j=1, 2, \dots, 50$ ) parameter that locates at the parameter  $x_i$  and  
 219  $x_{i+1}$  and  $A(x_{i,j})$  ( $i=1, 2, \dots, I; j=1, 2, \dots, 50$ ), the BC mass ratio at scale parameter  $x_{i,j}$ , is expressed  
 220 as the linear interpolation of the values at the measured diameters.

$$221 \quad A(x_{i,j}) = A(x_i) + P_i(x_{i,j} - x_i), \quad (11)$$

222 where  $P_i$  is the slope of the linear interpolation result of

$$223 \quad A(x_k^*) = C + P_i \cdot x_k^*. \quad (12)$$

224  $x_k^*$  refers to these five diameters that are nearest to the predetermined scale parameter  $x_i$ .  $C$  is the  
 225 intercept of the linear interpolation result.

226 With  $H_{i,j} = \beta_j \Delta x_i G(i, x_{i,j}) n(x_{i,j})$ , equation 11 can be written as

$$\begin{aligned} 227 \quad R_i &= \sum_{j=1}^J H_{ij} [A(x_i) + P_i(x_{i,j} - x_i)] = \sum_{j=1}^J H_{ij} A(x_i) + \sum_{j=1}^J H_{ij} P_i x_{i,j} - \sum_{j=1}^J H_{ij} P_i x_i \\ 228 \quad &= \sum_{k=1}^I (\sum_{j=1}^J H_{ij} \delta(i-k)) A(x_k^*) + \sum_{k=1}^I \left( \sum_{j=1}^J H_{ij} x_{i,j} \delta(i-k) \right) P_k - \sum_{k=1}^I \delta(i-k) P_k x_k^* \\ 229 \quad &= \sum_{k=1}^I Q_{ik} A(x_k^*) + \sum_{k=1}^I T_{ik} P_k - \sum_{k=1}^I Q_{ik} P_k x_k^*, \end{aligned} \quad (13)$$

230 where  $\delta(x) = \begin{cases} 0, & x \neq 0 \\ 1, & x = 0 \end{cases}$ ,

$$231 \quad Q_{ik} = \sum_{j=1}^J H_{ij} \delta(i-k), \quad (14)$$

$$232 \quad \text{and } T_{ik} = \sum_{j=1}^J H_{ij} x_{i,j} \delta(i-k). \quad (15)$$

233 By letting the

$$234 \quad S_i = R_i - \sum_{k=1}^I T_{ik} P_k + \sum_{k=1}^I Q_{ik} P_k x_k^* \quad (16)$$

235 This equation is then expressed as

$$236 \quad S_i = \sum_{k=1}^I Q_{ik} A(x_k^*), \quad (17)$$

237 or

$$238 \quad S = QA, \quad (18)$$

239 where S and A are  $I \times 1$  vectors and Q is an  $I \times I$  matrix. This matrix can be solved by using the  
240 non-negative least square method.

241 Finally, the A(x) can be determined and the corresponding size-resolved  $\sigma_{abs}$  that is multiple  
242 charging corrected can be calculated.

### 243 **3.1.3 Transform the size-resolved $\sigma_{abs}$ into BCMSD**

244 MAC of different size range is necessary when transform the size-resolved  $\sigma_{abs}$  into BCMSD.  
245 The MAC at different size should be different. When the size-resolved  $\sigma_{abs}$  is converted into  
246 BCMSD with a constant MAC, the derived BCMSD would be biased.

247 The size-resolved MAC was calculated using the Mie scattering model (Bohren and Huffman,  
248 2007). Based on the Mie scattering theory, MAC values vary for different aerosol core diameter and  
249 different total diameter. Results from SP2 measurement show that the size distribution of the BC  
250 core diameter peaked at around 120 nm in Beijing (Zhang et al., 2017). For each aerosol diameter,  
251 the MAC value with core diameter of 120 nm was used to transform the BCASD into the BCMSD.  
252 MAC values with core diameter at  $120 \pm 15$  nm were calculated and shown in Fig. 2. From Fig. 2, the  
253 MAC varied significantly between 3.6 and 9.2  $\text{m}^2/\text{g}$ . The constant MAC values 7.7  $\text{m}^2/\text{g}$   
254 corresponded to the aerosol diameter of 269 nm. The calculated mean MAC values in Fig. 2 under  
255 different diameter were used in this study.

### 256 **3.1.4 Validation of the multiple charging corrections**

257 An example of the multiple charging corrections of the size-resolved  $\sigma_{abs}$  was shown in Fig. 3.  
258 The corrections of aerosol PNSD were based on the work of Hagen and Alofs (2007). As shown in  
259 Fig. 2(a), the corrected aerosol PNSD was significantly different from the original uncorrected one.  
260 There were about half of the measured particles have multiple elementary charge in the size range  
261 between 100 and 200 nm. The raw uncorrected aerosol PNSD had a peak value of  $10920 \text{ cm}^{-3}$  at 98  
262 nm while the corrected aerosol PNSD reached its peak value of  $8450 \text{ cm}^{-3}$  at 98 nm. The peak



263 positions of the raw aerosol particle mass size distribution (PMSD, dm/dlogDp) peaked at 322 nm  
 264 with a peak value of 86.3  $\mu\text{g}/\text{m}^3$  and the corrected aerosol PMSD had a peak value of 53  $\mu\text{g}/\text{m}^3$  at  
 265 461 nm. The peak position of the aerosol PMSD shifted a lot before and after the multiple charging  
 266 corrections. The similar case for the size-resolved  $\sigma_{abs}$  was shown in Fig. 2(b). The shape of  
 267 size-resolved  $\sigma_{abs}$  had changed substantially due to the multiple charging corrections. The  
 268 measured raw BCMSD had a peak diameter near 320 nm and the magnitude of size-resolved  $\sigma_{abs}$   
 269 plateau reached 34.3  $\text{Mm}^{-1}$ , which was in accordance with the results of Ning et al. (2013), where the  
 270 multiple charging corrections were not involved. However, the corrected size-resolved  $\sigma_{abs}$  peaks  
 271 near 410 nm, with a peak value of about 29.5  $\text{Mm}^{-1}$ . According to the result, a small amount of  $\sigma_{abs}$   
 272 remained in particles with diameter between 100nm and 200nm. The measured size-resolved  $\sigma_{abs}$   
 273 changed a lot when multiple charging corrections were implemented, which highlighted the necessity  
 274 of implementation of appropriate multiple charging corrections

275 The  $\sigma_{abs}$  integrated from measured size-resolved  $\sigma_{abs}$  changed after multiple charging  
 276 corrections. Fig. S4 showed the comparison results of the  $\sigma_{abs}$  measured by AE33 and the  
 277  $\sigma_{abs}$  integrated from AE51 measurements. The  $\sigma_{abs}$  integrated from uncorrected and corrected  
 278 size-resolved  $\sigma_{abs}$  versus  $\sigma_{abs}$  measured by AE33 were shown in Fig.S4(a) and Fig.S4(b),  
 279 respectively. Before multiple charging corrections, the  $\sigma_{abs}$  from uncorrected size-resolved  $\sigma_{abs}$   
 280 increased linearly with the  $\sigma_{abs}$  from AE33, with  $R^2$  equaling 0.87, but it was 2.37 times that of  
 281 AE33 in average. As a comparison, overall magnitude of  $\sigma_{abs}$  integrated from corrected  
 282 size-resolved  $\sigma_{abs}$  agreed better with that from AE33 with a slope of 1.2. With the discussion above,  
 283 multiple charging corrections were essential for size-resolved  $\sigma_{abs}$  and BCMSD measurements.

### 284 3.2 Fitting the BCMSD by using two log-normal models

285 Based on the measurement results, the BCMSD had two modes for most of the conditions. The  
 286 BCMSD are assumed to be of two log-normal distributions as:

$$287 \quad m_{fit,Dp} = \sum_{i=1,2} \frac{m_i}{\sqrt{2\pi\log(GSD_i)}} \cdot \exp\left(-\frac{[\log(D_p)-\log(D_{m,i})]^2}{2\log^2(GSD_i)}\right), \quad (19)$$

288 Where  $D_p$  is the diameter of the aerosols;  $m_i$  is the mass of mode  $i$  ( $i=1,2$ );  $GSD_i$  is the geometric  
 289 standard deviation at mode  $i$  ( $i=1,2$ ), and  $D_{m,i}$  is the geometric mean diameter of the mode  $i$  ( $i=1,2$ ).  
 290 The  $GSD_i$  and  $D_{m,i}$  can be determined by using the least square method with the objective function  
 291 as :

292 
$$J = \sum_{i=1,n} (m_{Dp_i} - m_{fit,Dp_i}(D_{m1}, GSD_1, D_{m2}, GSD_2))^2, \quad (20)$$

293 Where  $m_{Dp_i}$  is the measured mass distribution at  $Dp_i$ , while  $m_{fit,Dp_i}$  is the fit mass distribution at  
 294  $Dp_i$ .

### 295 **3.3 Estimating aerosol optical properties with different BCMSD**

296 The Mie scattering model was used to study the influence of the BCMSD variation on the  
 297 aerosol optical properties. When running the Mie model, aerosol PNSD and BC were necessary. In  
 298 this study, The BCMSD was assumed to be log-normal distributed.  $D_m$  of the BCMSD was set to  
 299 vary from 100 nm to 600 nm. Geometric standard deviation (GSD) of the BCMSD was set to be in  
 300 the range between 1.3 and 1.8. BC was treated as partially externally mixed and the remaining  
 301 aerosols was treated as core-shell mixed. The ratio of externally mixed  $m_{BC}$  to core-shell  $m_{BC}$  was  
 302 determined by the method introduced in Ma et al. (2012) and a mean ratio of 0.51 was used. The  
 303 density and refractive index of BC were set as 1.5 g/cm<sup>3</sup> and 1.8+0.54i (Kuang et al., 2015),  
 304 respectively. The complex refractive index of non-absorbing aerosols was 1.53+10<sup>-7</sup>i (Wex et al.,  
 305 2002) at the wavelength of 525 nm. More details of calculating the aerosol optical properties by  
 306 using the aerosol PNSD and BCMSD, can refer to Kuang et al. (2016).

307 The aerosol PNSD and  $m_{BC}$  used here is the mean result of aerosol PNSD and  $m_{BC}$  over the  
 308 whole field measurement respectively. The amount of BC particle adopted in this study is the mean  
 309 value of the  $m_{BC}$  measured by AE33. For each BCMSD, extinction coefficient ( $\sigma_{ext}$ ), the scattering  
 310 coefficient ( $\sigma_{sca}$ ), the single scattering albedo (SSA), and the asymmetry factor ( $g$ ) could be obtained  
 311 from the output of Mie scattering model.

### 312 **3.4 Evaluating the DARF with different BCMSD**

313 In this study, the SBDART model (Ricchiuzzi et al., 1998) was employed to estimate the DARF.  
 314 In our study, the instantaneous DARF for cloud free conditions at the top of atmosphere was  
 315 calculated for irradiance wavelength range from 0.25 to 4  $\mu$ m. Input of the model required the  
 316 profiles of aerosol  $\sigma_{ext}$ , SSA,  $g$ . These profiles were calculated from the parameterization of the  
 317 aerosol vertical distributions. Details of calculating the  $\sigma_{ext}$ , SSA and  $g$  profiles can refer to part 4  
 318 in the supplementary material. In brief, the aerosol  $\sigma_{ext}$ , SSA and  $g$  profiles were calculated  
 319 based on the given aerosol PNSD and BCMSD. The DARF can be estimated using the above aerosol  
 320 optical profiles.

321 The aerosol optical properties and the corresponding aerosol optical profiles vary with different  
322 BCMSD. Then the DARF should be different for different BCMSD. By estimating the DARF using  
323 different aerosol BCMSD, the influence of BCMSD on the aerosol radiative properties can be  
324 studied.

## 325 **4 Results and Discussions**

### 326 **4.1 Measurement results of the BCMSD**

327 The time series of measured PM<sub>2.5</sub>, aerosol PNSD and BCMSD were shown in Fig. 3. During  
328 the observation period, the PM<sub>2.5</sub> varied from 0.06 to 220  $\mu\text{g}/\text{m}^3$ , with a mean value of  $71.5 \pm 52.56$   
329  $\mu\text{g}/\text{m}^3$ . Three periods of heavy PM<sub>2.5</sub> loading were observed: (1) PM<sub>2.5</sub> increased from around 100  
330  $\mu\text{g}/\text{m}^3$  to 200  $\mu\text{g}/\text{m}^3$  and decreased slowly to 1  $\mu\text{g}/\text{m}^3$  in the period 21-26, March; (2) the PM<sub>2.5</sub>  
331 accumulated slowly from 28 to 30, March and dissipated quickly from 30, March to 1, April; (3) the  
332 rapid accumulation and dissipation of PM<sub>2.5</sub> happened during 2 to 5, April. During the last five days,  
333 PM<sub>2.5</sub> fluctuated between 20 and 120  $\mu\text{g}/\text{m}^3$ . For each pollution condition, both the aerosol total  
334 number concentrations and the aerosol median diameter increased. The aerosol median diameter  
335 varied between 31 nm and 169 nm with a mean value of  $78 \pm 31$  nm.

336 Our measurements shew that the BCMSD had two modes with the coarser mode ranging  
337 between 430 nm and 580 nm in mobility diameter. Many field measurements had revealed that most  
338 of the BC mass locates in the aerodynamic diameter range of 320 nm and 560 nm using the MOUDI  
339 (Hu et al., 2012;Huang and Yu, 2008). When the aerodynamic diameter was transformed into  
340 mobility diameter with assumption a aerosol effective density of 1.3, the measured BC aerodynamic  
341 diameter range corresponded to mobility diameter range of 280 nm and 491 nm. Therefore, the  
342 measured size range for coarser mode of BCMSD agreed well with the previous measurement.

343 The measured aerosol in the field site was representative of the urban aerosol. The BC particles  
344 emitted by vehicles contributed significantly to the total aerosol BC mass. These BC particles were  
345 rarely coated or thinly coated, and the BC core diameter peaked around 120 nm (Zhang et al., 2017).  
346 Therefore, the BCMSD of the smaller mode measured in our study corresponded to these uncoated  
347 of thinly coated particles.

348 The total  $m_{BC}$  measured by AE33 ranged from 0.1 to 14  $\mu\text{g}/\text{m}^3$  with an average of  $5.04 \pm 2.64$   
349  $\mu\text{g}/\text{m}^3$ . Good consistence was achieved between  $m_{BC}$  measured by AE33 and  $m_{BC}$  calculated from

350 measured BCMSD as shown in Fig. 3(d).

## 351 **4.2 Evolution of the BCMSD under different polluted conditions**

352 Log-normal distribution was used to fit each mode of the BCMSD by using the least square  
353 method as introduced in section 3.2. For each mode, the geometric mean diameter ( $D_m$ ) and the  
354 geometric standard deviation (GSD) of the BCMSD were studied.

355 During the measurement period, both  $D_m$  and GSD of the two modes had changed significantly  
356 as shown in Fig S7. The  $D_m$  of first and second mode varied from 139 to 161 nm and from 420 to  
357 597 nm, respectively. The corresponding mean  $D_m$  was 151 and 520 nm. The  $D_m$  of the two  
358 modes was found to be positive correlated in Fig. S7(a). When the pollution was released from the  
359 beginning to 27, March, the  $D_m$  decreased from 597 to 420 nm and from 160 to 140 nm for the  
360 coarser mode and the smaller mode respectively. The BC containing aerosols tended to be aged and  
361 grew larger when the air surrounding get polluted.

362 GSD for the coarser mode and the smaller mode showed very different properties as shown in  
363 Fig. S7(b). For the second mode, GSD varied from around 1.49 to 1.68 with a mean value of 1.57.  
364 The GSD get decreased with the pollution condition, which indicated that BC containing aerosols  
365 tend to accumulate to a small range of diameters during the aging processing. This phenomenon was  
366 consistent with the fact that larger particles grew relative slower in diameter because the growth ratio  
367 of small aerosol particle is proportion to the negative power of it's diameter. For the first mode, GSD  
368 ranged from 1.41 to around 1.86 with a mean value of 1.63. However, GSD of the smaller mode tend  
369 to be larger when the surrounding air get cleaner, which might be related to the complex sources of  
370 the BC emission. A small amount of fresh emitted BC particles can have substantial influence on the  
371 mass size distribution of the smaller mode because the BC concentrations of the smaller mode were  
372 small, especially under clean conditions. In general, the GSD of coarser mode was a good indicator  
373 of the BC aging process and that of the smaller mode could partially reflect the complex sources of  
374 the BC fine particles.

375 The relationship between the  $D_m$  and the GSD for coarser mode was further analyzed by  
376 analyzing the distribution of the  $D_m$  and GSD. The GSD and  $D_m$  had opposite trends as shown in  
377 Fig 5. With the increment of the  $D_m$  from 420 to 540 nm, the mean value of GSD decreased from  
378 around 1.608 to 1.528 while the  $m_{BC}$  increased with the  $D_m$ . The statistical relationship between  
379  $D_m$  and GSD offered a reasonable representation of the BCMSD under different polluted conditions.

380 In the following work, mean values of the GSD at different  $D_m$  were used to for further discussion.  
381 The  $m_{BC}$  and GSD is positively correlated. The  $m_{BC}$  increased from 2.4 to 8.3  $\mu\text{g}/\text{m}^3$  when the  
382  $D_m$  increased from 420 to 540 nm.

383 Note that the GSD get slightly increased with the increment of  $D_m$  when  $D_m$  was larger than  
384 520 nm. This might be caused by the limit diameter range of BCMSD measuring system which was  
385 from 20 to 680 nm. The multiple charge corrections applied to the BCMSD could influence the  
386 BCMSD when  $D_m$  of the BCMSD was near the end of the scanned diameter and may lead to  
387 significant uncertainties to the BCMSD. The measurement results indicated that cases of measured  
388  $D_m$  of BCMSD larger than 520 nm were few, demonstrating that this multiple correction effect  
389 influenced little on shape of measured BCMSD in most cases.

#### 390 **4.3 Influence of BCMSD variation on the aerosol optical properties**

391 The aerosol optical parameters using the measured mean aerosol PNSD and mean  $m_{BC}$   
392 corresponding to different GSD and  $D_m$  values were shown in Fig. 6. In Fig. 6(a), the aerosol  $g$   
393 varied from 0.603 to 0.627 (variation of 4%). Recent work by Zhao et al, 2017 showed that the  
394 aerosol  $g$  value in the NCP may vary at a range of 10% due to the change of aerosol PNSD. Aerosol  
395  $g$  was more sensitive to  $D_m$  when the geometric mean diameter of the BCMSD was lower than 400  
396 nm. However, when the  $D_m$  was larger than 400 nm, the  $g$  become sensitive to both the  $D_m$  and  
397 the GSD of BCMSD. Overall, the  $g$  varied a little bit (0.02 to 0.609) under the representative  
398 conditions during the measurement period. For the aerosol SSA, it was sensitive to the  $D_m$  over the  
399 whole range as shown in Fig. 6(b). SSA varied between 0.90 and 0.94 under the representative  
400 measurement conditions. The  $\sigma_{\text{sca}}$  had large changes from 325.6  $\text{Mm}^{-1}$  to 364.4  $\text{Mm}^{-1}$ . The  $\sigma_{\text{sca}}$  was  
401 quite sensitive to variations in BCMSD when the  $D_m$  was larger than 450 nm as shown in Fig.6c. In  
402 addition, variations in  $\sigma_{\text{sca}}$  relied more on the variations in  $D_m$  when  $D_m$  was lower than 400 nm.  
403 Within the measurement conditions of BCMSD, the  $\sigma_{\text{sca}}$  varied from 328  $\text{Mm}^{-1}$  to 345  $\text{Mm}^{-1}$ . The  
404 measured GSD under different  $D_m$  went along with the gradient direction of the  $\sigma_{\text{sca}}$ , which mean  
405 that the evolution of BCMSD in the atmosphere influenced substantially on  $\sigma_{\text{sca}}$ . As for the  $\sigma_{\text{abs}}$ , it  
406 changed from 24.06  $\text{Mm}^{-1}$  to 37.27  $\text{Mm}^{-1}$  and the corresponding mass absorption cross section (MAC)  
407 was estimated to be in the range of 5.44 to 8.08  $\text{m}^2/\text{g}$ , suggesting that MAC of the BC aerosols  
408 should be carefully studied under different BCMSD conditions.

#### 409 **4.4 Influence of BCMSD on the direct aerosol radiative forcing**

410 The estimated DARF values for different GSD and  $D_m$  conditions were estimated. When  
411 estimating the DARF, the measured mean aerosol PNSD and mean BC mass concentration were used.  
412 The results of estimated DARF were shown in Fig. 7(a). DARF at the surface varied from  $-4.3 \text{ w/m}^2$   
413 to  $-3.59 \text{ w/m}^2$  for different BCMSD. Within the measured BCMSD range, the DARF varied from  
414  $-3.97 \text{ w/m}^2$  to  $-3.67 \text{ w/m}^2$ , which corresponding to 8.45% of variation. The heating rate within the  
415 mixed layer was a powerful indicator of the BC particles' absorbing effects, which may help evaluate  
416 the development of the boundary layer. The calculated mean heating rate within the mixed layer  
417 changed from 2.16 K/day to 2.65 K/day for different  $D_m$  and GSD, as shown in Fig. 7(b). The  
418 heating rate with the measured BCMSD range could change from 2.24 to 2.50 with a variation of  
419 11.6%.

420 Mixing states of BC play significant roles in calculations of aerosol optical properties and  
421 estimations of DARF (Jacobson, 2001). As a comparison, we estimated the DARF under different  
422 conditions of BC mixing state: (1) internally mixed, (2) externally mixed and (3) core-shell mixed.  
423 Table 1 gave the estimated DARF and mean heating rate within the mixed layer under different  
424 mixing state conditions. Results showed that the DARF under different BC mixing states conditions  
425 may change by 10.50%, which shared the same magnitude with 8.45% variation of DARF caused by  
426 BCMSD variations. In addition, the heating rate was estimated to vary by 9.71%. These results  
427 highlighted that the BCMSD plays significant roles in variations of aerosol optical properties and  
428 estimations of DARF as well as the air heating rate caused by the existence of BC particles. It was  
429 recommended that a real time measured BCMSD be used when estimating the aerosol DARF, instead  
430 of a constant one. The BCMSD was as important as that of the BC mixing states.

## 431 **5 Conclusions**

432 Knowledge of the BC microphysical properties especially the size-dependent information can  
433 help reduce the uncertainties when estimating the aerosol radiative effects. BCMSD is an important  
434 quantity in its own right, being directly and indirectly applicable to determination the sources, aging  
435 processes and mixing states of BC aerosols. In this study, the characteristics of BCMSD were studied  
436 from the field measurement results by using our own developed measurement algorithm.

437 The BCMSD measurement system was developed and validated based on the works of Ning et  
438 al. (2013) by using differential mobility analyzer (DMA) in tandem with Aethalometer (AE). When  
439 deriving the BCMSD, a comprehensive multiple charging correction algorithm was proposed and

440 implied. This algorithm was validated by closure study between the measured total  $m_{BC}$  from AE33  
441 and the  $m_{BC}$  integrated from the measured BCMSD using the datasets from field measurements.  
442 Results showed that the multiple charging corrections could significantly change the shapes and  
443 magnitudes of the raw measured BCMSD. The accurate BCMSD characteristics could be obtained  
444 by our proposed method in this paper.

445 The developed measurement system was employed in a field campaign in the North China Plain  
446 from 21 March to 9 April in 2017. The BCMSD was found to have two quasi-lognormal modes with  
447 peaks at around 150 nm and 500 nm, respectively. These two modes were consistent with the  
448 previous measurement results by MOUDI (Wang et al., 2015; Hu et al., 2012). The amount of the BC  
449 mass concentrations for the coarser mode peaks were about twice to that of the smaller mode.

450 The characteristic of the BCMSD was studied by fitting the shape of BCMSD with a bi-normal  
451 distribution. The relationships between the fitted  $D_m$  and GSD were statistically studied. During the  
452 aging processing, the opposite trends for the  $D_m$  and GSD were found for coarser mode. This is the  
453 first time that the coarser mode of the BCMSD were synthetically studied. The BCMSD of coarser  
454 mode varied significantly under different pollution conditions with peak diameter changed between  
455 430 and 580 nm. However, the relationship between the  $D_m$  and GSD for smaller mode BC aerosols  
456 were more complex due to the complex sources.

457 When the BCMSD were changed with the polluted condition, the corresponding aerosol optical  
458 properties changes significantly. Sensitivity studies found that the aerosol  $g$  varies from 0.603 to  
459 0.627 due to the variations in BCMSD. Aerosol  $g$  was more sensitive to  $D_m$  when the geometric  
460 mean diameter of the BCMSD is in the range of 300 nm and 370 nm. The SSA can changed from  
461 0.90 to 0.94. The  $\sigma_{sca}$  experienced significant change with the variation of BCMSD from  $325.6 \text{ Mm}^{-1}$   
462 to  $364.4 \text{ Mm}^{-1}$  and the  $\sigma_{abs}$  changed in the range between  $24.064 \text{ Mm}^{-1}$  and  $37.27 \text{ Mm}^{-1}$ . The  
463 corresponding BC MAC was calculated to be in the range between 5.44 and  $8.08 \text{ m}^2/\text{g}$ .

464 The variations in DARF were estimated due to the variations of the BCMSD by using the  
465 SBDART model. Results showed that the DARF can varies by about 8.45% for different BCMSD  
466 and the heating rate for different measured BCMSD conditions could change from 2.24 to 2.50,  
467 corresponding to a variation of 11.6%. At the same time, the variations in DARF due to the  
468 variations in the BC mixing state was estimated to be 10.5% and that of the heating rate is 8.45%.  
469 Thus, the variations of the BCMSD may had significant influence on the aerosol radiative budget and

470 an accurate measurement of BCMSD was very necessary.

471

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

473 **Data availability.** The data used in this study is available when requesting the authors.

474 **Author contributions.** GZ, CZ, JT and YK designed and conducted the experiments; CS, YY, CZ and GZ  
475 discussed the results.

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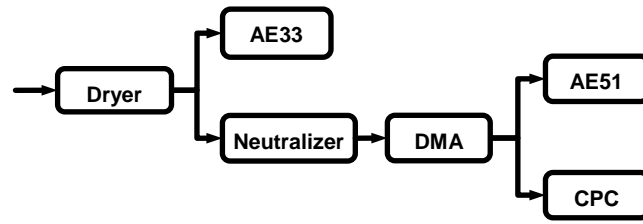
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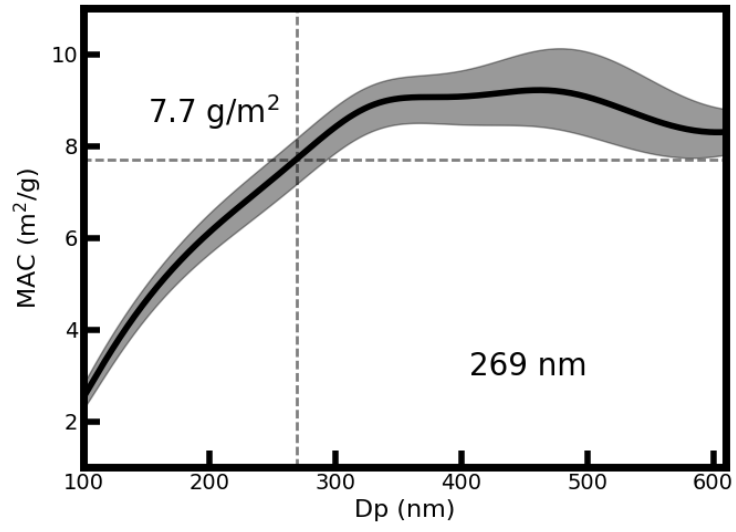


**Figure 1.** The schematic diagram of the instrument setup.

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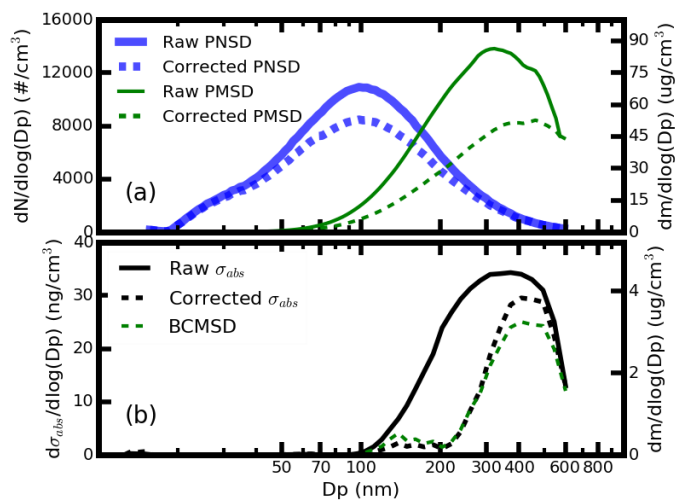


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735 **Figure 2.** Calculated mass absorption coefficient of different aerosol.

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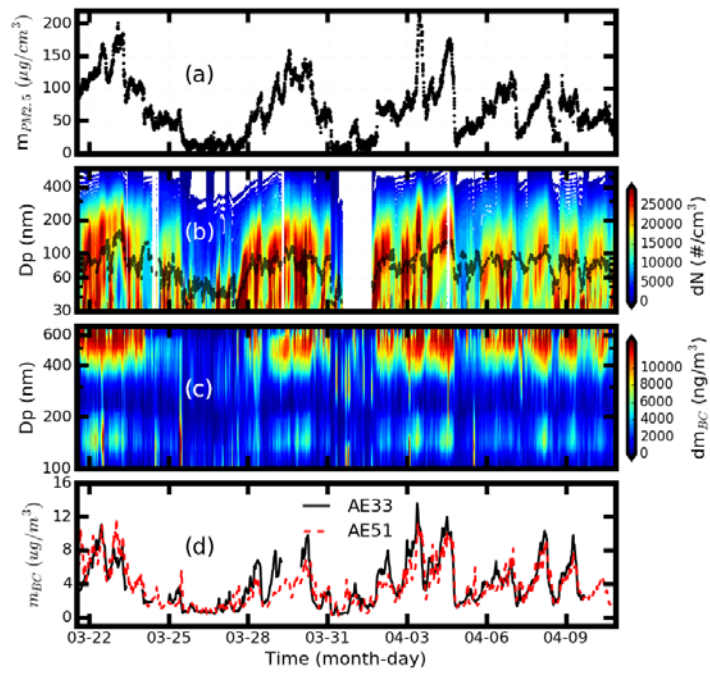
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739 **Figure 3.** Case of multiple charging correction processing. (a) the multiple charging correction of the  
 740 aerosol PNSD and aerosol PMSD, (b) the multiple charging correction of the size-resolved  $\sigma_{abs}$ .

741 The solid line is the measured results without multiple charging corrections and the dotted line is the  
 742 multiple charging corrections results.

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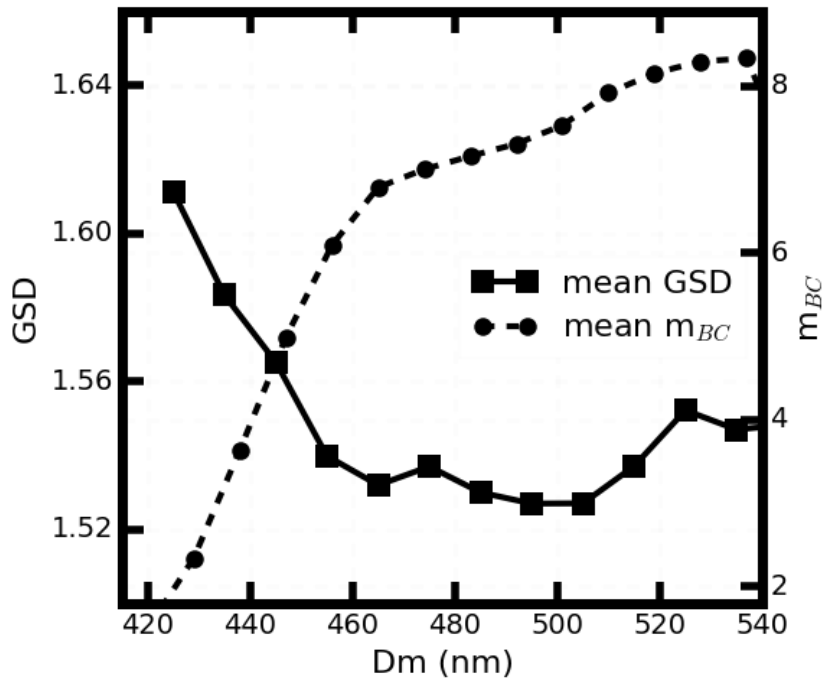
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**Figure 4.** The measured time series of mass concentrations for (a) the PM<sub>2.5</sub>; (b) the aerosol PNSD in filled color, the geometric median diameter in dotted line;(c) the BCMSD and (d) the  $m_{BC}$  by AE33 (black) and  $m_{BC}$  from integrated BCMSD from AE51 (red).

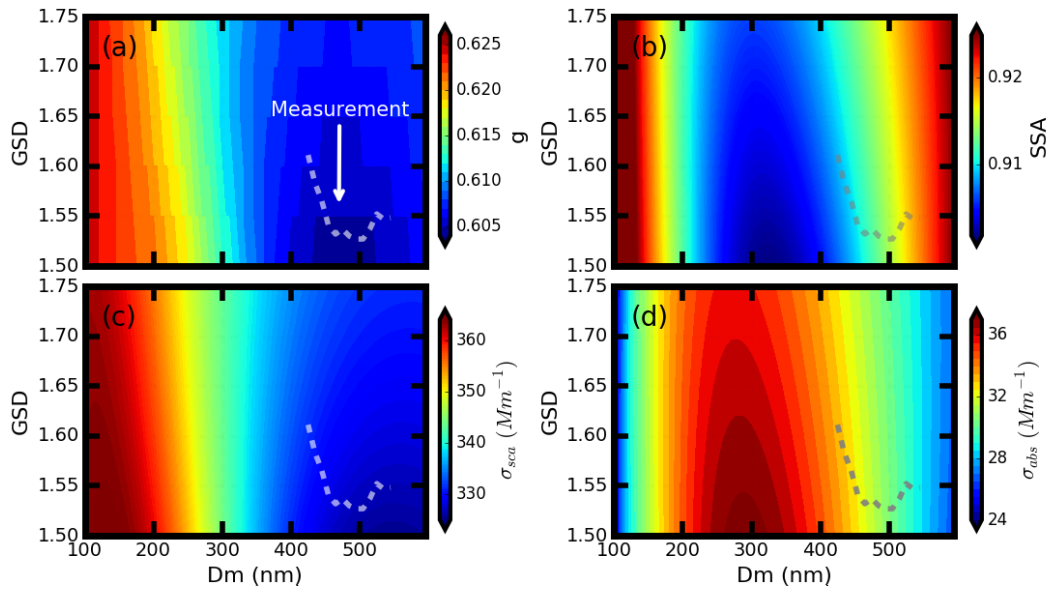
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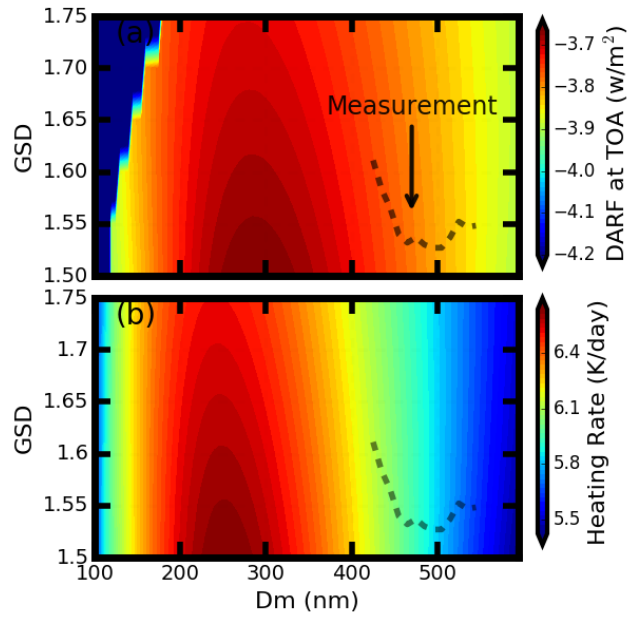
753 **Figure 5.** The relationship between the Dm and the GSD. The black dots show the real measured Dm  
754 and GSD. The black line shows the mean results of the GSD for different Dm. The black line marked  
755 with square shows the variation of mean  $m_{BC}$  with the Dm.

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757 **Figure 6.** Variations of aerosol optics properties using the measured mean aerosol PNSD and  $m_{BC}$   
 758 under different BCMSD conditions, which are represented by different Dm and GSD values: (a)  
 759 aerosol asymmetry factor, (b) single scatter albedo, (c) scattering coefficient and (d) extinction  
 760 coefficient . The grey dotted line in the figure shows the evolution path of the BCMSD according to  
 761 results of field measurements.

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764 **Figure 7.** Variations of (a) DARF and (b) heating rate under different BCMSD conditions, which are  
 765 represented by different Dm and GSD values. The black dotted line in the figure shows the evolution  
 766 path of the BCMSD according to results of field measurements.

767

768 **Table 1.** Comparison of the DARF and heating rate values under different BC mixing states and  
 769 different BCMSD conditions.

		Mixing State			BCMSD	
		Internal	External	Core-Shell	Minimum	Maximum
<b>DARF</b>	<b>Value(w/m<sup>2</sup>)</b>	<b>-3.45</b>	<b>-3.56</b>	<b>-3.81</b>	<b>-3.97</b>	<b>-3.67</b>
	<b>Variation</b>	<b>10.5%</b>			<b>8.45%</b>	
<b>Heat Rate</b>	<b>Value(K/day)</b>	<b>2.51</b>	<b>2.32</b>	<b>2.53</b>	<b>2.24</b>	<b>2.50</b>
	<b>Variation</b>	<b>9.71%</b>			<b>11.6%</b>	

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