

Interactive comment on “A novel study of the morphology and effective density of externally mixed black carbon aerosols in ambient air using a size-resolved single-particle soot photometer (SP2)” by Yunfei Wu et al.

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The paper by Wu et al. titled "A novel study of the morphology and effective density of externally mixed black carbon aerosols in ambient air using a size-resolved single particle soot photometer (SP2)" presents measurements of the mass of rBC particles with known mobility diameters, sampled from the atmosphere of urban Beijing. A selected portion of the measurements (the most common masses for a given diameter) are interpreted in terms of two parameters commonly used in the soot community, the effective density and mass-mobility exponent (erroneously called the fractal dimension

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by the authors). The remainder of the measurements (the shape of the overall distribution) are not interpreted.

The manuscript as submitted represents an intelligent and detailed analysis of one aspect of the data set. However, it is not a complete analysis as discussed below, and the comprehensiveness of the work could be significantly improved. Moreover, the conclusions that the authors reach are in some ways predetermined by the analysis method. Therefore, the conclusions are misleading and the manuscript should be substantially revised. The data set shows significant promise, but before publication in AMT needs to be re-analyzed by asking the question, "what can we learn from these measurements?" instead of "how can we calculate common soot diagnostics from these measurements?"

As I explain below, the authors have accidentally only analyzed uncoated particles. It is not clear whether coated particles could be separated from multiply-charged particles using this method. Therefore, I have recommended rejection unless the authors can show that the problems of restructuring/coating and multiply charging can be separated. With this potential improvement, the paper might become a substantial contribution to the literature.

We greatly appreciate the reviewer for providing very constructive comments which have helped us improve the paper. We have considered the comments carefully and revised the manuscript accordingly, as detailed below in our point-to-point responses to the specific comments.

Major comments

The authors have clearly thought carefully about their data and performed a careful analysis. The DMA was stepped instead of scanned, which avoids problems of data inversion otherwise associated with tandem DMA setups. My major comments are:

1. Limitations of the gaussian fitting The authors have performed gaussian fitting to the

number distributions of rBC-mass-per-particle measured by the SP2 after the DMA. But a huge part of this distribution is not described by the gaussian fit. From my estimation about 50% of particles are not described, at smaller masses. This needs to be addressed quantitatively and seriously in the analysis.

The first hypothesis for the non-gaussian shape is multiple charging. The authors blame this on the distribution change in Section 3.2. This is possibly important. But also important would be restructuring due to coatings. Larger rBC particles can have smaller mobility diameters (D_m) after condensation of coatings (citations were given by the authors already). This would cause a tail to the right of the mode in Figure 2, as observed.

Reply: The particles selected by the DMA at a given voltage are generally quasi-monodisperse instead of having a single mobility diameter due to the transfer function of DMA. Thus, the peak diameter is used to represent the typical diameter of the particles selected by the DMA at the given voltage. In this study, we intend to obtain the typical mass of the externally mixed BC (extBC) particles selected by the DMA at a given voltage. Therefore, the masses corresponding to the peaks of the number distributions of rBC-mass-per-particle were required. In our original analysis, we investigated the frequency distribution of extBC mass at each prescribed mobility to obtain the peak extBC mass. However, the peak extBC mass directly obtained from the frequency distribution depends on the bin-size set in the frequency calculation. Thus, the Gaussian fitting was employed to identify the peak extBC mass with fewer artificial uncertainties.

In current study, only the peak locations of the frequency distribution of extBC mass are required. We don't concern about the practical shape of the distribution. In our original analysis, we also found that a considerable fraction of the extBC masses cannot be characterized by the Gaussian distribution, e.g., at the right tail of the distribution and the extremely small masses. The right tail beyond the Gaussian distribution can be partly interpreted by the effects of multi-charged BC particles as we discussed in the

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manuscript. An extBC particle having double or more charges has a larger mass than the singly charged extBC. Fig. R1 shows the frequency distribution of the incandescence peak height values of Aquadag (a representative of bare rBC for the SP2 incandescence calibration) detected by the SP2, which are proportional to the rBC masses. The Aquadag particles were well mixed in the pure water and generated by an aerosol nebulizer, and then delivered to the SP2 for measurement after passing through a diffuse dryer. Thus, the Aquadag particles were mostly without coating and considered extBC. In this case, the doubly charged Aquadag particles exhibit a clear minor peak after the major one at a given mobility. The Gaussian fitting for the major peak can distinctly separate the singly charged particles from those with double or more charges. The multicharged Aquadag particles account $\sim 50\%$ of the total incandescence particles detected by the SP2 in the range of 140–250 nm in mobility diameter (d_{mob}). The proportion gradually decreases to only a few percentage for particles larger than 600 nm d_{mob} .

However, in the case of measurement for ambient particles, it is more complex. First, the time-delay approach used in this study can only distinguish the thickly coated BC particles from the BC particles with thinly or without coating. A fraction of thinly and/or even moderately coated BC particles cannot be absolutely separated from the bare BC particles. Thus, a fraction of extBC particles claimed in this study might be thinly-coated BC particles. We propose the right tail of the distribution is also associated with the thinly coated BC particles. These thinly-coated BC particles have relatively more compact structures, resulting in larger masses than the bare BC particles at a given mobility.

As for the distribution concentrated at the smaller extBC masses, we originally considered it was resulted from the detection noises of the SP2 for the smaller BC particles. As claimed by the manufacture, the lower BC detection limit is ~ 0.3 fg per particle. Thus, data filter was performed by eliminating the particles with incandescence signal peak heights lower than 100. This criterion is determined from the calibration curve

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by using Aquadag particles (Fig. S3 in the revised supplemental file). However, the distributions concentrated at the smaller masses still exist even when the data filter is processed. Initially, we suspected the smaller masses belonged to the thickly coated BC particles which were not separated from the extBC particles using the time-delay method. However, we found that the smaller masses also existed for the bare BC particles (i.e., Aquadag). As shown in Fig. R1, the particles with these smaller masses accounted for only 1~9% of the total particle number (after data filter by eliminating the signal with incandescence peak value lower than 100) in the 140–200 nm dmob range. This proportion gradually increased to ~37% at 700 nm mobility. Thus, these smaller masses should not be resulted only from the ‘remnant’ BC particles with thick coatings. We still attribute these smaller rBC masses to the measurement noises of the SP2.

In our revised manuscript, we make an improvement in data processing by investigating the number size distribution of the rBC core in extBC particles ($dN/d\log D_c$, where D_c is the mass-equivalent diameter of rBC core by assuming it is a void-free sphere with 1.8 g cm^{-3} density, N is the number concentration in a given D_c range) instead of the frequency distribution of extBC mass at each prescribed mobility (Fig. 2 and Fig. S6 in the revision). This improvement would reduce the artificial uncertainties in the determination of extBC masses roughly obtained from the frequency distribution. The effect of detection noises at the small rBC masses also appears to be weakened by using the number size distribution. The size distributions at different mobility sizes are generally used in the studies of size-resolved BC particles. For instant, Zhang et al. (2016) measured the morphology and density of internally mixed BC (In-BC for short in the reference) with SP2 and VTDMA. They used the peak diameter of the normalized volume size distribution of the In-BC core to represent the typical diameter of In-BC core at the prescribed dmob selected by the DMA.

As shown in Fig. 2 and Fig. S6 in the revision, a minor peak at the right side of the major peak can also be observed at a given mobility, especially in the range with dmob smaller than 400 nm. The minor peak is related to both the multiply charged extBC

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and the thinly coated BC particles which were also recognized as extBC by using the time-delay discrimination. To examine the effect of delay time threshold employed to discriminate extBC on the determined typical extBC mass, normalized number size distributions of extBC distinguished with delay times $< 2.0 \mu\text{s}$ (red), $< 1.2 \mu\text{s}$ (green) and $< 0.4 \mu\text{s}$ (blue) are compared (Fig. S6 in the revised supplemental file). Reducing the delay time threshold results in a significant reduction in the data volume that used to calculate the distribution (Fig. S5 in the revised supplemental file) and also a reduction in the extBC number fraction at the right tail of the size distribution (more significant when delay time threshold decreases to $0.4 \mu\text{s}$). Reducing the delay time threshold seems to affect little on the peak location of the size distribution, which is considered the typical mass-equivalent diameters (dme), in turn the typical mass of extBC at a given mobility. The discrepancies in the lognormal-fitted peak dme by using different delay time thresholds don't exceed 3% in the current study. Similar discussion is presented in detail in Section 3.2 of our revised manuscript. It implies the typical dme of extBC obtained from the peak dme of the number size distribution of the rBC core in extBC particles at each mobility is reliable in our study.

Fig. R1 Frequency distributions of incandescence peak height detected by the SP2 for size-selected Aquadag particles at the prescribed mobility sizes. The Gaussian fitting is performed for the major peak of each distribution to obtain the typical incandescence peak height at the corresponding mobility size.

The hypothesis of coatings means that the authors' selection of the mode diameter resulted in their analysis of fresh, uncoated particles only. Therefore it is no surprise that the results indicate consistency with literature reports of fresh, uncoated particles. Therefore, the authors' results, conclusions and abstract must be rewritten.

Reply: The initial purpose of our experimental setup is to study the microphysical and optical properties of size-resolved BC aerosols, including the sizes, mixing states and their impacts on light absorption at different mobility sizes, as well as the morphology and effective density of extBC aggregates that presented in the current study. Similar

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experimental setup for the measurement of ambient BC particles can also be found in the recent literature (e.g., Zhang et al., 2016; Raatikainen et al., 2017).

Indeed, the morphology of BC aggregates has been widely studied by using different techniques, including transmission electron microscopy (TEM) and tandem measurements (e.g., DMA-APM). The morphology of diffusion-limited cluster aggregation (DLCA), to which the BC aggregates belong, has even been well documented in the review literature (Sorensen, 2011). However, as mentioned in our revised introduction, previous tandem measurements generally investigated the morphology and density of BC aggregates at small mobility sizes with the d_{mob} not exceeding 350 nm. It is likely due to the large uncertainties in the measurement for larger particles which were less abundant in the atmosphere. In our study, we use a DMA-SP2 tandem system to study the mass-mobility relationship in a much larger mobility size range (140–750 nm) based on the sensitive and accurate SP2 measurement. The morphology and effective density determined in the relatively larger size range (140–750 nm) is compared to those in the smaller range (\sim 50–350 nm) in the literature to examine the applicability of mass-mobility relationship established at the smaller range in previous tandem measurements. In addition, variation in the morphology and effective density of ambient extBC aggregates in the atmosphere is also studied by comparing the mass-mobility relationship in a polluted episode to that a clean period. Although the DMA-SP2 is not a novel system (Gysel et al., 2011, 2012; Zhang et al., 2016; Raatikainen et al., 2017; and other associated references cited in our revised manuscript), this tandem system is seldom used to study the microphysical properties of ambient extBC aggregates which are also abundant in the atmosphere. Especially, similar studies are deficient in urban Beijing, where the particulate pollution is severe in recent years.

We have modified our manuscript substantially. Almost all the sections are rewritten.

It would be very interesting, for example, if the fraction of restructured particles could be separated from the fraction of multiply charged particles. This is also very difficult and may be impossible. I am not certain that it is impossible, but a very convincing

argument would be required to show that the two problems could be separated.

Reply: It is very difficult or even impossible to separate the fraction of restructured particles from the fraction of multiply charged particles. We have no idea how to deal with this issue, at least at the current stage. However, this problem should not have substantive impact on the results presented in current study. As mentioned above, the peak dme (i.e., mass) of the number size distribution of the rBC core in extBC particles were required and considered the typical dme (i.e., mass) of extBC at the prescribed mobility. Both the fractions of restructured particles due to thin coating and multicharged particles should only affect the shape of the distribution, e.g., cause an increase in the number size distribution at its right tail. They have few impacts on the peak location of the distribution as presented above by examining the effect of delay time threshold chosen to discriminate extBC particles.

I would like to note that the hypothesis of multiple charging would mean that smaller Dm should have a smaller fraction of total SP2 measurements explained by the Gaussian fit (since there are more pre-existing particles available to become multiply-charged in the DMA). From my inspection of Figure 2 I do not see a strong trend with Dm. This makes me suspect that coatings are involved, but is not strong enough evidence for the authors to interpret the data as such.

Reply: The frequency distribution of the rBC mass in extBC particles has been replaced by the size distribution, because the frequency distribution is too rough to presented in the scientific manuscript while the size distribution ($dN/d\log D$) is more practical. As presented in the responses above, we have explained the reasons of the high frequency at smaller rBC masses and attributed it to the measurement noises of the SP2. These smaller rBC masses influence little of the normalized number size distribution. Meanwhile, the increase in the size distribution at its right tail is interpreted as the co-effects of multicharged extBC and thinly coated extBC particles which cannot be absolutely separated from the bare BC aggregates using the time-delay discrimination. During the data reanalysis, we also calculated the normalized size distributions of rBC core

in thickly coated BC particles at the prescribed mobility sizes. As shown in Fig. R2, the rBC core of thickly coated BC (intBC for short) particles exhibit generally lower mass-equivalent diameters than extBC particles at each dmob. Note that the size distributions with $\text{dmob} < 200$ nm are not presented due to the lower detection limit of the SP2. It can be expected because the mobility of the whole intBC particle, which is composed of a rBC core and a considerable non-refractory materials, was selected by the DMA while only the mass of rBC core was measured by the SP2. For extBC, the mass of particle is mostly attribute to the rBC core.

Fig. R2 Normalized number size distributions of the rBC core in extBC (red) and thickly coated BC particles (magenta)

On this topic two important related points should be made. Thick coatings are more likely to be acquired by smaller particles (Fierce et al., 2016). And it is possible that core-shell coatings are more likely for larger particles (Liu et al., 2017).

Reply: We have read the two suggested references carefully. However, only the extBC aggregates without coating and a possible fraction of thinly coated BC aggregates are concerned in current study. The size-resolved mixing state of BC-containing particles will be discussed in our future study. In the revision, rough mixing states of size-resolved BC particles, calculated as the ratios of number concentration of extBC particles to the sum of extBC and intBC particles, at different mobility sizes are presented as shown in Fig. S9. Note that effects of multicharged particles are not eliminated. However, the multicharged particles should result in a similar effect on extBC and intBC at a given mobility. Thus, they should influence little of the number fraction of extBC in the total BC-containing particles. Size distribution of number fraction of extBC is used to auxiliary interpret the possible mechanism of the relatively higher masses and effective densities of extBC in the dmob range of 280–350 nm in the revision (Lines 566–591).

2. Interpretation of the 'effective density' and 'fractal dimension' The first major com-

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ment makes it clear that the 'effective density' and 'fractal dimension' results are biased towards fresh soot particles. In addition to this bias, the 'effective density' is a quantity which should correspond to the apparent density of a sphere with diameter equal to D_m . When using the DMA-SP2 setup employed in this study, the 'effective density' has virtually no meaning, since coatings are not measured by the SP2 as rBC. I do not see how this quantity could be useful for any future studies. If the authors wish to report such a quantity, they must explain in what context it should be interpreted. It should not be called 'effective density', which will confuse readers. The quantity called 'fractal dimension' has the same problem as the 'effective density.' In addition, the quantity should have been called 'mass-mobility exponent' (Sorensen, *Aerosol Sci Technol*, 45:765-779, 2011, doi:10.1080/02786826.2011.560909). It may be more interesting to compare the mixing state retrieved by asking 'is this particle similar to fresh soot?' (according to the 'effective density') with the mixing state retrieved by SP2 coating thickness analysis. But the usefulness of such an analysis is not guaranteed, the uncertainties may be too large.

Reply: Thanks for the professional comments. As presented in the original manuscript, the effective density and fractal dimension of externally mixed BC (extBC) were analyzed instead of the entire BC-containing particles. The morphology and effective density of thickly coated BC particles were not discussed in our study. They have been studied in the previous literature, e.g., Zhang et al. (2016), using the SP2 and VTDMA measurements. In our study, the extBC particles are identified according to the time-delay between the incandescence signal peak and the scattering signal peak measured by the SP2. Only the BC-containing particles with delay times less than the criterions ($2.0 \mu\text{s}$ or even lower) are considered extBC particles. The remaining BC-containing particles are considered thickly coated by nonrefractory components. As mentioned in our responses to the first major comment, a fraction of thinly coated BC particles might also be considered the extBC particles based on the time-delay approach. These thinly coated BC particles should influence the shape of the frequency or size distribution but have few impacts on the determined typical mass or mass-equivalent diameter of the

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extBC. Thus, we propose that the results presented in our current manuscript should be reliable.

Actually, the thickness of the BC-containing particle can be retrieved using a specific approach, i.e., lead-edge-only (LEO) fitting (Gao et al., 2007). The method can estimate the optical size of particles using the scattering signal detected by SP2 at the beginning stage (e.g., the first 5%) by assuming certain parameters, including the refraction indices of coating materials and the rBC core. Compared to the size of the rBC core determined using the incandescence signal, the thickness of the coating can be estimated. However, this approach needs the scattering signal measured by the split channel of the SP2 (a two-element APD). The scattering signal detected by this channel is used to determine the center location of the Gaussian-distributed laser beam. Unfortunately, this channel of our SP2 was out of work during the experiment. We failed to use this method to estimate the coating thickness of individual BC-containing particle in the present study. However, the ‘coating thickness’ is not a crucial quantity in our current study. Only the extBC particles without or possibly with thin coating are investigated. Moreover, because a number of assumptions are employed in the LEO fitting, large uncertainties also exist in the retrieved coating thickness. Similar statements have presented in our revised manuscript as shown from Line 248 to 267.

Since only the extBC particles are analyzed, the ‘effective density’ and ‘fractal dimension’ should have practical meanings. Actually, the ‘fractal dimension’ we defined on the basis of the mass-mobility relationship of extBC is the ‘mass-mobility scaling exponent’. It is different from the virtual ‘fractal dimension’ of BC aggregates (Sorensen, 2011), while also represents the morphology of BC aggregates. Although the ‘effective density’ and ‘mass-mobility scaling exponent’ of fresh soot particles were measured using a range of methods, the DMA-SP2 method was seldom used, especially in the ambient atmosphere. The advantage of the DMA-SP2 method is that the SP2 can distinguish the extBC from the thickly coated BC particles at the same time as measuring the masses of their rBC core, allowing to obtain the mass-mobility relationship in

different atmospheric environments. While previous studies using the DMA-APM-CPC method were mainly conducted in the laboratory or in the source environments (e.g., in the tunnel). Meanwhile, the DMA-APM-CPC generally measured the mobility size not exceeding 350 nm in $dmob$ due to the very number concentrations of soot particles at larger particle sizes with the $dmob > 350$ nm. The SP2 measures the particle-to-particle rBC mass, thus has a higher efficiency in the detection of larger BC-containing particles. The stepwise measurement of DMA-SP2 also provide a high time-resolution of the ‘effective density’ and ‘mass-mobility scaling exponent’ at different mobility size, which can be used to study the variation of these quantities and the possible reasons. Similar statements are presented in the last paragraph of the introduction in the revision.

In addition, we find that the effective densities of size-resolved extBC obtained from the DMA-SP2 measurement are slightly lower than those determined using the DMA-APM-CPC method. Although we have given reasonable interpretation to the difference in our manuscript, further measurements are needed to examine the difference between the two tandem methods, e.g., by measuring diesel exhaust particles synchronously. The uncertainty of the mass determination of extBC is estimated to be $\sim 20\%$ at each $dmob$, including the uncertainty ($\sim 10\%$) in the rBC mass which is converted from the incandescence peak height measured by the SP2 and the uncertainty ($\sim 10\%$) arisen from the discrimination of extBC by using different delay time criterions. We have added similar discussion on the measurement uncertainties in the revision (Lines 277–283, 371–376).

Minor comments: 1. It was not clear to me why the points in Figure S2 were quantized. Why does the number concentration nto vary smoothly?

Reply: Figure S2 (Fig. S1 in the revised supplemental file) showed an example of the number concentrations of particles (including scattering and incandescence ones) detected by SP2 during a short circle and a long one. The operated flow rate of the SP2 was set to 100 cc per minute during this experiment. The smooth number concentration

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is related to the quite low number concentration of the size-selected particles.

2. In the abstract: effective density is not morphology.

Reply: We have modified. We have checked throughout our manuscript to make the expression more rigorous. Moreover, we have written the abstract of the revised manuscript.

3. The discrepancy between DMA-APM and DMA-SP2 measurements cannot be explained by the SP2 only being sensitive to rBC. The SP2 is calibrated using an APM (or CPMA). The former DMA-APM studies used denuded soot. The discrepancy is due to the limitations of the SP2 calibration.

Reply: We calibrated our SP2 using the size-resolved Aquadag particles selected by the DMA as mentioned in the section 2.3 of our revision manuscript. The effective densities of mobility size -selected Aquadag particles were referred to Gysel. (2011). Calibrations were well conducted before and after the experiment. As shown in Fig. S3 in the revised supplemental file, the calibration curve obtained after the campaign is consistent to that obtained before the campaign (with difference in the calibration factor < 3%). Even when the denuded soot particles are measured, there also remain a possibly small fraction of non-BC components which can be measured by the APM but cannot be determined by the SP2. Rissler et al. (2014) revealed that the residual mass fraction of volatile and/or semivolatile materials in the soot aggregate was ~10% even when the sample air was heated to 300 °C before entering the system for measurement.

4. Line 55 – Thick and thinly coated needs to be defined. The authors may find that in fact most atmospheric BC is coated.

Reply: We used the time-delay method to distinguish the mixing state of refractory BC (rBC). Two type rBC-containing particles can be distinguished. One is the rBC thickly coated by nonrefractory components and the other is rBC thinly coated or without coat-

ing (bared). The thickness of the coatings cannot be accurately identified in current study. As mentioned in the introduction of the manuscript, there also be a considerable fraction of extBC in the atmosphere especially in the urban regions (Lines 77–82 in the revision). The extBC discriminated using the time-delay method accounted for even >50% of the total BC-containing particles which were detectable by the SP2 in urban Beijing, although a fraction of extBC might be thinly or even moderately coated. The TEM analysis also showed that the bare-like soot particles accounted for 25% of the total soot particles in urban, and as high as 64% in tunnel. 63% soot particles were partly coated in urban environment (Wang et al., 2017). Thus, the morphology and effective density of these extBC aggregates in the atmosphere are studied.

Actually, the thickness of the coat can be retrieved from the scattering signals of the SP2 using the lead-edge-only (LEO) fitting (Gao et al., 2007). However, the notch in the two-element APD of our SP2 failed to fix in an adequate position in this experiment. Thus, the optical size and the consequent coating thickness of the BC-containing particles cannot be estimated in current study. However, the coating thickness is not a crucial quantity in our current study on the morphology and density of uncoated BC aggregates. It can provide a validation of our discrimination of extBC but should have little influence on our final analysis and discussion presented in the current study. Similar statements are presented in Lines 248–267 in our revised manuscript.

5. Line 88 – explain the reasons for the detection limits.

Reply: As mentioned in Lines 111–123 in the revision, larger uncertainties exist in the DMA-APM-CPC measurement for larger particles (e.g., with a $dmob > 350$ nm) which are much less abundant in the atmosphere than the smaller particles. Thus, the previous tandem measurements generally provided the mass-mobility relationship of BC aggregates with a $dmob$ not exceeding 350 nm.

6. Line 110 – quasi-monodisperse needs to be quantified or specified precisely. AMT is a technical journal.

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Reply: Due to the effect of DMA transfer function, particles within a certain dmob range are selected by the DMA at a given voltage. Usually, the particle number presents a triangle distribution as the function of dmob at a given voltage. The peak of the triangle distribution corresponds to the certain dmob which we are used in this study. A slight modification to the statement is performed in the revision (Line 184). In the current manuscript, we won't specify the principle of DMA that used to quantify the quasi-monodisperse since it is out of our current research scope.

7. Line 149 specify the intensity of the laser in units of W/m² or similar

Reply: Usually, the intensity of the laser is not specified in the SP2 study. Although the intensity of laser would influence the detection efficiency of SP2, it is usually checked indirectly by using the monodisperse Polystyrene Latex (PSL) at a given size (e.g., 269 nm). The intensity of the laser depends on the laser current which can be set manually in the SP2 acquisition software.

After the experiment, we used the DMA-SP/CPC system to measure the bare BC (Aquadag) particles. The detection efficiency was obtained by comparing the particle number concentration detected by the SP2 to that measured by the CPC at each prescribed mobility, as shown in Fig. S4 in the revised supplement file. Adequate detection efficiencies (>~90%) of SP2 are found when the mass-equivalent diameter of rBC core is larger than ~90 nm.

8. Line 153 frequency is the wrong word.

Reply: We have changed the word 'frequency' to 'frequency distribution' in our revised manuscript (Line 234).

9. Line 185 define K.

Reply: Have defined in the revision (Line 405).

10. Line 255 discuss the Diffusion-Limited Cluster-Cluster Aggregation mechanism (see Sorensen citation above)

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Reply: We have read the review literature (Sorensen, 2011) carefully and found that the mass-mobility scaling exponents measured using the tandem techniques (e.g., DMA-APM, DMA-SP2) were sometime erroneously equivalent to the virtual fractal dimension of BC aggregates. The same mistake occurs in our current study. Corrections to this mistake have been performed in the revision (e.g., Lines 87–107, 393–396).

11. Line 261 organics do not 'fill in the gap', they cause restructuring

Reply: Corrected in the revision (Lines 432–434)

12. Line 357 the drag force, or uncertainty? Figure 6 needs error bars. Counting statistics will be poorer at the extreme sizes, possibly causing the observed trend.

Reply: We have rewritten Section 4.3 as shown in Lines 601–613 in the revision. A more reasonable interception is presented for the hiatus in the increase trend of dynamic shape factor.

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Please also note the supplement to this comment:

<https://www.atmos-meas-tech-discuss.net/amt-2018-408/amt-2018-408-AC1-supplement.pdf>

Interactive comment on Atmos. Meas. Tech. Discuss., doi:10.5194/amt-2018-408, 2018.

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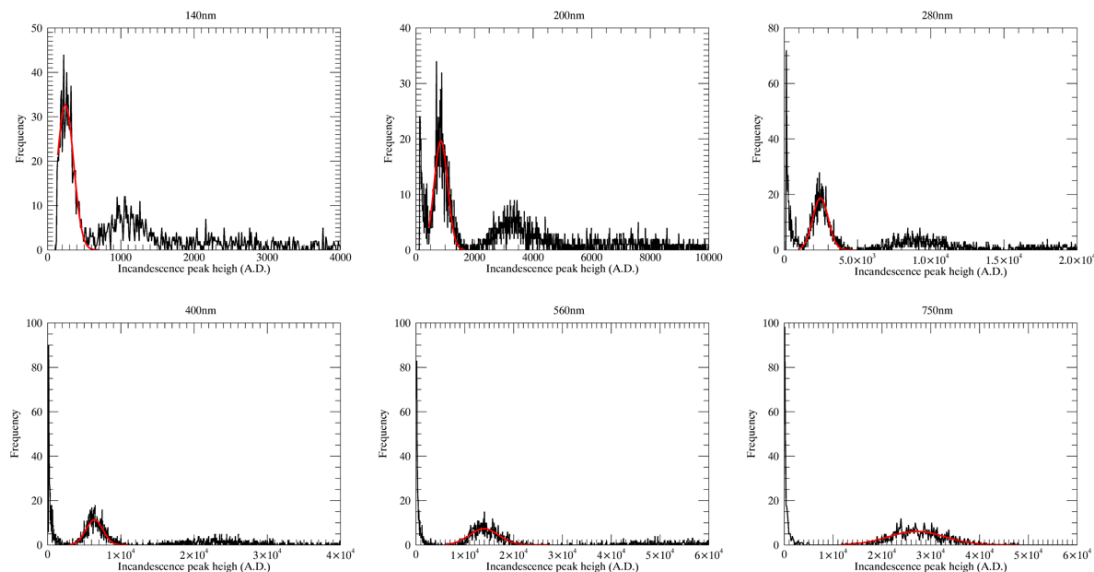


Fig. 1. Frequency distributions of incandescence peak height detected by the SP2 for size-selected Aquadag particles at the prescribed mobility sizes

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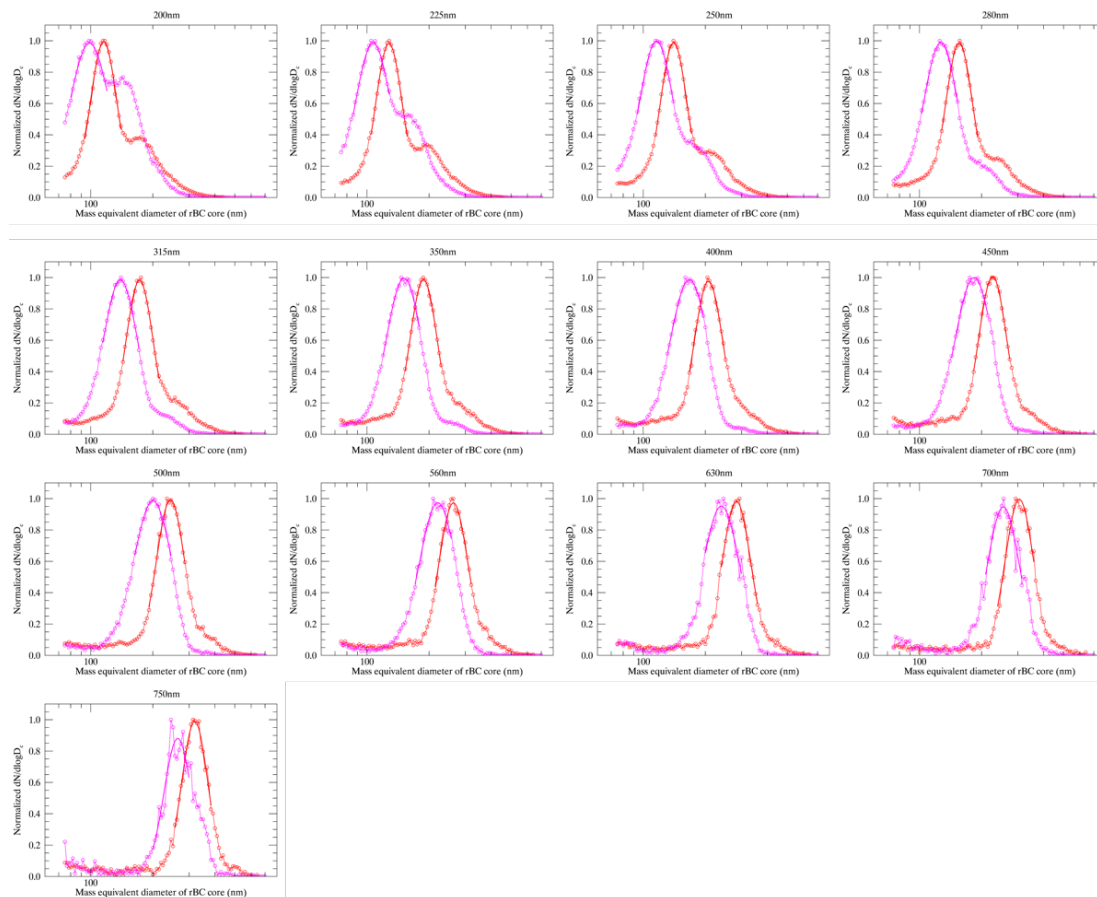


Fig. 2. Normalized number size distributions of the rBC core in extBC (red) and thickly coated BC particles (magenta)

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