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Atmospheric Measurement Techniques Discussions

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.

Anonymous Referee #2

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* General comments

The authors use a tandem DMA-SP2 system to measure the mass-mobility exponent, effective density, and shape factor of ambient BC particles in Beijing, China. Despite the claim to novelty made in the title similar combined DMA-SP2 measurements have already been discussed and reported in the literature, which the authors have failed to discuss. In addition, there are significant shortcomings in the manuscript itself, ranging from inadequate description of the experimental details and results to data quality issues. In my view the manuscript is at the 'early draft' stage rather than the 'under

review' stage. I believe major revisions are required before the manuscript can be considered for publication in AMT. Most importantly, the authors need to better demonstrate why this study is an original contribution to the literature on the properties of ambient BC aerosols, and why their measurements should be considered artifact-free and trustworthy.

I see the following major issues:

The study is not placed in appropriate context through citation of relevant literature. In the title and elsewhere (e.g. L101) the authors claim that this is a 'novel' system for measuring the morphology and effective density of black carbon particles. This is incorrect. Tandem DMA-SP2 measurements have been discussed and/or performed in a number of different studies (e.g. Gysel et al., 2011; Raatikainen et al., 2017; Zhang et al., 2016). A handful of references are cited for more general mass-mobility measurements (e.g. using an APM rather than an SP2). But the literature on this topic is more extensive than this small selection of studies would suggest, including a review on the mobility of fracal aggregates by Sorensen (2011). I suggest the authors read this review and the references therein and put more effort into placing their measurements into the context of these previous works. In particular, the authors need to demonstrate what is the original contribution of this work.

The quality of the writing is not at a suitable level for scientific publication. There are many English grammar issues - too many to list in a scientific review. Beyond this, the language is frequently too vague. To take just the first example I come across the authors state that BC can lead to 'Earth warming' on L39. I believe that authors mean 'warming of the Earth's atmosphere' or similar. There are many more examples of such lazy language throughout the manuscript.

The study design and experimental and analytical details are inadequately described, which makes it difficult to judge their suitability. For example from what I can gather, the results presented in Figs. 2 to 6 are only for rBC-containing particles that displayed

a delay time of less than 2us (defined as 'extBC'). This needs to be clarified and stated more explicitly (another example of lazy language). The suitability of using delay time to distinguish between externally and internally mixed BC particles then requires further discussion. It is not as simplistic as the authors make it out to be. The authors allude to the fact that thinly-coated BC will exhibit low SP2 delay times on L280. This is also true for moderately-coated BC (e.g. with BC volume fractions as low as 30%), which would certainly not be classified as 'externally mixed' (ExtBC). A better approach for classifying BC mixing state with the SP2 is the quantitative LEO-fit approach (Gao et al., 2007; Laborde et al., 2012). The authors need to at least discuss this more sophisticated method, the reasons why they chose to use the more simplistic delay time approach, and the consequences of this decision.

The material as presented gives cause to question the quality of the measurement data, but insufficient details are provided to fully make this assessment. Specific points are highlighted below in relation to Fig. 2, which contains features that require explanation (absence of clearly defined peaks for multiply charged particles, presence of peaks with rBC mass approaching 0). In addition for reasons that are not yet satisfactorily explained, the effective density results presented in Fig. 5 are systematically lower than previous measurements (both the previous studies already cited by the authors as well as previous DMA-SP2 measurements that were also conducted in Beijing but are not yet discussed; Zhang et al., 2016). Given these issues, the authors should include further data and explanations to build confidence in their results and to confirm that the measurements are artifact-free. For example, were PSL spheres or other monodisperse particles (e.g. aquadag for the SP2 measurements) used to confirm that the DMA was operating correctly? What quality checks were performed to ensure the SP2 was operating correctly? (E.g. laser and detector block properly aligned, laser power levels, flow rate checks, comparison of calibration curves with previous calibration curves of the instrument).

No uncertainty estimates are provided for the measurements and main results. This is

important as the SP2 counting statistics and therefore measurement errors will be size-dependent. How sensitive are the reported quantities (e.g. Df) to these measurement uncertainties?

* Specific comments

L69: 'Representativeness' instead of 'representation'

L80 - 82: The meaning of these sentences is not clear, rewording required.

L92: Schwarz misspelt.

L98: Statement needs qualification. This is not true when measuring pure BC particles.

L109: Please include information about the neutralizer that was used upstream of the DMA. This is especially required when considering the potential impacts of multiply charged particles as discussed in Section 3.2.

L135: Please include the length of the nafion dryer and the source of the dry sheath air.

L139: Some discussion is required about SP2 counting efficiency over this size range. It is not always 1, which will affect counting statistics, adding uncertainty to measurements reported at the limits of the range.

L153: The phrase 'frequency of the time lag' does not make sense. I guess the authors meant frequency distribution or histogram.

L156: Am I correct in assuming that the 'extBC' results presented later only include particles that displayed a lag time less than 2 us? If so please state this explicitly.

L162: More sensitive than what? To Fullerene soot I presume but statements like this need to be explicit, avoiding lazy language.

L166: Please include a Figure in the supplementary information of the two measured incandescence calibration curves. Were the incandescence calibrations performed all

the way up to 750 nm or were the calibration curves extrapolated? What sort of function was fit to the calibrations curves? Were the calibrations consistent with previous calibration curves measured for this instrument (an important check to make to ensure the SP2 was operating ok)

L176: Please also discuss what the prefactor k represents (e.g. Sorensen 2011).

L191: Please provide a reference for why this value of material density was chosen.

L199: The more common phrasing for this section would be 'Data processing'. And it seems to me that Section 2.4 would be a better fit in this section rather than the measurement methodology section above.

L220: It seems that this method was not used in this study. Why is it mentioned? Was it used to check the results processed with the first method?

L232: 'Minimizing the multicharged particles' is too vague. Suggest 'Correcting for the presence of multiply-charged particles' or something similar.

L240: Please be more specific. Exactly what parameter of the fitted curve was used to represent the mass of singly charged extBC particles?

Fig. 2: These curves contain features that require discussion to build confidence in the measurements. For example: related to the comment above about L156, do these frequency distributions only contain particles that display time lag less than 2us? If so, why are there sharp increases in the number of particles with rBC mass approaching 0 at diameters greater than 160nm? (With no filtering of the data applied I would assume these are heavily coated particles, which is a reason why I think the filtering process is not strict enough to be able to label these particles as 'extBC'). Why do the multiply-charged particles show up as a very fat tail that does not descend to a frequency of 0 until some point beyond the upper limit of the x-axis? What do these tails in the distribution represent? (E.g. they are in contrast to what is typically seen in SP2 calibrations, when doubly and triply charged particles are observed as clear, separate

gaussian peaks). I suggest adding vertical lines along the x-axis corresponding to the mass of particles selected by DMA (e.g. under the assumption of spherical particles with the material density of BC) to provide reference points to compare the measured mass distributions to.

L252: Please provide further details for how the fit was performed. Were both the prefactor k and mass-mobility exponent Df allowed to vary freely? The k value seems to be very low in comparison to previous measurements (Sorensen 2011 and references therein), which requires explanation. How is the standard deviation of 0.04 in Df determined? How sensitive is the fitted Df to the size-dependent measurement uncertainties in rBC mass?

Fig. 3: Error bars are required in this figure to indicate the measurement uncertainties.

L263: This is a speculative statement that is not examined as thoroughly as it should be. If the sensitivity of Df to measurement uncertainty is taken into consideration is it still possible to conclude that the Df calculated here is 'relatively lower' than the previous measurements? If this result stands, can the authors provide any evidence to support their claim that fuel quality is higher in Beijing than for the studies they have cited?

L337: 'extBC' as defined with respect to delay time could also be comprised of BC mixed with small or even moderate amounts of non-BC material (e.g. L280). Additionally, the measurements reported here are lower than previous DMA-SP2 measurements of effective density in Beijing. Therefore, I find this explanation of the low effective density values measured in this study to be problematic.

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