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Atmospheric Measurement Techniques Discussions
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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 #1

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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 by the authors). The remainder of the measurements (the shape of the overall distri-

bution) 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.

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 (Dm) 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.

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.

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.

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.

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).

Fierce, L. et al. Black carbon absorption at the global scale is affected by particle-scale diversity in composition. Nat. Commun. 7:12361 doi: 10.1038/ncomms12361 (2016).

Liu, D. et al. Black-carbon absorption enhancement in the atmosphere determined by particle mixing state. Nat. Geosci., vol 10, pp 184–188 (2017). doi:10.1038/ngeo2901

2. Interpretation of the 'effective density' and 'fractal dimension'

The first major comment 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 Dm. 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.

Minor comments

Several 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?
2. In the abstract: effective density is not morpholgoy.
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.
4. Line 55 – Thick and thinly coated needs to be defined. The authors may find that in fact most atmospheric BC is coated.
5. Line 88 – explain the reasons for the detection limits.
6. Line 110 – quasi-monodisperse needs to be quantified or specified precisely. AMT is a technical journal.
7. Line 149 specify the intensity of the laser in units of W/m2 or similar.
8. Line 153 frequency is the wrong word.
9. Line 185 define K.
10. Line 255 discuss the Diffusion-Limited Cluster-Cluster Aggregation mechanism (see Sorensen citation above)
11. Line 261 organics do not 'fill in the gap', they cause restructuring.
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.

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