The paper presents a new method to quantify elemental carbon mass size distribution using size-resolved absorption coefficient measured by an aerodynamic aerosol classifier in tandem with an aethalometer. It aims to demonstrate the feasibility of the method by comparing the measurement results with that of refractory black carbon mass size distribution measured by a differential mobility analyzer in tandem with a single particle soot photometer (DMA - SP2) during a field measurement in Yangtze River Delta. The impact of different assumptions in calculation processes on EC mass size distribution is also presented. The method presented in the paper is of general interest to the scientific community with application in the climate and air quality sectors. A complete understanding of the black carbon cycle and radiative impacts requires knowledge of processes related to the temporal and spatial evolution of BC size distribution. Although a variety of techniques are available to measure BC size distribution, several limitations of these techniques relates to time resolution, instrument cost and complex data analysis.

<u>Response:</u> Thanks for your comments.

Major comments:

1) The authors should follow the recommendation by Petzold et al. (2013) to properly clarify the terms used for BC. Due to their design the various BC measuring techniques used in the paper are based on different physical and chemical BC properties. This generates difficulties when comparing data from the different techniques because it may provide different BC concentrations. Since there is no recognized technique to quantify BC in the atmosphere, Petzold et al. (2013) established terminology recommendations for reporting BC results. The terms "refractory BC" (rBC) and "equivalent BC" (eBC) should be used to refer to the BC mass concentration quantified using SP2 and aethalometer, respectively. Internally mixed BC particles composed of both BC and non-BC material should be designated as "BC-containing particles". <u>Response:</u> Thanks for your recommendation. We agreed with you and the revisions were made according to your suggestion. The revisions improved the description of our study. BC measured by SP2 and aethalometer was termed as equivalent BC (eBC) and refractory BC (rBC), respectively. Internally mixed BC particles composed of both BC and non-BC material were referred to as BC-containing particles.

2) The significance of the study should be strengthened. The main motivation of the proposed new method is to extend the detection range of BC size distribution up to 1.5 μ m, while the widely used SP2 instrument can't measure rBC above 600 nm. The mass median diameter of fresh BC is normally distributed within the range of 30 – 200 nm. If they exist in the atmosphere, I guess superlarge-sized BC

may be formed by massive coating or superaggregation. What is the state of the art on these coarse BC particles? Have they already been observed in the atmosphere using other measurement techniques? Are they formed in the atmosphere by massive coating or superaggregation? What is their contribution to the total BC concentration, BC radiative forcing and CCN ability?

<u>Response:</u> Thanks for your insight comments. Yes, we agreed with you that coarse BC was formed by massive coating or superaggregation. Wang et al. (2022) found that coarse BC was formed by massive coating or superaggregation with transmission electron microscopy. Contribution of coarse BC to total BC concentration could be as large as 50 % (Wang et al., 2022). The contribution of coarse BC to radiative forcing and CCN ability was unclear so far.

3) There is lack of information on the measurement setup. There is no information on the aerosol sampling (inlet used for aerosol sampling, cut-off diameter of the sampling inlet, aerosol conditioner to dry,...), which need careful consideration for interpreting the results. In addition the authors did not provide any information regarding the calibration of the instruments. Have each instrument been calibrated individually using spherical monodisperse particles? Which detector was calibrated and used to analyze SP2 measurements? The accuracy of the rBC mass concentration measured by SP2 is strongly dependent on the calibration material chosen. This should be mentioned in the paper. Without these information, the reliability of the data is questionable.

<u>Response:</u> Thanks for your comments. We input more information on the aerosol sampling system in Sect. 2.1 and Fig. 1 was replotted accordingly.

AAC, AE33, APS and SMPS were calibrated by corresponding manufacturers before the field measurement. SP2 was calibrated by monodispersed Aquadag soot particles. Incandescence high gain channel and incandescence low gain channel were calibrated and used to analyze SP2 measurement. The above information was added to Sect. 2.1.3 of the revised manuscript.

4) There are some methodology and assumptions in calculation processes of rBC mass size distribution measured by the DMA-SP2 that are not discussed. Just as in a typical SMPS data analysis, the rBC mass size distribution need to be calculated from the raw observations of rBC number (or mass) versus voltage using an inversion code. This inversion depends on particle charging efficiencies and the DMA transfer function. The authors should clearly explain the method they used to correct the size distribution measured by the DMA-SP2 to output the information for singly charged particles only. In addition the particle size selected by the DMA is an electrical mobility diameter, which is highly sensitive to the particle shape. If the authors neglected the dependence of rBC asphericity, they shoud mention it in the paper. The authors could

also analyze in section 4 the possible uncertainty in deriving rBC diameters from DMA-SP2 owing to the dynamic shape factor they used.

<u>Response:</u> Thanks for your comments. We agreed with the comments. We explained the method to correct the size distribution measured by DMA – SP2 to output the information for singly charged particles in Sect. 2.1.3 of the revsied manuscript. We also metioned that rBC asphericity was neglected in Sect. 2.1.3.

Specific comments:

1) L80-85: Why did not you use the SP2 alone to obtain rBC mass size distribution ? Please add the size range of the DMA-SP2.

<u>Response:</u> Thanks for your comments. The reason why DMA – SP2 was used rather than SP2 alone was that D_p could be directly measured by DMA. If SP2 was used alone, D_p was calculated from Mie theory with assumed inputs (Taylor et al., 2015). We added the size range of DMA – SP2 in Sect. 2.1.3.

2) L98: Where was C measured in Zhao et al. (2020b)?

<u>Response:</u> Thanks for your comment. C in our manuscript was "multiple-scattering correction factor (C_f)" defined in the section "INTRODUCTION" of Zhao et al. (2020b), specifically in Page 1834. The discussion of C_f could be found in subsection "Multiple-scattering Correction Factor" of section "RESULTS AND DISCUSSION", specifically from Page 1837 to Page 1838. C was changed into C_f in our revised manuscript to consistent with Zhao et al. (2020b).

3) L205: Fig. S3b instead of S2b.

Response: Thanks for your comment. Fig. S2b was changed to Fig. S3b in the revised manuscript.

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