Response to Reviewer #1:

Reviewer 2, Second Review of Jing Wei et al., "Unraveling the impact of heterogeneity and morphology on light absorption enhancement of black-carbon-containing particles"

I comment the authors on what is clearly a lot of hard work in revising their manuscripts and writing good responses.

However, I have some remaining concerns. The general problem is that the authors have often responded to my comments by writing a rebuttal, rather than by improving the manuscript. In other words, if the authors have scientific arguments against my comments, these arguments belong in the manuscript, not only in the review response. For two specific comments I will ask the authors to add discussion of the issues, see below.

[Comment 1B] I quote from my original [Comment 1]: "The major gap is that this work does not cite, nor implement, the advances in[charger-]CPMA-SP2 data inversion which have been published since the original work by Liu etal., 2017 (cited by the authors). Those advances have been published in a series of papers, mostrecently Naseri et al. 2024, which are accompanied by open-source code for performing thecalculations."

The authors responded "we did not directly use the inversion code from Liu et al. (2017a) or Naseri et al. (2024)". In fact, Liu et al. 2017a did not publish any inversion at all, and did not do any data inversion. Nor have the authors done any inversion here. Inversion is a standard procedure in SMPS measurements, where signals from q=2 particles at q=1 are subtracted (i.e., corrected for). Is it appripriate to publish SMPS data without inversion? No. Nor is it appropriate to publish CPMA-SP2 data without inversion. Inversion is necessary to calculate the quantities like E_abs presented in this work.

If the authors wish to reject the concept of data inversion in favour of a different analysis scheme, they must at a bare minimum explicitly discuss previous inversion work and present clear, quantitative arguments against it. The authors cite the previous work for "applying the tandem CPMA-SP2" but not for data inversion.

Ideally, the authors would BOTH use a classical inversion scheme AND their bimodal-peak-fitting approach. While I hope to see this ideal in future work, it is not necessary for publication of the present work.

Response: We thank the reviewer for the continued evaluation of the revised manuscript and apologize for the lack of clarity in our previous response. We would like to clarify that the CPMA transfer function was already calculated and applied in the original data analysis, and all mass-resolved quantities presented in this work were derived from transfer-function-corrected CPMA–SP2 measurements. However, the explicit description of the transfer function calculation and its role in the data processing workflow was not sufficiently detailed in the earlier version of the manuscript. In response to this concern, we have substantially revised the Methods section to explicitly describe the calculation and application of the CPMA transfer function, as well as the subsequent treatment of multiply charged particles (Lines 133-148 and Text S1). The CPMA transfer function was calculated based on the CPMA geometric and operational parameters, using a set mass resolution R_m=8, and applied to correct for the finite mass resolution and transmission characteristics of the CPMA prior to further data processing. Following the transfer-function correction, contributions from multiply charged particles (q>1), arising from the known bipolar charge distribution produced by the upstream X-ray aerosol neutralizer, were identified as distinct

mass modes and quantitatively removed using a peak-resolved subtraction approach. Under the operating conditions of this study, the CPMA mass resolution was sufficient to clearly resolve charge-dependent mass modes, allowing the singly charged mode corresponding to the CPMA setpoint to be retained for subsequent analysis. This sequential treatment—explicit correction for the CPMA transfer function followed by charge-resolved subtraction—constitutes a simplified but physically consistent alternative to full matrix-based inversion schemes (Naseri et al., 2024; Naseri et al., 2021). While matrix-based inversion provides a unified framework for simultaneously treating transfer function convolution and charge distribution, the approach adopted here is robust when charge-dependent mass modes are well separated and non-overlapping. Additional corrections, including SP2 detection efficiency and instrumental time delay, were also applied as described in Text S1. We have now explicitly cited recent CPMA-SP2 inversion studies and clarified in the manuscript how our applied correction scheme relates to, and differs from, full matrix-based inversion approaches.

"*In the subsequent data processing, measurements from the CPMA-SP2 system were first corrected for multiple charging effects using a peak-resolved subtraction approach. An X-ray aerosol neutralizer (TSI 3088) installed upstream of the CPMA produced a known bipolar charge distribution, resulting in discrete mass modes corresponding to singly and multiply charged particles in the CPMA mass spectra. The CPMA transfer function was calculated based on the CPMA geometric and operational parameters, with a set mass resolution* R_m*=8 (Text S1), ensuring that charge-dependent mass modes were sufficiently resolved. Under these conditions, mass peaks attributable to multiply charged particles (q > 1) could be explicitly identified based on their expected mass-to-charge relationships and quantitatively subtracted, while the singly charged mode corresponding to the CPMA setpoint was retained for subsequent analysis. This peak-resolved subtraction represents a simplified, charge-resolved inversion, which differs from full matrix-based inversion schemes (Naseri et al., 2024; Naseri et al., 2021) , but is appropriate when charge-dependent mass modes are well resolved and non-overlapping. Additional corrections, including SP2 detection efficiency, and instrumental time delay, were applied as described in Text S1*."

We hope that these revisions clarify the CPMA-SP2 data processing procedure and adequately address the reviewer's concerns regarding the treatment of instrument response and data inversion.

[Comment 2B] My [Comment 2]: The MAC vs M_R plot of Figure S6 shows that MAC did not increase even at M_R of 5.5. Mie theory and lab experiments show that coated soot has E_abs 2.0 at M_R of 5.5 (Cappa et al., 2012; doi:10.1126/science.1223447). So the fundamental premise of this paper seems to be violated by Figure S6. The author's response was "In the atmosphere, factors such as particle mixing state, coating composition, hygroscopic growth, and measurement uncertainties can influence the observed MAC values."

Response: We thank the reviewer for this comment and apologize for our previous explanation. In the revised manuscript, we have added a detailed explanation in the Methods section addressing why the measured MAC does not increase at high bulk-averaged M_R, in comparison with laboratory and Mie-theory predictions (Cappa et al., 2012). Specifically, we discuss the relative contributions of particle mixing state, morphology, coating composition, and instrumental uncertainties, and provide quantitative estimates of their impact on absorption enhancement,

consistent with previous ambient studies (Cappa et al., 2019; Huang et al., 2024). These additions clarify the underlying causes of the observed limited enhancement (E_{abs} ≈ 1.1-1.5) and highlight the value of our field measurements for constraining BC absorption under realistic atmospheric conditions.

The detailed revised text and supporting quantitative discussion are provided below (Lines 239-250):

"*Importantly, at high bulk-averaged M_R (≈5), the measured MAC did not approach the laboratory-based absorption enhancement (E_{abs} ≈ 2) reported for idealized core-shell soot particles (Cappa et al., 2012). The observed limited E_{abs} ≈1.1~1.5 is consistent with ambient studies and can be explained by the complexity of ambient particles: the heterogeneity of particle-resolved M_R and morphology can reduce absorption enhancement by ~20-70% relative to idealized internal mixtures (Cappa et al., 2019; Huang et al., 2024), while variations in coating composition and instrumental uncertainties contribute additional variability but are considered secondary factors. These high-time-resolution field measurements therefore provide quantitative constraints on BC E_{abs} under realistic atmospheric conditions, complementing laboratory studies and informing model evaluations of aerosol optical properties and regional radiative effects.*"

References:

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