Response to Reviewer #3:

The manuscript treats an important aspect of black carbon optical properties with innovative techniques. Overall, the topic and novelty fulfil the requirements for ACP publication. Some work still to be done improve the readability of the manuscript and especially ensure that robustness of the measured properties. The article requires consistent modification.

[Comment 1] NOMENCLATURE AND READABILITY: The use of abbreviations and symbols requires careful attention. Several parameters (particularly those distinguishing measured from modeled quantities) are difficult to follow, which hinders comprehension, especially in the results section. I strongly recommend introducing a summary table listing each property, its abbreviation, and whether it refers to a measurement or a modeled value. Improving consistency here would greatly enhance clarity for readers unfamiliar with the measurement framework.

Response: We appreciate the reviewer's suggestion regarding the clarity of abbreviations and symbols. Following this recommendation, we have now compiled a comprehensive summary table that lists all parameters, their abbreviations, and whether they refer to measured or modeled quantities. This table has been added as Table S1 in the revised Supplementary Information. We believe this addition substantially improves the readability and consistency of the manuscript, particularly for readers who are less familiar with the measurement framework.

Table S1. *The abbreviation of this study*

Abbreviation	*Full Name*
BC	*Black carbon*
E_{abs}	*light absorption enhancement*
M_R	*Coating-to-core mass ratio of single BC-containing particle*
DRF	*direct radiative forcing*
bulk-averaged M_R	*Averaged* M_R *of bulk BC-containing particle*
C_{sca}	*the scattering cross-section*
$C_{sca\_measured}$	*the measured scattering cross-section*
$C_{sca\_modeled}$	*the modeled scattering cross-section by core-shell Mie model*
M_p	*the mass of a BC-containing particle*
M_{BC_core}	*the mass of BC core*
MAC	*the mass absorption cross section*
$E_{abs\_measured}$	*the measured light absorption enhancement*
$MAC_{BC\_core\_measured}$	*the MAC of uncoated BC particles extrapolated from measured MAC values of BC-containing particles*
D_c	*the BC core size*
D_p/D_c	*BC coating thickness*
RI	*the refractive index*
$MAC_{BC\_coated\_modeled}$	*modeled MAC of coated BC*
WS	*wind speed*
RH	*relative humidity*
$E_{abs\_uniform}$	*the* E_{abs} *calculated using the traditional core-shell Mie model*
ΔE_{abs}	$E_{abs\_uniform}$-$E_{abs\_measured}$
$E_{abs\_resolved}$	*the* E_{abs} *calculated from each BC-containing particle using the core-shell Mie*

	model
E_{abs_param}	*the E_{abs} obtained based on the empirical scheme developed in this study*
CPMA	*Centrifugal Particle Mass Analyzer*
SP2	*single-particle soot photometer*
PSL	*polystyrene latex spheres*
TOF-ACSM X	*time-of-flight aerosol chemical speciation monitor*
CAPS-ALB	*Multi-Wavelength Cavity Attenuated Phase Shift Single-Scattering Albedo Monitor*
DMA	*Differential Mobility Analyzer*
LEO	*the leading-edge-only technique*

[Comment 2] INSTRUMENTAL DESCRIPTION AND UNCERTAINTIES: The methodology section currently lacks sufficient citations and discussion of measurement uncertainties. Given that the study combines multiple instruments in a novel configuration, these details are crucial. The uncertainties of the CPMA–SP2 tandem system, as well as of derived quantities such as M_p and M_BC, should be clearly stated and discussed in the context of previous literature. Since many key quantities in the analysis are expressed as ratios, unquantified uncertainties may propagate and influence the reported variability. In line with other reviewers' remarks, I encourage the authors to describe the CPMA–SP2 system and data processing steps in greater detail, including how calibration and error propagation were handled.

Response: We appreciate the reviewer's insightful comment. In the revised manuscript, we have substantially expanded the methodological description of the CPMA-SP2 system and clarified all associated measurement uncertainties. Specifically, we now report the uncertainties of M_p (5%), M_{BC_core} (10%), single-particle M_R (~11%), and bulk-averaged M_R (~7%), and the absorption coefficient (15-20%), as well as the propagated uncertainties for $MAC_{BC\_coated\_measured}$ (18-22%), $MAC_{BC\_core\_measured}$ (19-23%), and $E_{abs\_measured}$ (26-32%). The corresponding uncertainty-estimation procedures and calibration details have been added to Text S2, and the resulting values are clearly stated at the relevant locations in the revised manuscript (Lines 98-225). Some key description can be found as follows:

Lines 111-112 (M_p): *"According to the instrument manual, the mass accuracy of the CPMA is approximately 5%."*

Lines 114-116 (M_{BC_core}): "……., *and the uncertainty associated with the SP2-derived BC core mass (M_{BC_core}) is approximately 10% (Laborde et al., 2012).*"

Lines 152-155 (the absorption coefficient): "*Absorption was calculated as the difference between extinction and scattering, with estimated uncertainties of ~1-10% for both extinction and scattering (Modini et al., 2021), leading to a conservative absorption uncertainty of ~15-20% for the submicron BC particles considered.*"

Lines 180-181 (single-particle M_R): "*Considering the uncertainties of M_p (5%) and M_{BC_core} (10%), the uncertainty of M_R for a single BC-containing particle was approximately 11% (Text S2).*"

Lines 185-187 (bulk-averaged M_R): "*Propagating these uncertainties to the hourly mass-weighted calculation resulted in an uncertainty of approximately 7% for the bulk-averaged M_R (Text S2).*"

Lines 221-224 ($MAC_{BC\_core\_measured}$ and $E_{abs\_measured}$): "*Based on our error propagation analysis,*

which accounts for measurement uncertainties in particle absorption and BC mass as well as the standard error of the extrapolation, the estimated uncertainty of $MAC_{BC\_core\_measured}$ *is approximately 19-23% (Text S2). And the uncertainty of* $E_{abs\_measured}$ *is approximately 26 - 32% (Text S2).*"

I hope this revision improves the clarity for readers.

[Comment 3] RESULTS AND INTERPRETATION: The results section shows promising potential for impact, but several points require clarification. The authors could strengthen the manuscript by expanding the discussion on how the proposed "transition-state" correction scheme might be generalized to other atmospheric environments. For example, conditions in rural or biomass-burning regions, or during other seasons, may produce distinct coating compositions and morphology evolution pathways. Explaining how the correction parameters (e.g., MR thresholds or morphology indicators) could adapt to such conditions would enhance the broader applicability of the method. The method for defining the three "cases" should also be revisited. It appears that only Case 2 represents a specific or anomalous event compared to the rest of the campaign. I suggest first describing the overall meteorological and bulk aerosol conditions and then examining how the optical properties vary under those regimes. This would provide a more physically grounded interpretation of the case classification. At present, the combined issues of unclear nomenclature and insufficient methodological detail reduce the understanding of the results, being the ultimate limiting factor of the manuscript.

Response: We thank the reviewer for these constructive comments. Following this suggestion-and in conjunction with the feedback from the other reviewers—we have substantially revised the Results section. Specifically, we have: (1) expanded the discussion on the potential generalization of the transition-state correction scheme to other atmospheric environments (e.g., rural, biomass-burning, and different seasonal conditions); (2) clarified how key parameters such as M_R thresholds of transition state may adapt under varying coating conditions; and (3) reorganized the case definitions by first presenting the overall meteorological and bulk aerosol characteristics before examining the case-dependent optical behavior. We believe these revisions effectively address the reviewer's concerns and greatly improve the readability and interpretability of the Results section. Please refer to the revised manuscript (Section 3.1-3.3).

SPECIFIC COMMENTS

[Comment 4] L1: title. I am not convinced by "heterogeneity". What does it mean in this context? It is a very general term that, alone, does not convey a unanimous message.

Response: We thank the reviewer for this comment. In our study, "heterogeneity" specifically refers to the variability in the mass ratio of coatings to BC cores among particles. To clarify this and in line with the suggestions of Reviewer 2 (Comment 3), we have revised the title to:

"*Effects of Mass Ratio Heterogeneity and Coating-Related Optical Characteristics on the Light Absorption Enhancement of Black Carbon-Containing Particles.*"

[Comment 5] L35-38: Eabs is not contextualized, not properly described. Enhancement with respect to? Please provide a short description.

Response: We thank the reviewer for pointing this out. In the revised manuscript, we have clarified the definition of BC E_{abs} in the context of particle coatings. Specifically, E_{abs} is defined as the ratio

of absorption by coated BC particles to that of uncoated BC cores, capturing the increase in absorption due to the presence of coatings. This definition is now included in the revised manuscript (Lines 35-41):

"*According to IPCC assessments, the global effective DRF of BC ranges from −0.28 to 0.41 W/m^2 (Szopa et al., 2021). To represent the effect of BC particle coatings on absorption, most climate models (Bauer et al., 2013; Chen et al., 2024; Stier et al., 2005; Wang et al., 2023; Zhang et al., 2025b) estimate BC light absorption enhancement (E_{abs}) using Mie theory, defined as the ratio of absorption by coated BC to that of uncoated BC cores, under the assumption of a uniform core-shell structure where BC core is fully coated by coating materials.*"

[Comment 6] L51: what do you mean with heterogeneity?

Response: We thank the reviewer for the comment. In this study, "heterogeneity" specifically refers to the variability or dispersion in the mass ratio (M_R) of the coating to BC cores among individual particles. This definition clarifies that we focus on the distribution of core-shell mass ratios. Notably, several previous studies have also used the term "heterogeneity" to characterize similar variability in BC particle properties (Fierce et al., 2020; Fierce et al., 2016; Zeng et al., 2024), supporting our usage of the term in this context.

[Comment 7] L67: please avoid the use of "fortunately" , it undermines the preparation and thoughts behind your research. Leaving the reason of the positive outcome of your work to luck.

Response: We thank the reviewer for the comment. The word "fortunately" has been removed from the manuscript, as suggested. The revised sentence can be found at lines 88-89:

"*Field measurements revealed the coexistence of high, medium, and low E_{abs} under high bulk-averaged M_R conditions.*"

[Comment 8] L80-84. If available, I suggest adding 1 or 2 references describing the site and its representativity.

Response: We thank the reviewer for the suggestion. References describing the Hangzhou site and its representativeness have been added (Zhang et al., 2025a; Qian et al., 2025). These are previous studies from our group, conducted at the same observation location, further supporting the representativity (Lines 83-85).

"*In this study, a suite of state-of-the-art instruments were employed to simultaneously capture the magnitude and temporal of BC M_R in Hangzhou, China (Zhang et al., 2025a; Qian et al., 2025).*"

[Comment 9] L86-109. I suggest describing a bit more each instrument alone. With the use of references, which is limited. Here some old works about SP2 describing its principle: (Stephens et al., 2003; Moteki and Kondo, 2010) and calibrations: (Gysel et al., 2011). I suggest a recent paper exploring the operational limits of the SP2 (Schwarz et al., 2022). This is the major reference of the CPMA: (Olfert and Collings, 2005). For the tandem combination of SP2 with mass analysers I suggest a relatively old review (Cross et al., 2010) and a more recent set of papers (Liu et al., 2022; Naseri et al., 2022; Zanatta et al., 2025). DMT 2011. This is an odd reference. The ACQUADAG scaling factor to fullerene should be slightly better accounted for. DMT 2011 is and odd reference. The original reference should be (Baumgardner et al., 2012; Laborde et al., 2012). Please do the

same for the ACSM. The CAPS-SSA is fully detailed by (Modini et al., 2021). Overall, these works report all the error associated with the single measurements, which will propagate substantially for the application intended in the current manuscript. None of these are described.

Response: We thank the reviewer for the suggestions regarding instrument descriptions and relevant references. All suggested references have now been added. Regarding DMT 2011, this refers to the Aquadag aerosols supplier and production year rather than a literature reference; its previous placement led to confusion, which has now been clarified. Additionally, we have provided concise descriptions of each instrument and performed a rough assessment of the propagated errors associated with individual measurements, as relevant for the applications in revised manuscript (see **[Comment 2]**).

[Comment 10] L93: I would expect the SP2 showing the multi charged particles. How exactly was the mass MBC calculated?

Response: We thank the reviewer for the comment. The treatment of multiply charged particles has already been detailed in Fig. S10 and Fig. S11 and the accompanying Supplementary Text S1. The BC core mass (M_{BC}) was calculated based on the linear relationship between the SP2 incandescence peak height and BC mass established during calibration. Relevant description has been added to the revised manuscript (Lines 135-141).

"In the subsequent data processing, measurements from the CPMA-SP2 system were first corrected for multiple charging effects. Additional corrections, including transfer function, detection efficiency, and time delay, were also applied, as detailed in the Text S1. The mass of each BC core (M_{BC_core}) was then calculated from the SP2 incandescence signal using the calibration described above, with a correction factor of 0.75 applied to the peak height (Liu et al., 2020; Liu et al., 2014; Zhang et al., 2018; Gysel et al., 2011)."

[Comment 11] L115. What model and company? Please be consistent with previous notation.

Response: We have added the model and company information for the Nephelometer to be consistent with previous notation (Aurora 3000, Acoem; see Line 161).

[Comment 12] L122: CAPS and nephelometer may well respond to PSL, especially small PSL. Truncation error may become more and more important with larger particles and especially irregular particles…such as ramified fresh BC. Please provide a small statement about it. Was truncation corrected?

Response: We thank the reviewer for the comment regarding truncation error. As reported in Madini et al. (2021), the CAPS PMssa instrument can measure extinction (σ_{ext}) and scattering (σ_{sca}) coefficients with high accuracy, with errors on the order of 1-10%. However, absorption derived via the extinction-minus-scattering method is subject to substantial subtractive error amplification, particularly for highly scattering or irregular particles such as ramified fresh BC. The main source of this uncertainty is the truncation of near-forward and near-backward scattered light that is not captured by the instrument. Using standard error propagation for subtraction, the resulting uncertainty in σ_{abs} can be estimated as $\sqrt{\sigma_{ext}^2 + \sigma_{sca}^2}$. For submicron BC particles in this study, this leads to a conservative estimate of absorption uncertainty on the order of 15-20%, accounting for both instrument precision and potential truncation effects. We did not apply explicit truncation

corrections, because our focus is on the relative enhancement of absorption with increasing M_R rather than absolute absorption values. Besides, the nephelometer measurements in this study were used solely to compare with CAPS results and assess the stability and reliability of the CAPS measurements, and no truncation correction was applied. The presence of CAPS measurement uncertainties and other contributing factors to absorption uncertainty have been noted in the Methods section (lines 149-157).

"*The aerosol extinction and scattering coefficient (Fig. S3) at wavelength of 440, 530 and 630 nm were measured by Multi-Wavelength Cavity Attenuated Phase Shift Single-Scattering Albedo Monitor (CAPS-ALB, Shoreline) (Weber et al., 2022). Absorption was calculated as the difference between extinction and scattering, with estimated uncertainties of ~1-10% for both extinction and scattering (Modini et al., 2021), leading to a conservative absorption uncertainty of ~15-20% for the submicron BC particles considered. No explicit truncation correction was applied, as the analysis focuses on the relative enhancement of absorption with M_R rather than absolute values.*"

[Comment 13] L127: Well, we "assume" that everything is working properly. This is why is important to estimate, even roughly, the uncertainty.

Response: Thank you for your comment. We agree that estimating the uncertainty is important. We have performed a rough evaluation of the uncertainties associated with the parameters discussed in the manuscript. The details of this assessment are provided in **[Comment 2].**

[Comment 14] L132-133: I have a couple of questions here. The CPMA is capable of selecting particles based on their mass to charge ratio. Hence, it is recommended, even by the manufacturer, to run the CPMA after a neutralizer/charger. From the schematics of Figure S1 it looks like there was no neutralizer. What it is the additional uncertainty of running the CMPA-SP2 setup without a charger? Single and multi-charged peaks, should be visible in the mass distribution provided by the SP2. I wonder if the authors quantified the single particle BC mass (MBC, by fitting the SP2 mass distribution, including only the first peak (single charge) or fitting the full distribution). This technical detail may influence all the result sections. Hence need to be fully and properly described. Regarding the sampling collection. Unfortunately, Figure S2 shows that the counting efficiency of the SP2 is far from 100%, especially below 100 nm (Figure S2a). It is also surprising that the SP2 counts systematically more than the CPC. I presume that some setting in the SP2 were not properly configured or that the CPC had some counting issues. Moreover, why the two incandescence detectors should have a different (linear and non-linear) mass/incandescence relationship?

Response:

1. Regarding the CPMA-SP2 multiple charge issue: We would like to clarify that the influence of multiply charged particles ($q > 1$) has been explicitly considered in our CPMA-SP2 measurements. An X-ray aerosol neutralizer (TSI 3088) was installed upstream of the CPMA to charge the aerosols before entering the classifier (Lines 121-123). Although we did not directly use the inversion code from Liu et al. (2017) or Naseri et al. (2024), we have applied an independent multiple charge correction procedure to the data, allowing us to identify and select only singly charged particles. All relevant details had already been included in Text S1 and Figure S10 and Figure S11. To facilitate readers' understanding of the data correction procedures and the CPMA-SP2 system configuration,

we have added the relevant descriptions in the Methods (Lines 135-138) and Text S1 and included the aerosol neutralizer in the schematic diagram of Figure S1.

2. Regarding CPC and SP2 particle counting: We note that the SP2 and CPC instruments have inherently different counting principles, which can lead to systematic differences in particle counts. The SP2 measures single-particle BC core properties via incandescence, whereas the CPC detects total particle number via condensation, which explains why SP2 counts can sometimes exceed CPC counts under certain conditions. Additionally, the SP2 has reduced detection efficiency for small particles, particularly below ~100 nm, as shown in Figure S2a. To ensure data reliability, our analysis was restricted to particles with BC core diameters larger than 80 nm, where the SP2 counting efficiency is sufficiently high.

3. why the two incandescence detectors have a different (linear and non-linear) mass/incandescence relationship: The two incandescence detectors of the SP2 exhibit different mass-signal behaviors due to their gain settings and the underlying physics. The low-gain detector operates within the linear dynamic range of the photomultiplier, so the incandescence signal increases linearly with BC mass. In contrast, the high-gain detector is designed to enhance sensitivity for small particles, but for larger BC masses the PMT enters a non-linear response regime, causing the signal to deviate from linearity. This difference in detector behavior is intrinsic to the SP2 design and explains why the mass-incandescence relationship appears linear for one detector and non-linear for the other.

Lines 121-123: "*In this setup, particles with known mass (M_p) selected by CPMA were injected into the SP2. An X-ray aerosol neutralizer (TSI 3088) was installed upstream of the CPMA to charge the aerosols to a Boltzmann equilibrium before entering the classifier.*"

Lines 135-138: "*In the subsequent data processing, measurements from the CPMA-SP2 system were first corrected for multiple charging effects. Additional corrections, including transfer function, detection efficiency, and time delay, were also applied, as detailed in the Text S1.*"

[Comment 15] L140, why distorted. It is attenuated due to the evaporation of absorbing-refractory material.

Response: We thank the reviewer for pointing this out. The scattering signal measured by the SP2 can indeed appear distorted due to partial evaporation of absorbing-refractory material as BC-containing particles pass through the laser beam. To address this, we have clarified the text and revised the sentence as follows (Lines 177-202):

"The measured scattering cross section ($C_{sca\_measured}$) was obtained from the SP2 using the leading-edge-only (LEO) technique, which reconstructs the scattering signal as BC-containing particles pass through the SP2 laser beam due to partial evaporation of refractory-absorbing material. The validity of this reconstruction relies on the assumption that the leading-edge data used for fitting represents an unperturbed particle, as extensively reported in previous studies."

[Comment 16] L142-146: the LEO-fit relies on many assumptions, as correctly stated by the authors. It would be nice if they could elaborate, shortly, about the reason behind these choices. Moreover, with a similar number of assumptions (density of coating and BC cores), the optical coating thickness could be derived directly from the Mp and Mbc was this performed? Are the results coherent?

Response: We thank the reviewer for the comment. The validity of the LEO-fit method indeed relies on the assumption that the leading-edge data used for fitting represents an unperturbed particle. In our study, the LEO reconstruction was used solely to obtain the scattering signal of BC-containing particles, which is necessary for multi-charged particle distribution statistics and the corresponding multi-charge corrections. For the coating thickness, we directly calculated it from the CPMA-selected particle mass (M_p) and the BC core mass (M_{BC}). Previous study have demonstrated that this approach provides higher accuracy and reliability compared with estimating coating thickness based on LEO-fit reconstruction (Naseri et al., 2024). Therefore, the LEO-fit was not used for coating thickness calculations.

Besides, we have now added a description of the LEO-fit assumptions to the main revised manuscript (Lines 197-205).

"The measured scattering cross section ($C_{sca\_measured}$) was obtained from the SP2 using the leading-edge-only (LEO) technique, which reconstructs the scattering signal as BC-containing particles pass through the SP2 laser beam due to partial evaporation of refractory-absorbing material. The validity of this reconstruction relies on the assumption that the leading-edge data used for fitting represents an unperturbed particle, as extensively reported in previous studies (Liu et al., 2014; Zhang et al., 2016; Brooks et al., 2019; Gao et al., 2007; Zhang et al., 2020). Note only particles with successfully fitted LEO signals are considered in the optical property calculations."

[Comment 17] L149: define the MAC.

Response: We thank the reviewer for the comment. We have clarified the definition of the mass absorption cross section (MAC) in the revised manuscript (Lines 212-215).

"*The light absorption enhancement of BC-containing particles is defined as the ratio of the mass absorption cross section (MAC) of the coated and uncoated BC-containing particles (Eq. 3), Here, MAC is defined as the particle light absorption cross section normalized by the BC mass, representing the light absorption per unit mass of BC.*"

[Comment 18] L154: I like the approach of deriving the MAC of uncoated BC using this extrapolation. However, this MAC (no units) for uncoated BC results to be slightly higher than previous estimations in European urban (Savadkoohi et al., 2024) rural (Zanatta et al., 2016) and the canonical 7.5 m/ g of (Bond and Bergstrom, 2006). This could also be due to the high variability of MAC itself across sites and seasons, but also to strong uncertainties related with MAC (absorption and BC mass) and bulk MR (width of the distribution of particles exiting the CPMA and method to quantify the single particle mass with the SP2). Overall, I notice a lack in providing context to these findings and assumptions. The MACBC_core is fundamental to all the results presented in the paper, hence, even small errors may substantially modify the quantification of the enhancement. The authors must provide more details on their methods and uncertainties, and put all of these consideration with the context of recent literature.

Response: We thank the reviewers for their valuable comments. The reported $MAC_{BC\_core\_measured}$ of 9.08 m^2/g at 630 nm corresponds to uncoated BC, representing the intrinsic light absorption efficiency of BC without influence from any coating materials. This value is independent of the mixing state and reflects the inherent optical property of BC itself. The slightly higher value compared to 7.5 m^2/g reported by Bond and Bergstrom (2006), can be attributed to several plausible

factors, including variations in BC morphology (aggregate compactness, fractal dimension, and cluster structure), instrumental and calibration effects (SP2 calibration with Aquadag standards, detector response, and limited sampling statistics), and site-specific environmental variability (differences in BC sources and seasonal atmospheric conditions).

Regarding uncertainties, we conducted a quantitative error propagation analysis. Considering relative uncertainties of 15-20% for absorption, 10% for SP2-measured BC mass, and the extrapolation standard error of 0.54 $m^2\ g^{-1}$, the $MAC_{BC\_core\_measured}$ uncertainty is estimated to be ~19% - 23%. Consequently, the $E_{abs\_measured}$ has a propagated uncertainty of ~26% - 32% (Text S2). This analysis demonstrates that, while the $MAC_{BC\_core\_measured}$ value is slightly higher than the commonly cited value of 7.5 $m^2\ g^{-1}$ (Bond and Bergstrom, 2006), it remains well constrained and representative of the local atmospheric BC properties.

We have added this discussion in the revised manuscript (Lines 217-228), providing details on methods, uncertainties, and comparisons with recent literature to give proper context and highlight the robustness of our findings (Text S2).

"*The value of $MAC_{BC\_core\_measured}$ was obtained by extrapolating $MAC_{BC\_coated\_measured}$ to the limit of bulk-averaged $M_R = 0$ using linear regression. The $MAC_{BC\_core\_measured}$ at wavelength of 630 nm was 9.08 ± 0.53 $m^2\ g^{-1}$ (mean ± 90% confidence Interval) (Fig. S6). Based on our error propagation analysis, which accounts for measurement uncertainties in particle absorption and BC mass as well as the standard error of the extrapolation, the estimated uncertainty of $MAC_{BC\_core\_measured}$ is approximately 19-23% (Text S2). And the uncertainty of $E_{abs\_measured}$ is approximately 26 - 32% (Text S2). For comparison, the $MAC_{BC\_core\_measured}$ is slightly higher than the value of 7.5 $m^2\ g^{-1}$ recommended by Bond and Bergstrom (2006) but still within the range reported by other study (~6.5 - 17 $m^2\ g^{-1}$) (Zanatta et al., 2016), likely due to variations in measurement methods, and site-specific atmospheric conditions.*"

[Comment 19] L169-175: I am genuinely confused on how the MACbc introduced in equation 3 was calculated. Use a logic order when presenting variables. Is the MAC of equation 4 the same presented in equation 3?

Response: We thank the reviewer for the comment. We apologize for the confusion caused by the previous equation. The corrected equation has now been included in the revised manuscript. The formula has been revised as follows:

$$E_{abs\_measured} = \frac{MAC_{BC\_coated\_measured}}{MAC_{BC\_core\_measured}} \quad (3)$$

$$MAC_{BC\_coated\_modeled} = \frac{\sum_i MAC_{BC\_coated\_modeled,i} \times M_{BC\_core,i}}{\sum_i M_{BC\_core,i}} \quad (4)$$

[Comment 20] L174: The nomenclature is a bit confusing here. MACBC-core of line 174 is the same used int Equation 3 ? Or the MA_core, presented in Line 154 is used in equation 3? So, the "Mie MACBC_core" is it similar to 9.08 m2 /g. This aspect is extremely important and influences with a different weight MACMie and MAC observed with a different weight, especially in figure 2d where the delta enhancement is presented.

Response: We thank the reviewer for pointing out the confusion in the nomenclature. We fully agree

that the inconsistent notation could affect the interpretation of Equation 3. In the revised manuscript, we have thoroughly standardized and clarified all relevant symbols. The updated nomenclature and corresponding explanations are now explicitly provided in the Methods 2.2 and 2.3.

[Comment 21] L188: At what wavelength are these values provided. Could the authors state something about the absorption enhancement at different wavelengths? The high presence of organic material may change the Eabs at lower wvalenght?

Response: We thank the reviewer for this insightful comment. All absorption-related parameters and the subsequent analysis of E_{abs} in our study are based on the optical measurements at 630 nm. This clarification has been explicitly stated in the revised manuscript. (Lines 157-159 and Lines 261-265)

Lines 157-159: "*In this study, only the measurements at 630 nm were used for subsequent analysis, as this wavelength is minimally affected by brown carbon absorption.*"

Lines 261-265: "*The average $E_{abs\_measured}$ during the sampling period in Hangzhou is 1.28 ± 0.02 (mean ± 90% confidence Interval, the same below) at wavelength of 630 nm, and the bulk-averaged M_R is 3.32 ± 0.06, with average $E_{abs\_measured}$ values of 1.09 ± 0.02, 1.84 ± 0.07, and 1.55 ± 0.04 for Case 1, Case 2, and Case 3, respectively.*"

[Comment 22] L220-221: I recommend caution when mentioning morphology, especially in a section title. This scattering cross-section ratio is a far approximation for morphology assessment. It may be a proxy, but nothing more and must be confirm by real morphology observations such as microscopy fractal dimension or, at least DMA/CPMA density/fractal exponent measurements.

Response: We thank the reviewer for the helpful comment. A similar concern was also raised by Reviewer 2 (Comment 3). In response, we have removed the morphology-related descriptions in both the Abstract and the main text. These statements have been revised to explicitly reflect that our correction scheme is based on the M_R-dependent optical transition behavior, rather than particle morphology. We believe these changes improve the accuracy and clarity of the revised manuscript. Please see the revised manuscript.

[Comment 23] L222-223: what is the meaning of the sentence?

Response: We thank the reviewer for this comment. The sentence serves as a summary and overview for this subsection, providing a general introduction to the subsequent descriptions. To improve clarity and readability, we have revised the sentence in the manuscript as follows (Lines 315-318):

"*The coating-to-BC mass ratio (M_R) and the ratio of measured to modeled scattering cross sections were used to quantify the mixing state and associated optical transitions behavior of BC-containing particles, with M_R serving as an important indicator of BC aging.*"

[Comment 24] L239: The authors states that the difference between observed and modelled enhancement depends on the variability of the standard deviation of MR. First why the log10(Mr) was used ? Second, I all honesty, it is difficult to observe any sort of correlation in the scatterplot presented in figure 2d. Especially considering that correlation coefficient and slope changes substantially among the periods. In my opinion, Figure 2s does not support the claims of the authors.

Response: We appreciate the reviewer's insightful question.

1. To why use $\log_{10}(M_R)$: The logarithmic transformation of M_R ($\log_{10}(M_R)$) was applied to reduce the strong skewness in the M_R distribution, which typically spans several orders of magnitude. Using $\log_{10}(M_R)$ allows the data to approximate a normal distribution, making the standard deviation a more representative measure of heterogeneity. This approach is also consistent with previous studies (Fierce et al., 2020; Huang et al., 2024).

2. To Figure 2: We appreciate the reviewer's careful evaluation of Fig. 2d. We would like to clarify that the purpose of this figure was not to demonstrate a statistically strong linear correlation. Instead, the figure was intended to illustrate the overall trend that the ΔE_{abs} ($E_{abs\_uniform}$-$E_{abs\_measured}$) increases with increasing SD when considering all cases together. The previously shown regression line was only for visual guidance and may have unintentionally implied a stronger correlation.

Importantly, our result is consistent with earlier studies that used mixing-state entropy to characterize BC coating diversity (Zhao et al., 2021; Riemer et al., 2019; Zeng et al., 2024). These studies reported that the model-measurement deviation in E_{abs} decreases as the mixing-state entropy approaches 1, meaning that larger variability in BC coating (higher heterogeneity) leads to larger bias between modeled and measured E_{abs}. This is in good agreement with the trend observed in our analysis.

To avoid any potential misunderstanding, we have removed the regression equations from Fig. 2d and revised the corresponding text in the manuscript to clarify this point (Lines 327-337). We hope this revision more accurately reflects our intended message.

"*The results showed that the SD of Case 1 (0.63 ± 0.004) was greater than that of Case 3 (SD = 0.52 ± 0.012), followed by Case 2 (SD = 0.48 ± 0.005). In contrast, the $E_{abs\_measured}$ exhibited an opposite trend, suggesting that greater M_R heterogeneity of BC-containing particles leads to a lower $E_{abs\_measured}$. As shown in Fig. 2d, the discrepancy between the modeled (uniform core-shell Mie model) and the measured E_{abs} increases with SD, with this trend being most pronounced in Case 1, where M_R heterogeneity is highest. This suggests that greater M_R heterogeneity may lead to larger deviations from the uniform core-shell assumption, thereby increasing the mismatch between the modeled and measured E_{abs}. Such discrepancies likely due to the uniform core-shell model's simplified treatment of M_R heterogeneity in BC (Romshoo et al., 2024; Wang et al., 2021).*"

[Comment 25] L254: please provide the wavelength

Response: We thank the reviewer for the careful examination. The wavelength has now been added in the revised manuscript. Specifically, the measurement was conducted at 1064 nm, and this information has been included in Line 349-351 accordingly.

"*Fig. 3 presents the variation of the ratio $C_{sca\_measured}/C_{sca\_modeled}$ at wavelength of 1064 nm with M_R under different M_p.*"

[Comment 26] L252-276: although the results shown in figure 3 are interesting, this section is very confusing. It is hard to understand what causes the decrease in the transition regime. Try to restructure your though in a more logic process. Could this "transition state" represent the compaction due to coating formation. This sort of natural process will reduce the optical and geometrical cross section of the particles. It is usually observed in chamber studies (e.g. Schnaiter et al., 2005; Zanatta et al., 2025) and rarely, up to my knowledge, observed in ambient conditions

(Bhandari et al., 2019). This process description could be developed further.

Response: We thank the reviewer for this valuable comment. We have clarified in the revised manuscript that the observed "transition state" reflects the BC aging process. During clean periods, limited secondary formation and low coating material mean that BC particles require more mass to reach an optically core-shell-like state, resulting in higher M_R values for transitional-state particles. In contrast, during polluted periods, enhanced secondary formation produces sufficient coatings, so BC reaches the core-shell configuration at lower M_R. These differences in coating availability and atmospheric processing allow us to infer the progression of BC aging from optical observations, even though direct morphological compaction is not measured. Note while chamber studies (Zanatta et al., 2025; Schnaiter et al., 2005) directly observe coating-induced compaction, our study demonstrates that field-based optical transitions provide insight into particle aging under ambient conditions. These revisions improve the physical interpretation of the results. Incorporating the comments from other reviewers, this section has been substantially revised; see Section 3.2 and 3.3 for details.

[Comment 27] Section 3.3 soffers a similar issue with readability. I am not fully convinced by the reasoning behind the period separation. Only period 2 looks different from the others.

Response: We thank the reviewer for this comment. In response, and based on feedback from you and the other reviewers, we have substantially revised Section 3.3 to improve readability and clarify the reasoning behind the different period. The revisions provide a more physically grounded explanation of the different periods and their classification. Please see the revised manuscript (Section 3.3, Lines 383-495) for details.

[Comment 28] F1: are these enhancement measured all at the same wavelength?

Response: We appreciate the reviewer's insightful question. As different instruments were employed in these studies, the E_{abs} shown in Figure 1 were not measured at exactly the same wavelength. However, to ensure consistency and minimize the influence of brown carbon (BrC) on the estimation of E_{abs}, we selected measurements obtained within the visible–near-infrared range for comparison. This wavelength selection greatly reduces the potential spectral bias associated with BrC absorption. The specific wavelengths used for each instrument have been summarized in Table S2.

[Comment 29] F2: please improve the labelling of the axis. Number counts and SD of…?

Response: We thank the reviewer for the helpful suggestion. The axis labels in Figure 2 have been revised to improve clarity. Specifically, "Number counts" has been replaced with "Normalized number counts of BC-containing particles", and "SD of …" has been updated to "SD of $\log_{10}(M_R)$".

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