Reviewer #1

This manuscript presents a novel hybrid framework that couples in situ PAX with ISS to apportion aerosol light absorption into BC, BrC, and lensing-related contributions. The topic is relevant and timely because accurate separation of these components is indeed important for constraining aerosol radiative forcing and for interpreting absorption enhancement by internally mixed carbonaceous particles. However, the current version still has room for improvement in several aspects, particularly in the clarity of the methodological framework, the organization of key definitions and equations, and the treatment of assumptions and uncertainties. Strengthening these points would improve both the rigor and readability of the manuscript and help support the main conclusions more convincingly.

We sincerely thank the reviewer for the careful evaluation and constructive comments on our manuscript. We are encouraged that the reviewer recognizes the scientific significance of the proposed PAX-ISS hybrid framework for distinguishing the contributions of BC, BrC, and the lensing effect. We also agree that the clarity of the methodological framework, the organization of key definitions and equations, and the treatment of assumptions and uncertainties can be further improved. In the revised manuscript and Supplement, we have addressed each comment point by point and described the corresponding changes below.

1. The manuscript treats the ISS-derived absorption as a lensing-free state containing only BC and BrC absorption. This assumption is fundamental to the entire framework. However, the paper does not provide direct validation that the ISS treatment removes lensing without simultaneously altering the intrinsic optical behavior of BC and BrC, or changing the original particle/coating structure in a way that biases the result. The authors should more rigorously justify the physical meaning of this “uncoated” state and discuss possible artifacts introduced by solvent processing.

We thank the reviewer for raising this important point. We agree that treating the ISS-derived absorption as a lensing-free absorption state is fundamental to the PAX-ISS framework proposed in this study. We address the reviewer’s concern from two

perspectives.

First, regarding the reliability and feasibility of the methodological framework, both PAX, which provides in situ absorption measurements, and ISS, which is based on an integrating sphere system, are well-established techniques (Lack et al., 2006; Moosmüller et al., 2009; Sun et al., 2017, 2021; Wonaschütz et al., 2009). Our study couples these two established techniques to apportion total aerosol absorption into three absorption-related contributors: BC, BrC, and the lensing effect.

Specifically, PAX does not require particle deposition on filters and can therefore avoid artifacts caused by filter scattering and particle shadowing. PAX-derived absorption is widely regarded as a reliable measurement of ambient aerosol absorption and has often been used as a reference in aerosol light-absorption measurements, instrument intercomparisons, and studies of BC absorption enhancement (Arnott et al., 2003, 2005; Lack et al., 2006, 2009; Li et al., 2023; Liu et al., 2015; Pokhrel et al., 2017). Regarding the ISS method, its development has progressed from BC absorption measurements to BC/BrC optical separation and, more recently, to the retrieval of lensing-free absorption. Fischer (1970) first applied the integrating sphere (IS) technique to quantify aerosol absorption. Andre et al. (1981), Heintzenberg (1982), and Hitzenberger et al. (1996) subsequently combined IS with solvent systems, namely ISS, to measure aerosol absorption properties or BC concentrations. Hitzenberger and Tohno (2001) further improved the ISS system by introducing a water/isopropanol rinsing step before acetone dissolution for studies of ambient BC concentrations. After BrC was recognized as an important light-absorbing component, Wonaschütz et al. (2009) used carbon black (CarB) and humic acid sodium salt (HASS) as optical reference materials for BC and BrC, respectively, and established the ISS method to distinguish the absorption contributions of BC and BrC in atmospheric aerosols. Sun et al. (2017, 2021) applied the ISS method to samples from residential solid-fuel combustion in China to quantify the emission factors and light-absorption contributions of BC and BrC. More recently, Li et al. (2023) coupled ISS with in situ PAX absorption observations to quantify differences in absorption efficiency between coated and uncoated aerosols and explicitly described the mechanism as “de-lensing through solvent dissolution and

solvent de-refraction". Building on these methodological advances, the present study uses the hybrid PAX-ISS framework to resolve the mixed light absorption into contributions from BC, BrC, and the lensing effect (lines 89–102). In the revised manuscript, we have supplemented the description of this methodological development context (lines 124–139).

Second, regarding whether solvent treatment may significantly alter the intrinsic absorption characteristics of BC or BrC while removing the lensing effect, available studies that used solvent dissolution to remove coatings have not reported solvent-induced changes in the absorption characteristics of BC or BrC (Chen et al., 2017; Cui et al., 2016; Kong et al., 2024; Lan et al., 2024). From a theoretical perspective, the 3 mL solvent mixture used in this study consisted of acetone, water, and 2-propanol at a ratio of 5:4:1. Because BC is generally considered chemically inert and poorly soluble in such solvent mixtures (Bond et al., 2013; Petzold et al., 2013; Wonaschütz et al., 2009), it is unlikely that the absorption properties of the BC core would be altered. For BrC, light absorption mainly arises from organic chromophores with conjugated π-electron systems and aromatic functional groups (Alang and Aggarwal, 2024; Laskin et al., 2015, 2025). These chemical structures are not expected to be destroyed or reconstructed by the acetone/water/2-propanol solvent mixture under room-temperature conditions (Alang and Aggarwal, 2024; Laskin et al., 2015, 2025). Moreover, to the extent that solvent treatment could induce systematic optical shifts, the same solvent environment and measurement protocol applied to both ambient samples and reference materials (CarB and HASS) should reduce the related bias.

2. Could the authors clarify how much coating is actually removed and discuss whether the observed waveband-dependent differences may partly reflect size changes rather than lensing alone? This issue may be particularly relevant in light of recent studies emphasizing the importance of size-resolved constraints and particle microphysics for aerosol absorption estimates (doi:10.1029/2025GL117418; doi:10.1002/2017JD027833).

We thank the reviewer for this valuable comment. The comment raises two

specific concerns: how much coating is actually removed, and whether the observed waveband-dependent differences may partly reflect size changes rather than lensing alone. We address these two concerns separately below.

Regarding the first concern—how much coating is actually removed—we directly cite the description by Wonaschütz et al. (2009): "In the IS method, samples are either suspended in a liquid or surrounded by a liquid. Soluble transparent material will be dissolved leaving the BC particles either without coatings or with coatings of insoluble material. The refractive indices of these coatings relative to the liquid, however, are small for typical aerosol material. Most organic aerosol substances have refractive indices around 1.4 (D'Almeida et al., 1989), and if they are submerged in the liquids (we use 2mL acetone+2mL of an80%water and 20% 2-propanol mixture; refractive index of mixture: 1.35) the relative refractive index of the organics drops from 1.4 in air to 1.04 in the liquid, making the coating nearly 'invisible' to the incident light. Little enhancement of absorption is expected in this case". In this context, it is neither feasible nor necessary to provide a quantitative value for "how much coating is removed". To avoid potential ambiguity, we now describe the ISS-treated state more precisely as a "solvent-mediated lensing-free state" in the revised manuscript (lines 8–9 and 95).

Regarding the second concern, i.e., whether the observed waveband-dependent differences may partly reflect size changes rather than lensing alone, this issue cannot be directly resolved with the current PAX-ISS instrumentation, because the framework does not provide simultaneous size-resolved constraints. Nevertheless, we fully acknowledge the influence of aerosol size on aerosol absorption estimates, as reflected in the following two revisions:

(1) In the revised manuscript, we have refined the original statement "However, previous source-related experiments and Mie model calculations (Li et al., 2016; Liu et al., 2018b; Wang et al., 2021b) have reported a wide range of values for AAE_{BC} (e.g., 0.6−1.6) due to various confounding factors, implying the possibility of *AAE* values below 1.0 for coated aerosol" to "However, previous source-related experiments and Mie model calculations (Li et al., 2016; Liu et al., 2018b; Wang et al., 2021b) have reported a wide range of values for AAE_{BC} (e.g., 0.6−1.6) due to various confounding

factors (e.g., aerosol size, morphology, mixing state, or core-shell structure, etc.) (Curci et al., 2019; Guan et al., 2026; Helin et al., 2021; Liu et al., 2018; Zhang et al., 2020), implying the possibility of *AAE* values below 1.0 for coated aerosol" (lines 284–289). This revision explicitly includes aerosol size among the relevant confounding factors, thereby acknowledging the established view in the literature.

(2) In response to the editor's earlier request concerning aerosol-size effects, we have added a specific discussion of the caveats and limitations of the current PAX-ISS framework in the Conclusions section, where we explicitly state that our approach does not provide size-resolved insights:

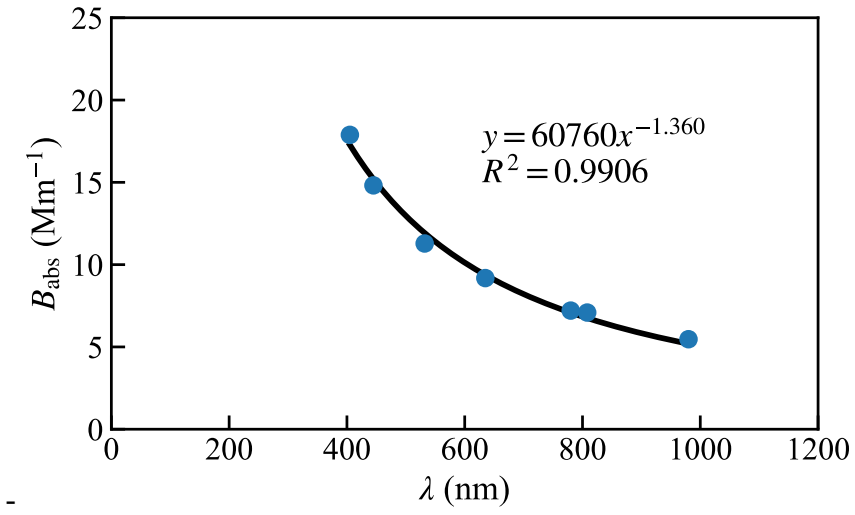
"The PAX-ISS hybrid method offers a powerful tool for deconvolving the complex sources of aerosol absorption without requiring detailed assumptions or information about particle microphysical properties, such as size distributions, morphology, mixing state, or core-shell structure. This characteristic is, on one hand, an advantage over methods that rely on such parameters. On the other hand, it constitutes a limitation, as our framework currently cannot directly link the observed absorption enhancement to these fundamental particle properties. For instance, while particle size critically influences the magnitude and spectral dependence of absorption enhancement (e.g., Chen et al., 2025; Fu et al., 2024), our approach does not provide size-resolved insights" (lines 494–503).

3. The authors use PAX to measure total coated absorption at 870 nm and then extrapolate that value to 380-880 nm using a single AAEcoated. This reconstructed coated spectrum is then directly used in the subtraction that defines the lensing contribution. The concern is that coated aerosol absorption in this study is itself a mixture of BC, BrC, and enhancement effects, all of which may have different spectral behavior. It is therefore unclear whether a single AAE is sufficient to represent the full spectral dependence.

We thank the reviewer for raising this important concern. Coated aerosol absorption is indeed the sum of BC absorption, BrC absorption, and coating-induced enhancement, each of which may have its own spectral dependence. In this study,

AAE_{coated} here was used only to reconstruct $B_{abs\_coated}(\lambda)$, not to retrieve $B_{abs,BC}(\lambda)$ and $B_{abs,BrC}(\lambda)$. Because the PAX instrument used here is a single-wavelength device (PAX-870), only the in situ $B_{abs\_coated}(870)$ can be measured directly. We therefore used the power-law relationship between wavelength and the corresponding light-absorption signals measured by the multi-wavelength thermal/optical carbon analyser (TOA) to calculate AAE_{coated} (Chen et al., 2015; Chow et al., 2018, 2021; Shen et al., 2025). $B_{abs\_coated}(870)$ was then extrapolated to absorption coefficients at wavelengths between 370 and 880 nm, namely $B_{abs\_coated}(\lambda)$, constrained by AAE_{coated} (lines 182–189).

To test whether an overall AAE_{coated} can reasonably represent the full spectral dependence over 370–880 nm, we selected the data from 19 February 2023 as an example. As shown in the figure below and in Figure S3 of the Supplement, the single AAE_{coated} derived from the seven-wavelength data reasonably captures the broad spectral dependence of B_{abs_coated}, as the TOA-measured absorption coefficients are generally close to those reconstructed using AAE_{coated}.



Note: Absorption coefficients and the fitted curve measured by the DRI Model 2015 Multi-Wavelength Thermal/Optical Carbon Analyser on 19 February 2023.

We acknowledge that extrapolating $B_{abs\_coated}(\lambda)$ using the AAE method introduces a certain degree of uncertainty. In the revised manuscript, we explicitly state that extrapolating single-wavelength PAX measurements to other wavelengths is a source of methodological uncertainty (lines 187–189). The uncertainty associated with AAE was estimated to be 3.6% and is included as one contributor to the overall uncertainty

in the calculation of the absorption enhancement factor. The detailed calculation procedure is provided in the Supplement section entitled "Assessment of uncertainty of PAX-ISS methodology."

4. The DWIC method uses 420 and 650 nm as the two anchor wavelengths, described as BrC-sensitive and BC-dominated, respectively. However, the manuscript does not explain why this pair was selected over other possible combinations, nor whether the results depend strongly on this choice. This should be justified more carefully.

We appreciate this valuable question. The Double-Wavelength Iterative Calculation (DWIC) approach is based on the difference in spectral dependence between BC and BrC. Two reference materials are introduced: CarB for BC and HASS for BrC. Two wavelengths in the short- and long-wavelength ranges are then used to separate the absorption contributions of BC and BrC through iterative calculation. Therefore, the key requirement of the DWIC method is to designate a wavelength pair with sufficient spectral separation and different sensitivities to BC and BrC absorption.

Previous studies have not fixed a single wavelength pair for DWIC. Instead, eligible wavelength pairs have been selected flexibly according to the wavelength range of the specific instrument and the spectral characteristics of the reference materials (Sun et al., 2017; Wang et al., 2021; Wonaschütz et al., 2009).

In this study, the selection of 420 and 650 nm was based mainly on the following considerations. The wavelength 420 nm lies in the short-wavelength region of visible light, where BrC has much higher light-absorption efficiency than at 650 nm; thus, it can serve as a BrC-sensitive wavelength. By contrast, 650 nm lies in the longer-wavelength visible region, where BrC absorption is much weaker than at 420 nm, whereas BC still maintains strong absorption. Therefore, 650 nm can be regarded as a BC-dominated wavelength. In addition, the instrument provides stable light-intensity signals at both wavelengths, which also supports this choice. In the revised version, we give a special explanation of the wavelength-pair choice (lines 227–235).

5. The DWIC separation relies on carbon black and humic acid sodium salt as surrogate

standards for BC and BrC. This is practical, but the paper does not discuss how representative these materials are for ambient Beijing aerosol in different seasons. The limitations of these proxies should be addressed explicitly.

We thank the reviewer for this constructive comment. Previous studies have used these two substances as proxies because of their similarities to observed BC and BrC, particularly in their optical properties. For example, CarB was used as a proxy for BC in diesel exhaust by Medalia et al. (1983), and HASS was used as a proxy for BrC in wood combustion by Wonaschütz et al. (2009). In two previous studies by our group, CarB and HASS were used as proxies for BC and BrC, respectively, in residential solid-fuel combustion samples (Sun et al., 2017; 2021). We adopted the same rationale in the current study. In practice, the most critical requirement for reference materials in this methodology is that they separately represent the distinct wavelength dependencies of BC and BrC, thereby serving as a bridge to decompose the total absorbance of both species. Fully reproducing the chemical composition of actual atmospheric particles is neither feasible nor necessary for this purpose.

In the revised manuscript, we have added explanatory sentences on the selection of these two reference materials (lines 207–214). The details of the DWIC method are also provided in the Supplement (see Section S4).

6. The apparent lensing term is derived as the difference between the extrapolated coated absorption and the ISS-derived uncoated absorption. Because this residual may be sensitive to accumulated uncertainties, especially at short wavelengths, could such uncertainties substantially affect the retrieved lensing-related term and thereby the robustness of the conclusions?

We appreciate the reviewer's attention to this critical issue. We fully agree that the apparent lensing-related absorption ($B_{\mathrm{abs,lensing}}(\lambda)$) is not measured independently by an instrument; rather, it is a residual term obtained by subtracting the ISS-derived uncoated absorption ($B_{\mathrm{abs\_uncoated}}(\lambda)$) from the extrapolated coated absorption ($B_{\mathrm{abs\_coated}}(\lambda)$):

$$B_{\text{abs,lensing}}(\lambda) = B_{\text{abs\_coated}}(\lambda) - B_{\text{abs\_uncoated}}(\lambda)$$

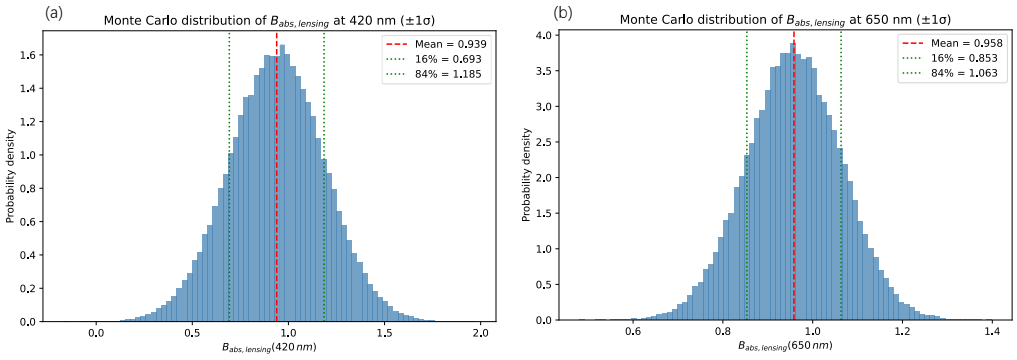
where

$$B_{\text{abs\_coated}}(\lambda) = B_{\text{abs\_coated}}(870) \times \left(\frac{\lambda}{870}\right)^{-AAE_{\text{coated}}}$$

$$B_{\text{abs\_uncoated}}(\lambda) = \frac{100 \times ABS_{\text{ISS}}(\lambda) \times A}{CF \times F \times T}$$

Therefore, the uncertainty of $B_{\text{abs,lensing}}(\lambda)$ does not originate from a single measurement process, but from the combined uncertainties of multiple variables, including $B_{\text{abs,coated}}(870)$, AAE_{coated}, CF, and $ABS_{\text{ISS}}(\lambda)$. To assess the uncertainty inherent in the PAX-ISS methodology comprehensively, we used Monte Carlo uncertainty propagation to quantify the uncertainty associated with the key result—the absorption enhancement factor (E_{abs})—and obtained the probability distribution of $E_{\text{abs}}(\lambda)$ using data from 19 February 2023. The uncertainty of $B_{\text{abs,lensing}}(\lambda)$ was also incorporated. Details are provided in the Supplement section entitled "Assessment of uncertainty of PAX-ISS methodology."

Here, we present the results for the uncertainty of $B_{\text{abs,lensing}}(\lambda)$ only, as shown in the figure below and in Figure S5. At 420 nm, the mean $B_{\text{abs, lensing}}$ is 0.939 ± 0.246 $Mm^{-1,}$ corresponding to an uncertainty of 26.2%; at 650 nm, the mean $B_{\text{abs, lensing}}$ is 0.958 ± 0.105 Mm^{-1}, corresponding to an uncertainty of 11.0%.



Note: Probability distribution of $B_{\text{abs,lensing}}$ at 420nm (a) and 650nm (b) on 19 Febrary 2023). The ±1σ range (68% probability) is used to represent uncertainty.

7. The hybrid framework combines real-time in situ PAX observations with offline filter-based ISS measurements collected over 23.5-hour sampling periods. The authors implicitly assume that both datasets represent the same aerosol population and can be merged into a common spectral decomposition framework. The concern is that the manuscript does not discuss possible mismatch between online and offline observations, since the method depends on cross-platform consistency, this issue deserves much more attention.

We appreciate the reviewer's valuable feedback. The reviewer is correct that a key assumption of the PAX-ISS hybrid framework is that online PAX observations and offline filter-based ISS measurements represent a comparable aerosol population and can therefore be merged into a common spectral decomposition framework. Specifically, the shared basis of the two datasets is that they represent the same daily integrated aerosol population at the same sampling site and within the same particle size range over a 23.5 h sampling period, allowing 30 min for filter replacement. The framework does not require exact minute-to-minute or hour-to-hour alignment. This consistency has been explicitly clarified in the revised manuscript (lines 196–201). We have also added a quality-control section to the Supplement, including procedures to ensure that PAX and ISS evaluate the same aerosol population (see Section S5).

8. The CF is important because it converts ISS absorption into a quantity comparable with PAX. Is CF wavelength dependent? A brief summary of the uncertainty introduced by CF in the final apportionment results would also be helpful.

We thank the reviewer for highlighting this important issue. In the ISS method, a filter punch is placed inside the integrating sphere, where light can be absorbed by BC and BrC from multiple directions. In contrast, PAX measures absorption corresponding to a conventional one-way optical path. Thus, the primary physical role of CF is to convert the multi-directional absorption measured within the integrating sphere into a conventional one-way absorption quantity that is comparable with PAX.

Regarding whether CF is wavelength dependent, CF primarily reflects the relationship between two optical measurement geometries: ISS-measured multiple-

angle absorption and PAX-measured conventional one-way absorption. In principle, this geometric conversion is not expected to depend strongly on wavelength.

Regarding the uncertainty introduced by CF into the final apportionment results, the CF value may be affected by several practical factors, including the state of the integrating sphere, internal wall reflectivity, cuvette position, light-source stability, detector response, solvent background, and system assembly conditions. These factors may introduce uncertainty in practical experiments. We refer readers to our previous study (Li et al., 2023), where the methodology for determining CF was described in detail. A more comprehensive assessment of the composite uncertainty of the PAX-ISS hybrid methodology is provided in the Supplement section "Assessment of uncertainty of PAX-ISS methodology."

9. Section 3.3.2 compares the derived enhancement factors with previous literature values from different sites and methods. Could the authors clarify more explicitly whether the definitions of enhancement are fully consistent across these studies?

We thank the reviewer for raising this important issue. There is broad agreement on the standard definition of the enhancement factor, E_{abs}, which refers to the enhancement of BC absorption caused by the lensing effect of coatings:

$$E_{abs} = \frac{B_{abc,BC}+B_{abc,lensing}}{B_{abc,BC}} = 1 + \frac{B_{abc,lensing}}{B_{abc,BC}} \quad (A)$$

where $B_{abs,lensing}$ represents the absorption equivalent due solely to the lensing effect, and $B_{abs,BC}$ represents the absorption coefficient of BC in the absence of lensing.

If BC is assumed to be the only light-absorbing carbonaceous component in atmospheric aerosols, the absorption enhancement factor for BC can be understood simply as the ratio of absorption with and without a coating layer:

$$E_{abs} = \frac{B_{abs\_coated}}{B_{abs\_uncoated}} \quad (B)$$

where, in this expression, the numerator B_{abs_coated} is $B_{abc,BC} + B_{abc,lensing}$, and the denominator $B_{abs\_uncoated}$ is $B_{abc,BC}$. This ratio can be obtained through thermal denuding or solvent dissolution.

However, as BrC has received increasing attention in recent years, results obtained from Equation (B) using either thermal denuding or solvent dissolution approaches are effectively equivalent to those from Equation (C):

$$E_{\text{abs}} = \frac{B_{\text{abs\_coated}}}{B_{\text{abs\_uncoated}}} = \frac{B_{\text{abs,BC}}+B_{\text{abs,BrC}}+B_{\text{abs,lensing}}}{B_{\text{abs,BC}}} \quad \text{(C)}$$

Compared with the standard definition of E_{abs} in Equation (A), the numerator in Equation (C) contains an additional contribution, namely $B_{\text{abs,BrC}}$. As a result, E_{abs} values calculated using Equation (C) are theoretically higher than those calculated using Equation (A). Numerous studies have calculated E_{abs} following the logic of Equation (C), including studies using thermal denuding (Cappa et al., 2012; Fu et al., 2021; Xie et al., 2019) and solvent dissolution (Chen et al., 2017; Cui et al., 2016; Lan et al., 2024).

Another ISS-based approach directly divides the absorption coefficient measured by PAX ($B_{\text{abs,BC}}+B_{\text{abs,BrC}}+B_{\text{abs,lensing}}$) by that obtained from ISS ($B_{\text{abs,BC}}+B_{\text{abs,BrC}}$) to derive E_{abs} (Li et al., 2023):

$$E_{abs}=\frac{B_{\text{abs\_coated}}}{B_{\text{abs\_uncoated}}}=\frac{B_{\text{abs,BC}}+B_{\text{abs,BrC}}+B_{\text{abs,lensing}}}{B_{\text{abs,BC}}+B_{\text{abs,BrC}}}=1+\frac{B_{\text{abs,lensing}}}{B_{\text{abs,BC}}+B_{\text{abs,BrC}}} \quad \text{(D)}$$

In this definition, both the numerator and the denominator include BrC absorption. Compared with Equation (A), Equation (D) represents the enhancement relative to ($B_{\text{abs,BC}}+B_{\text{abs,BrC}}$) rather than solely relative to $B_{\text{abs,BC}}$. Consequently, values calculated using Equation (D) are slightly lower than those calculated using Equation (A).

Based on the above analysis, the expected high-to-low sequence of reported E_{abs} values is (C) > (A) > (D) in theory.

Each equation above is valid within its own methodological framework and provides useful insight into BC absorption enhancement, provided that its meaning is clearly understood. We have added Section S6 to the Supplement to compare different operational definitions. In the revised manuscript, we have also added the following sentence: "Note that the definitions of E_{abs} vary a little in literature depending upon how to remove coatings or how to deal with BrC (a comprehensive comparison on these operational definitions is provided in Section S6 of the Supplement)" (lines 424–426).

10. Some key terms and formulas are explained repeatedly after already being defined,

which affects the clarity of the presentation. It would be helpful to streamline these repeated explanations and better organize the related equations and terminology so that the manuscript is easier to follow. Please check carefully

We appreciate this helpful suggestion. We carefully revised the manuscript to remove repeated explanations and to streamline the organization of the related definitions, equations, and terminology.

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Reviewer #2

Major Comments:

This manuscript combines in situ and filter-based techniques to address important questions about aerosol optical properties. They present long term sampling of Beijing with important insights about how seasonal changes can affect light absorption by aerosol beyond the first-order changes in black carbon concentrations.

However, while it is clear that great care was taken with the calibration and execution of each individual analyses, the propagation of uncertainty for each method is not presented. Without a thorough discussion of the uncertainties in each step of the process, the conclusions remain unsupported. This is a common challenge in trying to establish the relative contributions of different aerosol absorption mechanisms, and one that I hope that this dataset and the ISS-PAX Hybrid method can help resolve with additional analysis.

Referee Comment #1 provides an excellent list of potential sources of uncertainty that need to be addressed; I have nothing to add.

We thank the reviewer for emphasizing this critical issue regarding uncertainty propagation. Using the PAX-ISS hybrid framework, this study decomposes total aerosol absorption into contributions from BC, BrC, and the lensing effect and ultimately determines the absorption enhancement factor (E_{abs}) from multiple measured and derived quantities. Therefore, the evaluation should follow error-propagation principles and should be based on a clear identification of the uncertainty sources and characteristics of each relevant sub-variable.

In the manuscript, E_{abs} is expressed as follows:

$$E_{abs}(\lambda) = \frac{B_{abs,BC}(\lambda) + B_{abs\_lensing}(\lambda)}{B_{abs,BC}(\lambda)}$$

$$= \frac{B_{abs,BC}(\lambda)+(B_{abs\_coated}(\lambda)-B_{abs\_uncoated}(\lambda))}{B_{abs,BC}(\lambda)}$$

$$= \frac{B_{abs,BC}(\lambda)+(B_{abs\_coated}(870)\times\left(\frac{\lambda}{870}\right)^{-AAE_{coated}} - \frac{100\times ABS_{ISS}\times A}{CF\times F\times T})}{B_{abs,BC}(\lambda)}$$

where $B_{abs,BC}(\lambda)$ is the absorption coefficient of BC; $B_{abs,BrC}(\lambda)$ is the absorption coefficient of BrC; $B_{abs,lensing}(\lambda)$ is the absorption coefficient due to the lensing effect; $B_{abs,coated}(\lambda)$ is the total absorption coefficient of coated aerosols; $B_{abs,uncoated}(\lambda)$ is the lensing-free absorption coefficient; λ is the wavelength; AAE_{coated} is the absorption Ångström exponent of coated aerosols; CF is the correction factor; A is the deposited area on the filter; F is the sampling flow rate; T is the sampling duration, and ABS_{ISS} is the absorbance of the filter sample measured by ISS.

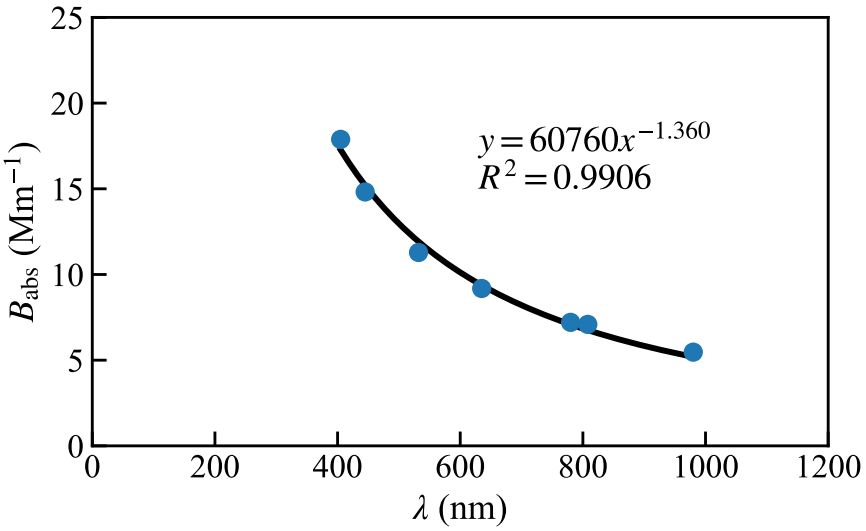
Thus, the uncertainty of $E_{abs}(\lambda)$ results from the propagation of uncertainties in the five sub-variables $B_{abs,BC}(\lambda)$, $B_{abs\_coated}(870)$, AAE_{coated}, CF, and ABS_{ISS}. We discuss the probability distributions of these five sub-variables in turn.

(1) $B_{abs\_coated}(870)$. The value of $B_{abs\_coated}(870)$ is obtained primarily from direct PAX measurements and may be influenced by instrument calibration, laser-intensity stability, flow-rate stability, cavity contamination, relative humidity, and acoustic response. According to Lack et al. (2006), the PAX technique has become an accepted reference for comparisons with filter-based and difference methods (Arnott et al., 1999, 2003, 2005a, b; Moosmüller et al., 1998; Schmid et al., 2006; Schnaiter et al., 2005; Sheridan et al., 2005). Reported accuracies are typically 5–10%, depending on the laboratory or field setting and the calibration method. The overall instrument accuracy with respect to aerosol absorption is estimated to be about 5%. Lack et al. (2009) later reconfirmed this estimate. Therefore, the uncertainty in B_{abs_coated} was taken as 5%.

(2) AAE_{coated}. Because the PAX instrument used in this study is a single-wavelength PAX-870, only the in situ absorption at 870 nm ($B_{abs\_coated}(870)$) was measured directly. We calculated AAE_{coated} from the power-law relationship between wavelength and the light-absorption signals obtained using the multi-wavelength thermal/optical carbon analyser (Chen et al., 2015; Chow et al., 2018, 2021; Shen et al., 2025). Based on $B_{abs\_coated}(870)$ and AAE_{coated}, we then calculated the absorption coefficient at any wavelength between 370 and 880 nm, namely $B_{abs\_coated}(\lambda)$.

We examined the absorption-coefficient fitting results for a representative day, as

shown in the figure below and in Figure S3 of the Supplement. Based on the differences between the measured absorption coefficients at the seven wavelengths and the corresponding values derived from the fitted curve, we estimated the uncertainty of *AAE* to be 3.6%.



Note: Absorption coefficients and the fitted curve measured by the DRI Model 2015 Multi-Wavelength Thermal/Optical Carbon Analyser on 19 February 2023.

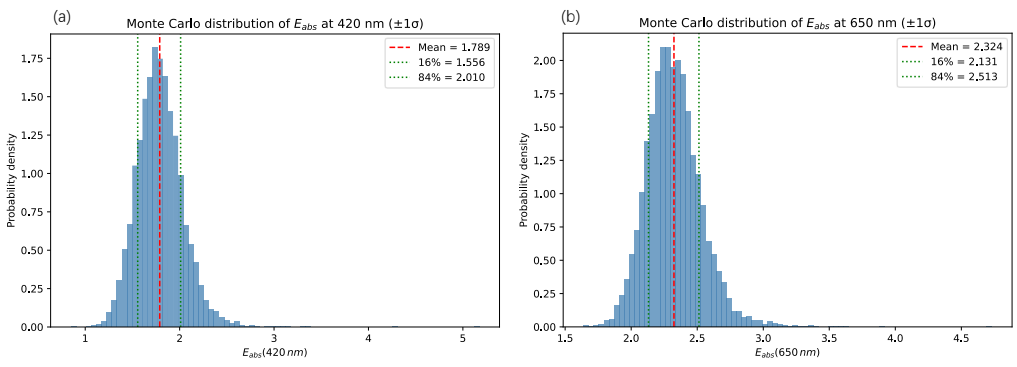
(3) ABS_{ISS}. ABS_{ISS} is the fundamental quantity used to calculate $B_{abs\_uncoated}(\lambda)$ (defined as $B_{abs\_uncoated}(\lambda) = \frac{100 \times ABS_{ISS} \times A}{CF \times F \times T}$). The uncertainty in ABSISS measurements arises from factors including the stability of the integrating-sphere light source, detector response, repeatability of sample punches, uniformity of filter deposition, solvent treatment time, blank-filter subtraction, and variations in sample positioning within the integrating sphere. According to Wonaschütz et al. (2009), the precision of ISS-measured absorbance is 4%.

(4) CF. As described in our manuscript, the punch placed in the cuvette inside the integrating sphere chamber can absorb light from multiple directions rather than from a single direction. This causes the absorbance measured by the ISS system to differ from that measured by a conventional method, in which the incident light travels along one direction. The conversion factor (CF), which converts ISS-measured absorbance to conventional one-way absorbance, was determined by comparing the absorbance values of Humic Acid Sodium Salt (HASS) obtained using ISS (ABS_{ISS}) and a conventional setup (ABS_o; e.g., a cuvette outside the integrating sphere). We refer to

our previous article (Li et al., 2023), where the determination of CF was described. The relative standard deviation (RSD) of the measured ABS_{ISS} was 5.67% (Li et al., 2023).

(5) $B_{abs,BC}(\lambda)$. The uncertainty in $B_{abs,BC}(\lambda)$ primarily stems from the decomposition of ABS_{ISS} value during the DWIC process, i.e., partitioning it into the contribution shares of BC and BrC. For evaluation, we used the data from Wonaschütz et al. (2009), who measured a series of reference samples containing known proportions of carbon black and humic acid sodium salt using the DWIC method and assessed the performance of DWIC in decomposing the absorbance contributions of BC and BrC. They plotted attenuation signals calculated from calibration curves against attenuation signals directly measured by ISS, as shown in Figure 3 of Wonaschütz et al. (2009), with one scatter plot for long-wavelength values and another for short-wavelength values. Using the open-source WebPlotDigitizer software, we extracted the x- and y-coordinates of each data point directly (see Table S3 in the Supplement). The results show that the average relative difference for $B_{abs,BC}(\lambda)$ was 10.4% at the low wavelength and 8.1% at the high wavelength.

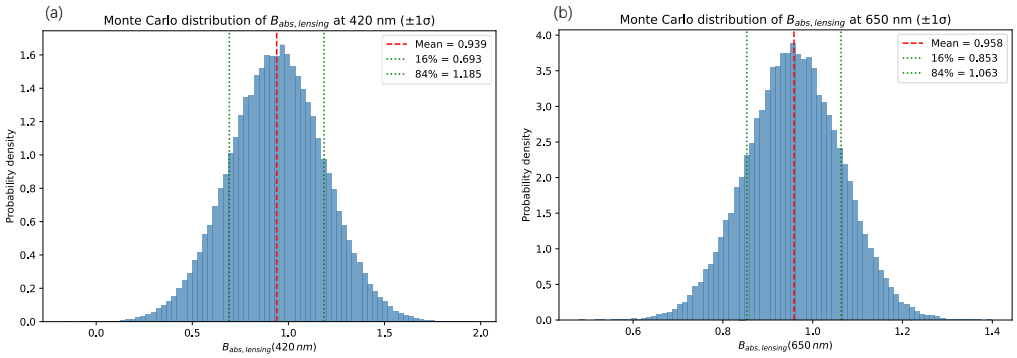
After determining the uncertainties of all five sub-variables, we propagated them to estimate the uncertainty of E_{abs}. We used Monte Carlo simulation to calculate the probability distribution of E_{abs} for the sample collected on 19 February 2023. As shown below and in Figure S4, the mean E_{abs} at 420 nm is 1.789 ± 0.221, corresponding to an uncertainty of 12.4%; at 650 nm, the mean E_{abs} is 2.324 ± 0.189, corresponding to an uncertainty of 8.1%.



Note: Probability distribution of E_{abs} at 420nm (a) and 650nm (b) on 19 February 2023. The

±1σ range (68% probability) is used to represent uncertainty.

For $B_{\mathrm{abs,lensing}}(\lambda)$ alone, the uncertainty results from the propagation of uncertainties in the four sub-variables ($B_{\mathrm{abs_{coated}}}(870)$, AAE_{coated}, CF, and ABS_{ISS}) used to calculate $B_{\mathrm{abs,lensing}}(\lambda)$. Because the uncertainties of these four sub-variables have been determined, we used Monte Carlo simulation to calculate the probability distribution of $B_{\mathrm{abs,lensing}}(\lambda)$ for the sample collected on 19 February 2023. As shown below and in Figure S5, the mean $B_{\mathrm{abs,\ lensing}}$ at 420 nm is 0.939 ± 0.246 Mm^{-1}, corresponding to an uncertainty of 26.2%; at 650 nm, the mean $B_{\mathrm{abs,\ lensing}}$ is 0.958 ± 0.105 Mm^{-1}, corresponding to an uncertainty of 11.0%.



Note: Probability distributions of $B_{\mathrm{abs,lensing}}$ at 420 nm (a) and 650 nm (b) on 19 February 2023. The ±1σ range (68% probability) is used to represent uncertainty.

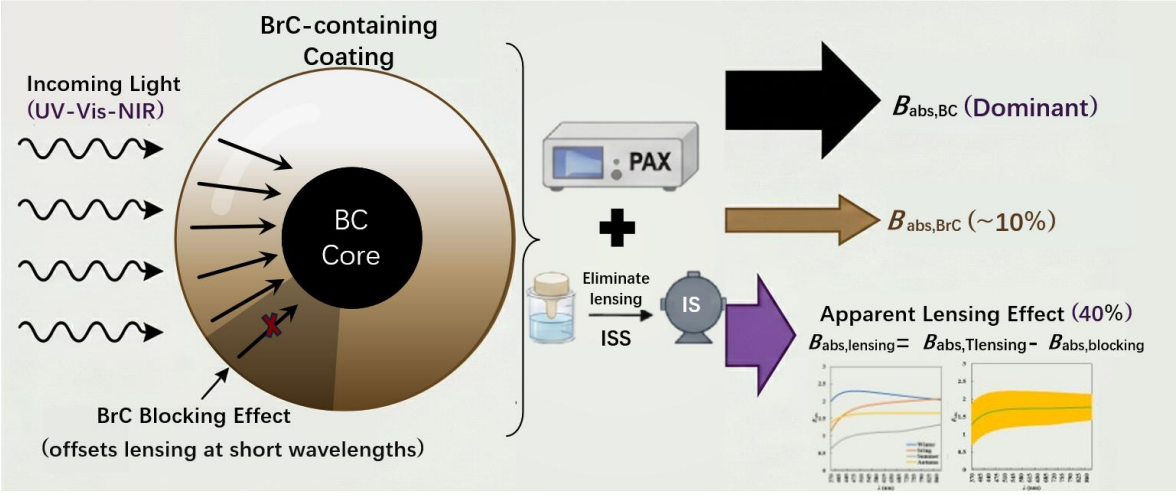
To comprehensively address the uncertainty inherent in the PAX-ISS methodology, we have added a new subsection, "3.4 Assessment of uncertainty", to the revised manuscript (lines 450–465). In addition, the Supplement now includes a section entitled "Assessment of uncertainty of PAX-ISS methodology", which details the methods used to determine the magnitude of uncertainty for each parameter or variable, as well as the probability distribution of the final E_{abs} values.

Minor comments:

Graphical abstract: "(offsets lensing at short __?_)"

We have revised the phrase to 'offsets lensing at short wavelengths' in the

graphical abstract.



Line 50: Instead of “stability”, maybe “vapor pressure”?

We agree that vapor pressure is a critical factor influencing the volatilization or removal of coatings during heating. However, we prefer to retain the term "stability," because our intended meaning is broader than vapor pressure alone. Specifically, whether coating materials can be removed during thermal denuding or thermal treatment depends not only on their vapor pressure but also on whether they undergo thermal decomposition. Therefore, we employ “stability” to characterize the overall differences in thermal and volatility-related stability between the BC core and coating materials, rather than focusing solely on vapor pressure as a single parameter.

Line 51: Swap “former” and “latter”

We thank the reviewer for pointing out this mistake; it has been corrected in the revised manuscript (lines 51–54).

Figure 1. Bottom left-most box should be – $B_{abs,lensing} = B_{abs\_coated} - B_{abs\_uncoated}$

This has been corrected.

We again thank the reviewers for their constructive and instructive comments, which have helped us improve the manuscript.

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