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Apportioning Light Absorption of Ambient Aerosols to Black Carbon, Brown Carbon, and Lensing Effect Using a PAX-ISS Hybrid Method: Insights into Absorption Enhancement

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Abstract

Accurately apportioning aerosol light absorption to black carbon (BC), brown carbon (BrC), and the lensing effect is crucial for constraining aerosol radiative forcing, yet existing methods often fail to resolve all three components simultaneously. Here, we introduce and demonstrate an integrated measurement framework that couples photoacoustic spectroscopy (PAX) with an integrating sphere system using solvent mediation (ISS). This PAX-ISS hybrid method quantifies the total absorption of ambient aerosols (B_{abs_coated}) and removes the lensing effect via solvent-mediated coating removal to obtain the lensing-free absorption ($B_{abs\_uncoated}$). The absorption solely due to the lensing effect ($B_{abs,lensing}$) is then directly quantified as their difference: $B_{abs,lensing}=B_{abs\_coated}-B_{abs\_uncoated}$. The lensing-free absorption ($B_{abs\_uncoated}$) is further spectrally decomposed into BC and BrC contributions ($B_{abs,BC}$ and $B_{abs,BrC}$) using a dual-wavelength iterative algorithm. Applied to seasonal samples in Beijing during 2023, the method revealed that BC dominated light absorption, with BrC contributing approximately 10% annually. The apparent lensing-induced enhancement averaged 40% of total absorption but exhibited strong seasonal (4.6–52.0%) and spectral variations, contracting sharply at shorter wavelengths—a pattern indicative of a BrC "blocking effect" that offsets lensing enhancement. Our field measurements provide, for the first time, direct observational evidence supporting this blocking effect, which was initially proposed by other researchers based solely on numerical simulations. The annual wavelength-averaged absorption enhancement factor (E_{abs}) was 1.69 ± 0.10. This methodology provides a robust, observationally constrained approach to apportion aerosol absorption, offering refined insights for climate modeling.

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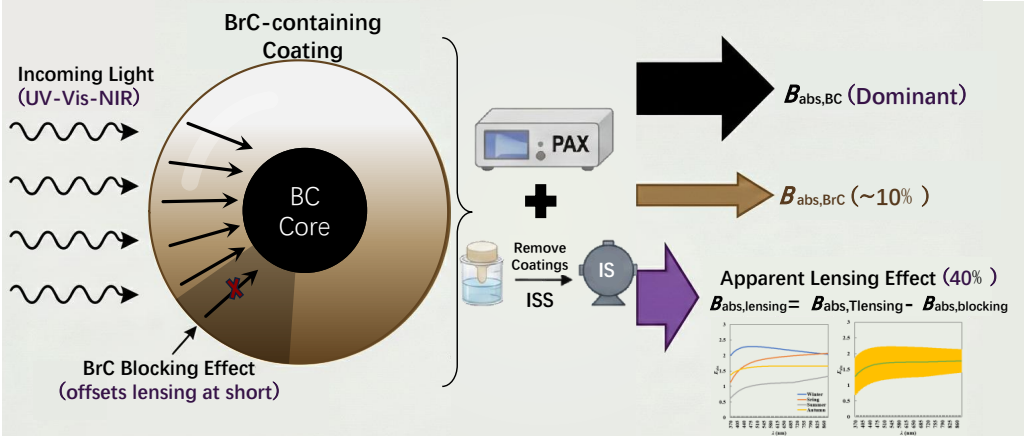
Graphical Abstract



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1 Introduction

Black carbon (BC) and brown carbon (BrC) are critical components of atmospheric particulate matter due to their strong absorption of solar radiation (Laskin et al., 2025; Xie et al., 2025; Yang et al., 2025). BC is a potent, broad-spectrum absorber, while BrC exhibits strongly wavelength-dependent absorption, primarily in the near-ultraviolet to visible range (Bond et al., 2013; Laskin et al., 2015). Accurately quantifying their respective contributions to total light absorption is fundamental for assessing aerosol radiative forcing, understanding atmospheric heating rates, and constraining climate models (Li et al., 2023a; Yang et al., 2025; Zhu et al., 2021). A key complexity, however, arises from the atmospheric ageing of BC. This process frequently results in the internal mixing of BC with non-absorbing or weakly absorbing coatings and enhances BC's absorption because of the lensing effect (Cappa et al., 2012; Jin et al., 2025; Yus-Díez et al., 2022). Consequently, the total measured absorption coefficient of ambient aerosols (B_{abs_coated}) is an aggregate signal from three distinct physical processes: absorption by bare BC cores, absorption by BrC, and the lensing-driven enhancement of BC absorption due to coatings. Thus, disentangling these three contributors—referred to as apportionment of total aerosol light absorption—is essential for advancing our understanding of aerosol mixing state, ageing processes, and ultimately, climate impacts (Liu et al., 2015; Pokhrel et al., 2017).

However, most methodological frameworks attempting to decompose aerosol total absorption could only resolve one or two of these contributors, but not all three. For example, a common way to obtain the information of one contributor is by accounting for the contributions of the other species based on their own known physicochemical properties. These properties are usually solubility in water or organic reagents or thermal stability at a certain temperature (Cappa et al., 2019; Liu et al., 2015; Xie et al., 2019). For the former, BC light absorption can be obtained as organic species are vaporized (Cappa et al., 2019; Liu et al., 2015; Xie et al., 2019) and for the latter, the optical properties of solutions can be measured to estimate the light absorption of BrC

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(Choudhary et al., 2017; Wu et al., 2019; Yue et al., 2019). Being inherently designed to isolate a single component, these approaches typically focus on either BC or BrC alone. Another approach for separating the contributions of BC and BrC from the total measured aerosol light absorption is the absorption Ångström exponent (AAE) attribution method (Lack and Langridge, 2013; Wang et al., 2019a, b). This approach assumes that only BC absorbs solar radiation at near-infrared wavelengths (e.g., 880 nm) and the BC's AAE (AAE_{BC}) is 1.0 or other preset values. The light absorptions of BC at other wavelengths ($B_{abs,BC}(\lambda)$) are extrapolated with the help of AAE_{BC}, enabling the light absorption of BrC ($B_{abs,BrC}(\lambda)$) to be calculated by subtracting the $B_{abs,BC}(\lambda)$ from the measured total absorption ($B_{abs\_coated}(\lambda)$). Considering that the AAE_{BC} value is actually variable (Liu et al., 2018b; Wang et al., 2021b), this approach is not perfect yet.

Thus far, methodological frameworks capable of resolving all three contributors include the applications of pure numerical calculation or the combination of a thermal denuder with a photoacoustic soot spectrometer (PASS-3) in field measurements. The former category is usually embedded with too many assumptions (Luo et al., 2018, 2021; Moschos et al., 2021; Zhang et al., 2021) and needs further improvement based on field verifications. The latter includes the combined use of photoacoustic spectroscopy (PAX) and thermal denuding (TD), which allows for the calculation of an absorption enhancement factor (E_{abs}) and, with additional spectral constraints, the separation of BrC and lensing contributions from BC (Cappa et al., 2012; Liu et al., 2015; Pokhrel et al., 2017). Even if the TD process may be subject to an incomplete separation of BrC from BC (Liu et al., 2015) and is therefore not conducive to an accurate apportionment, it is still appreciable in distinguishing the absorption contributors and calculating absorption enhancement factors.

In this study, we introduce and demonstrate an integrated methodological framework designed to apportion total carbonaceous aerosol light absorption into its three constituent parts. Two established but typically separate techniques: PAX and integrating sphere with solvent extraction (ISS) are synergistically coupled to provide

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a self-consistent framework to apportion absorption to BC, BrC, and the lensing effect. We present this methodology, detail its implementation, and demonstrate its applications such as in the understanding of absorption enhancement due to absorptive or non-absorptive coatings. We anticipate that this methodology will provide robust constraints for aerosol optical properties and climate modelling.

2 Methodology

2.1 Methodological architecture

As mentioned in the Introduction, the methodology adopted here is based on coupling two mature techniques: PAX and ISS. The PAX provides an accurate, in situ measurement of the total ambient absorption coefficient at 870 nm ($B_{abs\_coated}(870)$) (Lack et al., 2012; Selimovic et al., 2019). This value is subsequently extrapolated to derive absorption coefficients across wavelengths from 380 nm to 880 nm ($B_{abs\_coated}(\lambda)$). Concurrently, the ISS system measures the absorption of the same aerosol population after the lensing effect has been eliminated by solvent-mediated coating removal and refractive index matching, yielding $B_{abs\_uncoated}(\lambda)$. The reason why the mixed solvent eliminate lensing has been detailed in a few publications (Li et al., 2023b; Sun et al., 2021; Wonaschütz et al., 2009). The difference [$B_{abs\_coated}(\lambda) - B_{abs\_uncoated}(\lambda)$] directly quantifies the absorption enhancement due solely to the lensing effect ($B_{abs,lensing}(\lambda)$). Furthermore, the ISS system, employing a Double-Wavelength Iterative Calculation (DWIC) algorithm with carbon black and humic acid as reference materials, spectrally decomposes $B_{abs\_uncoated}(\lambda)$ into the contributions of bare BC ($B_{abs,BC}$) and BrC ($B_{abs,BrC}$) (Sun et al., 2021; Wonaschütz et al., 2009). A schematic of the methodological architecture is shown in Figure 1.

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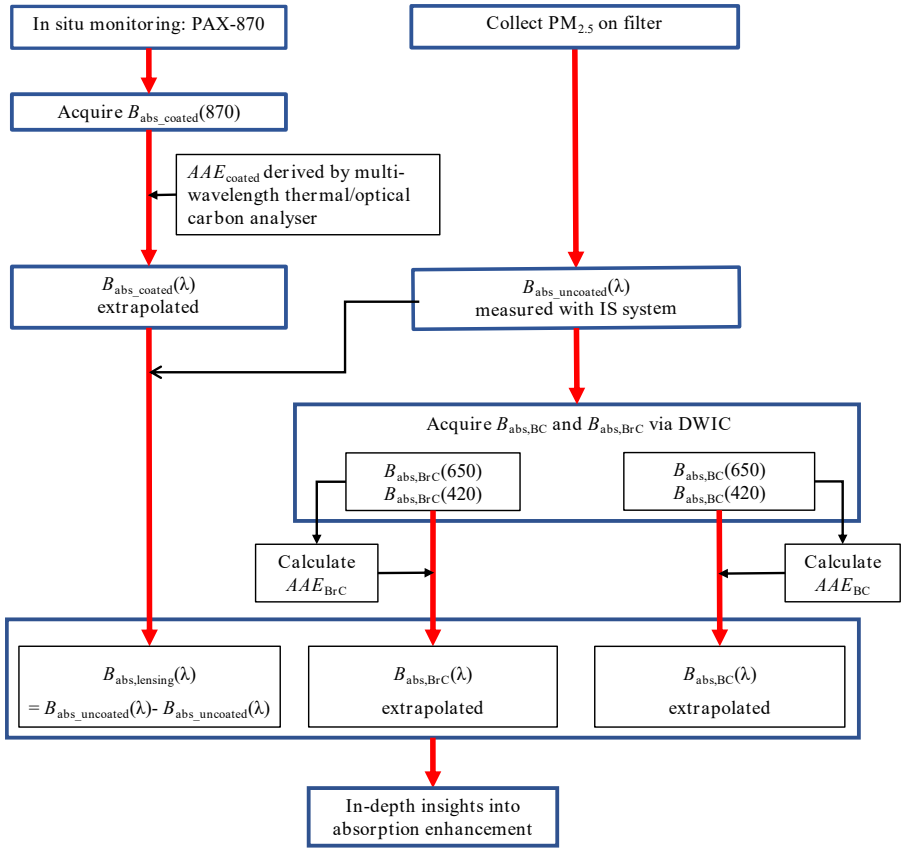

Figure 1. Methodological architecture. Note: The λ ranges from 370 nm to 880 nm

2.2 Measurement of lensing-free absorption using ISS system

2.2.1 Sample collection

Ambient $PM_{2.5}$ (particulate matter with an aerodynamic diameter ≤ 2.5 μm) was sampled at the Chinese Research Academy of Environmental Sciences (CRAES), Beijing (40.04°N, 116.42°E). A high-volume air sampler (Tisch Environmental Inc., OH, USA) was used at an air flow rate of 1.13 m^3/min. The sampling site was situated atop a smog chamber facility (~10 m above ground), with an 80-meter radius free of anthropogenic emission sources nearby (Li et al., 2023b; Wang et al., 2021a; Zhang et al., 2020). Quartz fibre filters (Pallflex™ Tissuquartz 2500QAT-UP, 8″ × 10″, Pall Corporation) were deployed for 23.5-hour repeated sampling cycles, commencing at 09:00 Beijing Time (UTC+8) every day and concluding at 08:30 the following morning.

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Seasonal campaigns in 2023 spanned the winter period of 9–24 February, the spring period of 7 April–9 May, the summer period of 7 July–6 August, and the autumn period of 12 October–5 November. The selected four time periods are representative of the meteorological conditions and emission regimes of four quarters of the year (Li et al., 2023b; Wang et al., 2021a; Zhang et al., 2020). The quality assurance measures for sampling filters and eligible days (Table S1) are detailed in Supporting Information (Sect. S1).

2.2.2 Measurement of absorption for uncoated aerosol

As previously described, the ISS system eliminates lensing-induced absorption ($B_{\mathrm{abs,lensing}}(\lambda)$) while preserving bare BC and BrC absorption components (i.e., $B_{\mathrm{abs\_uncoated}}(\lambda) = B_{\mathrm{abs,BC}}(\lambda) + B_{\mathrm{abs,BrC}}(\lambda)$) (Sun et al., 2017; Wonaschütz et al., 2009; Li et al., 2023). The system comprises a light source, a custom-made 150 mm IS, and a UV–Vis-NIR spectrometer (Li et al., 2023b; Sun et al., 2017, 2021; Wang et al., 2021a). For each measurement, a transparent quartz cuvette (1 cm × 1 cm × 4 cm), holding 3 mL of a solvent mixture (acetone, water, and 2-propanol in a ratio of 5:4:1) was fixed in the center of the integrating sphere chamber. A filter punch (30×8 mm) was immersed in the solvent mixture within the cuvette for optical detection. The wavelength-resolved absorbance ($ABS_{\mathrm{ISS}}(\lambda)$) was acquired from 370 nm to 880 nm, defined as:

$$ABS_{ISS}(\lambda) = -ln\frac{I(\lambda)}{I_0(\lambda)}, \quad (1)$$

where $I(\lambda)$ and $I_0(\lambda)$ denote the transmitted light intensities through the loaded and blank filters, respectively. The absorption coefficient of uncoated aerosols ($B_{\mathrm{abs\_uncoated}}(\lambda)$) is then converted as:

$$B_{\mathrm{abs\_uncoated}}(\lambda) = \frac{100\times ABS_{\mathrm{ISS}}\times A}{CF\times F\times T}, \quad (2)$$

where A is the total deposited area on the filter used by the high-volume sampler described in Sect. 2.2.1 (cm^2); F is the flow rate of filter sampling (m^3/min); T is the duration of each filter sampling (min); and CF is the correction factor converting the ISS-measured absorption (multiple-angle absorption within the integrating sphere

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chamber) to the conventional one-way absorption (as measured by PAX). The method for determining the *CF* value has been described in our previous publication (Li et al., 2023b) and is represented in Supporting Information (Sect. S2).

2.3 Measurement of total absorption via PAX

2.3.1 Thermal/optical analysis

A DRI Model 2015 Multi-Wavelength Thermal-Optical Carbon Analyser was used to split aerosol total carbon (TC) into organic carbon (OC) and elemental carbon (EC) employing temperature-regulated pyrolysis under controlled atmospheres. A 0.5024 cm^2 punch from a quartz filter was heated stepwise to 140 °C (OC1), 280 °C (OC2), 480 °C (OC3), and 580 °C (OC4) in a pure helium atmosphere and then combusted stepwise at 580 °C (EC1), 740 °C (EC2), and 840 °C (EC3) in a 2% oxygen + 98% helium atmosphere following the IMPROVE_A temperature protocol. The mass values of OC and EC were calculated as OC = OC1 + OC2 + OC3 + OC4 + OPC and EC = EC1 + EC2 + EC3 - OPC, where OPC represents pyrolyzed OC (Chow et al., 2007).

To acquire *AAE*s, filter transmittance and reflectance were continuously monitored using 7-wavelength diode lasers (405, 445, 532, 635, 780, 808, 980 nm), and the wavelength-specific light attenuation (*ATN*) was calculated accordingly. To mitigate filter matrix artefacts, including loading effects and multiple scattering (Chen et al., 2015; Chow et al., 2018; Peng et al., 2020), wavelength- and EC loading-dependent adjustment factors were applied to the raw *ATN* to obtain corrected *ATN*s (see Supporting Information Sect. S3, Table S2). Based on these, the *AAE*s for coated aerosols (AAE_{coated}) were calculated. The AAE_{coated} was then used to extrapolate $B_{abs\_coated}(\lambda)$ from $B_{abs\_coated}(870)$ in the following section.

2.3.2 Acquisition of total absorption at broad wavelength range

A photoacoustic extinction spectrometer (PAX-870, Droplet Measurement Technologies) was deployed to monitor the in situ atmospheric absorption coefficient at 870 nm ($B_{abs\_coated}(870)$) in real-time. Considering that we have only a PAX set with a single wavelength (870 nm), the absorption coefficients at other wavelengths

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($B_{abs\_coated}(\lambda)$) had to be extrapolated using the AAE_{coated} obtained in Sect. 2.3.1:

$$B_{abs\_coated}(\lambda) = B_{abs\_coated}(870) \times \left(\frac{\lambda}{870}\right)^{-AAE_{coated}} \quad (3)$$

2.4 Decomposition of coated aerosol absorption by coupling ISS-PAX

2.4.1 Lensing-equivalent absorption

The availability of $B_{abs\_coated}(\lambda)$ from PAX and $B_{abs\_uncoated}(\lambda)$ from ISS allows the acquisition of $B_{abs,lensing}(\lambda)$. As shown in Figure 1, $B_{abs,lensing}(\lambda)$ is obtained by calculating the difference between $B_{abs\_coated}(\lambda)$ and $B_{abs\_uncoated}(\lambda)$, as:

$$B_{abs,lensing}(\lambda) = B_{abs\_coated}(\lambda) - B_{abs\_uncoated}(\lambda) \quad (4)$$

2.4.2 BC and BrC absorptions

To decompose $B_{abs\_uncoated}(\lambda)$, which is lensing free, into the individual contributions of BC and BrC, the DWIC algorithm was employed. Details of the DWIC algorithm have been described in our previous publications (Sun et al., 2017, 2021; Wang et al., 2021a) and in the Supporting Information (Sect. S4 and Figure S1). Briefly, the iterative calculation uses two wavelengths (650 and 420 nm) and two reference materials: Carbon Black (CarB) as the proxy for BC, and Humic Acid Sodium Salt (HASS) as the proxy for BrC (Reisinger et al., 2008; Sun et al., 2017; Wang et al., 2021a, a; Wonaschütz et al., 2009). Calibration curves for CarB and HASS masses were established based on their respective absorption signals measured by the ISS at 650 (BC-dominated) and 420 nm (BrC-sensitive). Iterative optimization yielded the absorption coefficients of BC and BrC at these two wavelengths, i.e., $B_{abs,BC}(650)$, $B_{abs,BC}(420)$, $B_{abs,BrC}(650)$, and $B_{abs,BrC}(420)$.

The absorption coefficients of BC and BrC at wavelengths other than 420 nm and 650 nm (i.e., $B_{abs,BC}(\lambda)$, $B_{abs,BrC}(\lambda)$) can be extrapolated from the values at these two wavelengths, constrained by their respective *AAE*s (AAE_{BC} and AAE_{BrC}). The *AAE* for BC or BrC is calculated as:

$$AAE_X = -\frac{\ln\left(\frac{B_{abs,x}(650)}{B_{abs,x}(420)}\right)}{\ln\frac{650}{420}} \quad (5)$$

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The absorption coefficient at any wavelength is then given by:

$$B_{\mathrm{abs,x}}(\lambda) = B_{\mathrm{abs\_uncoated}}(420) \times \left(\frac{\lambda}{420}\right)^{-AAE_{\mathrm{X}}} \quad (6)$$

where X refers to BC or BrC. The wavelength 420 nm in Eq. (6) can be replaced by 650 nm.

2.5 Calculation of absorption enhancement factors

With the total aerosol absorption apportioned to BC, BrC, and lensing effect, the absorption enhancement factor of BC ($E_{\mathrm{abs}}(\lambda)$) is available via:

$$E_{\mathrm{abs}}(\lambda) = \frac{B_{\mathrm{abs,BC}}(\lambda) + B_{\mathrm{abs,lensing}}(\lambda)}{B_{\mathrm{abs,BC}}(\lambda)} \quad (7)$$

3 Results and discussion

3.1 Concentrations and total absorptions of ambient aerosol

3.1.1 Concentrations

Every filter-collected sample was weighed to determine $PM_{2.5}$ concentration and analysed for aerosol carbon speciation (Figure 2). Representative seasonal heterogeneity was observed in aerosol loadings and compositions.

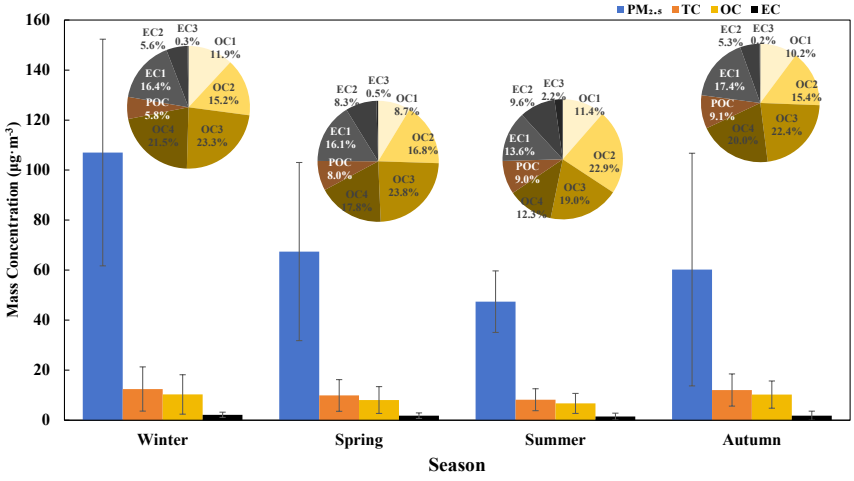

Figure 2. Aerosol loadings and compositions in 2023.

As shown in Figure 2, $PM_{2.5}$ mass concentrations demonstrate pronounced seasonal variations, peaking in winter (107.03 ± 45.31 μg·m^{-3}) and reaching a minimum in summer (47.40 ± 12.30 μg·m^{-3}), consistent with regional emission patterns and

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meteorological conditions (Chen et al., 2017; Wu et al., 2016b). The seasonal trends in TC, OC, and EC concentrations generally mirrored those of $PM_{2.5}$, although the relative order of spring and autumn was occasionally reversed. For example, unlike $PM_{2.5}$ concentrations of spring > autumn, EC concentrations are of autumn > spring, implying differences in $PM_{2.5}$ composition arising from varied sources, transport, and ageing processes (Arhami et al., 2018; Ravindra et al., 2022; Song et al., 2022).

Thermal-optical carbon analysis provided distinct source signatures, as indicated by the char-EC/soot-EC ratios. Elevated ratios during high-combustion seasons like winter (1.81 ± 1.61) and autumn (1.50 ± 2.67) reflect predominant contributions from coal and biomass burning, as established by source apportionment studies (e.g., Han et al., 2010). In contrast, reduced ratios during spring (0.93 ± 0.77) and summer (0.39 ± 0.58) accord with vehicular-dominated emissions (Han et al., 2008; Liu et al., 2018a). Further speciation of OC to OC1, OC2, OC3, and OC4 suggests the influence of atmospheric processing. Low-volatility OC3–OC4 fractions constitute 40–54% of total OC across all seasons, indicating the pathways of photochemical ageing and aqueous-phase processing that generate oxygenated semi-volatile compounds (Aswini et al., 2019; Li et al., 2018).

3.1.2 Total absorption

Having established the mass concentration and compositional context, we now examine the optical absorption properties of the coated aerosols. The daily light absorption of coated aerosol calculated in Sect. 2.3.2 was averaged seasonally and annually. Figure 3 shows the spectral dependence of total light absorption ($B_{abs\_coated}(\lambda)$) across seasons and the annual average. Both the absorption intensity and the absorption Ångström exponent (AAE_{coated}) exhibit a consistent seasonal order: winter > autumn > spring > summer. This order aligns with the seasonal trend in TC concentrations shown in Figure 2. Since both BC and BrC are spectrally dependent components of TC, it is consistent that the AAE_{coated} values follow the same order as TC concentrations.

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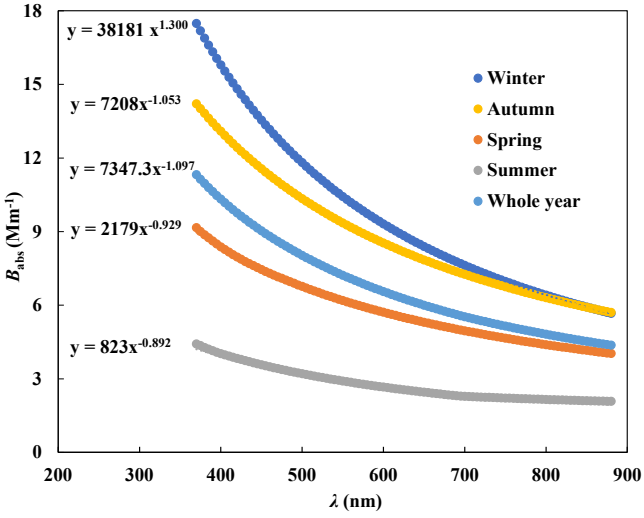

Figure 3. Total absorption of coated aerosol.

The AAE_{coated} values were highest in winter (1.300) and lowest in summer (0.892), with an annual mean of 1.053. *AAE*s lower than 1.0 are observed in spring and summer, seemingly challenging the traditional view that the *AAE* of carbonaceous aerosol should exceed 1.0, given that BC's *AAE* is typically 1.0 and BrC's *AAE* >1.0 (Lack and Langridge, 2013). 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. While referencing this broad range is instructive, the consistently low AAE_{coated} values in our spring and summer data may also be partially influenced by the net spectral effect of lensing enhancement and BrC blocking, which our subsequent deconvolution analyses aim to clarify.

To sum up this section (Sect. 3.1), the distinct seasonal characteristics of aerosol concentrations, carbon ratios, and *AAE*s reflect the diversity of the local ambient aerosol population, providing a suitable context for a comprehensive study of overall and decomposed light absorptions using the PAX-ISS hybrid method.

3.2 Deconvolved light absorption

Total absorption ($B_{abs\_coated}(\lambda)$) was deconvolved into $B_{abs,BC}(\lambda)$, $B_{abs,BrC}(\lambda)$, and

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$B_{abs,lensing}(\lambda)$ on daily, seasonal, and annual bases. Figure 4 shows the seasonal and annual decomposed absorption, where panels a−d display the wavelength-resolved contributions for winter, spring, summer, and autumn, respectively; panel e shows the annual wavelength-resolved contribution; and panel f shows the wavelength-averaged contributions for each season and the whole year. As noted, the wavelength range considered spans 370−880 nm, representing a most energetic part of solar radiation (Thuillier et al., 2003). To enable direct comparison across time periods, all panels showing wavelength-resolved absorptions (a−e) share a common y-axis upper limit of 18 Mm^{-1}.

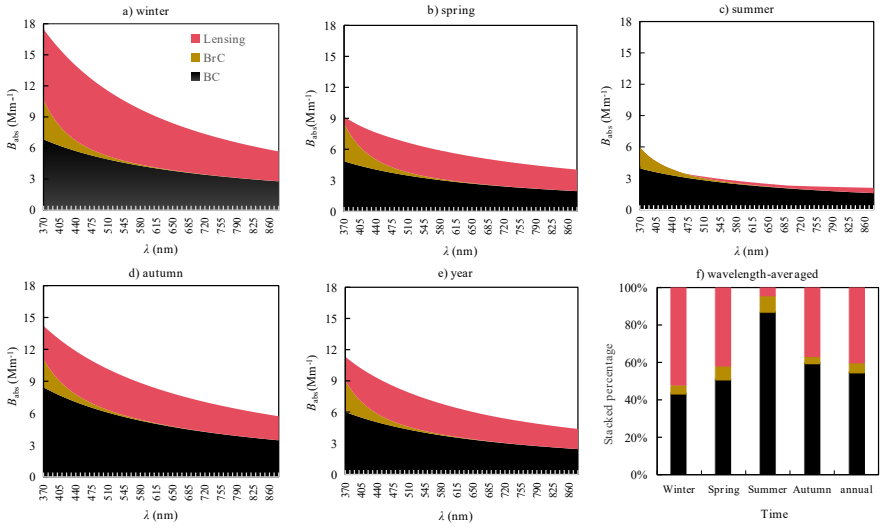

Figure 4. Deconvolved aerosol light absorption in wavelength-resolved (a−e) and wavelength-averaged (f) forms.

Both BC and BrC exhibited clear spectral dependence seasonally and annually (panels a−e), with BrC's dependence being more pronounced than BC's, consistent with the established knowledge that AAE_{BrC}>AAE_{BC} (Kaskaoutis et al., 2021; Lack and Langridge, 2013; Wu et al., 2016a). In all seasons, BC absorption exceeded BrC absorption even at ultraviolet wavelengths, indicating BC's dominant role in light absorption for the urban aerosol studied. This is further corroborated by Figure 4f, where the wavelength-averaged BrC absorption value was approximately 10% of BC absorption value for both individual seasons and the annual average. Regarding the

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apparent lensing-equivalent absorption ($B_{abs,lensing}$), Figure 4f shows an annual mean contribution of 40.0% to the total light absorption (B_{abs_coated}), with seasonal means of 52.0% in winter, 41.8% in spring, 4.6% in summer, and 36.7% in autumn. These differing seasonal contributions of $B_{abs,lensing}$ suggest varying degrees of BC absorption enhancement, discussed in Sect. 3.3.

More significantly, and central to one of our key findings, Figure 4a−e reveals a pronounced contraction of the apparent lensing effect toward shorter wavelengths. For example, at the annual level (Figure 4e), the lensing-equivalent absorption at 370 nm was only 66.3% of its maximum value at 455 nm. In spring (Figure 4b), lensing-equivalent absorption at 370 nm shrank to 22.9% of the maximum at 505 nm. The most extreme case was observed in summer (Figure 4c), where the lensing-equivalent absorption disappears at wavelengths below 465 nm. In this scenario, the summed absorption of BC and BrC at specific wavelengths (e.g., below 465 nm) exceeded the total measured absorption (summed absorption of BC, BrC, and lensing), suggesting that the intrinsic total lensing effect ($B_{abs,Tlensing}$) was overbalanced by other factors to the extent that the apparent lensing effect ($B_{abs,lensing}$) became negligible or negative. Although an accurate and reliable explanation for this spectral contraction phenomenon remains unavailable, the multiple effects of BrC internally mixed with BC cores are attracting increasing attention. Generally, coatings on BC can enhance its absorption via the lensing effect. When coatings are themselves absorptive (e.g., BrC), this enhancement constitutes one of the largest uncertainties in estimating aerosol radiative forcing (e.g., Curci et al., 2019; Wang et al., 2018). It has been established that the absorption enhancement of BC due to absorptive coatings (BrC) is lower than that induced by non-absorbing coatings (Lack and Cappa, 2010).

Further studies propose classifying BrC's impacts on BC absorption into a BrC-induced lensing effect and a BrC blocking effect (Luo et al., 2018; Zhang et al., 2021). In our framework, the lensing effect caused by BrC combines with that from non-absorbing coatings to constitute the intrinsic total lensing-equivalent absorption

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($B_{abs,Tlensing}$). The blocking effect ($B_{abs,blocking}$) occurs when BrC coatings block some photons that would otherwise be focused onto the BC core by the lensing effect, akin to a shadow. Therefore, total aerosol absorption should be defined as:

$$B_{abs\_coated} = B_{abs,BC}(\lambda) + B_{abs,BrC}(\lambda) + \left(B_{abs,Tlensing}(\lambda) - B_{abs,blocking}(\lambda)\right) \quad (8)$$

Consequently, the apparent lensing effect ($B_{abs,lensing}$) we measure represents the net balance between total lensing enhancement ($B_{abs,Tlensing}(\lambda)$) and BrC blocking ($B_{abs,blocking}$). Accordingly, $B_{abs,lensing}(\lambda)$ becomes negative whenever $B_{abs,blocking}(\lambda)$ exceeds $B_{abs,Tlensing}(\lambda)$. Thus, the observed contraction of $B_{abs,lensing}$ in Figure 4, which is precisely ($B_{abs,Tlensing} - B_{abs,blocking}$), is not surprising. The disappearance of $B_{abs,lensing}(\lambda)$ in summer below 465 nm (Figure 4c) likely does not indicate the absence of intrinsic total lensing but rather suggests that $B_{abs,Tlensing}(\lambda) - B_{abs,blocking}(\lambda) \leq 0$. The plausibility of such a BrC-induced blocking effect is supported by prior numerical investigations. For instance, Luo et al. (2018), employing the multiple-sphere T-matrix (MSTM) method, demonstrated that the total absorption of a BC core with brown coatings can be less than the sum of separately calculated absorptions ($B_{abs,BC} + B_{abs,BrC}$), attributable to blocking. Similarly, Zhang et al. (2021) numerically found that the lensing effect may not increase further with shell/core ratio beyond a threshold due to blocking. Furthermore, the wavelength dependence of the apparent lensing in Figure 4 (apparent lensing decreases toward shorter wavelengths) is largely complementary to that of BrC absorption (BrC absorption increases toward shorter wavelengths), implying an interplay with BrC blocking effects. Further investigations are needed to clarify the conditions for blocking effects, their interaction with the lensing from non-absorptive coatings, and their relationship with the direct light absorption from BrC.

3.3 Insight into absorption enhancement

The absorption enhancement of BC due to lensing is considered one of the largest uncertainties in assessing the climate impact of aerosol light absorption (Fan et al., 2024; Kong et al., 2024; Zhao et al., 2021). Here, we examine absorption enhancement from both wavelength-resolved and wavelength-averaged perspectives, followed by a

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discussion of why some enhancement quantification methods require correction.

3.3.1 Wavelength-resolved enhancement: insight into blocking effects

Using the daily wavelength-resolved absorption coefficients of BC, BrC, and apparent lensing effect (i.e., $B_{abs,BC}(\lambda)$, $B_{abs,BrC}(\lambda)$, and $B_{abs,lensing}(\lambda)$) obtained via Eqs. (4) and (6), the daily, seasonal, and annual wavelength-resolved enhancement factors ($E_{abs}(\lambda)$) were calculated using Eq. (7).

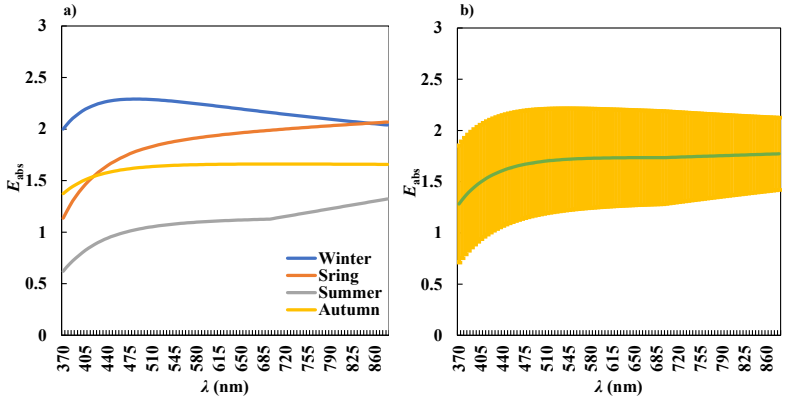

Figure 5. for seasonal (a) and yearly (b) terms.

Figure 5 shows the wavelength-resolved enhancement factors for the seasons and the year. A clear wavelength dependence is evident. Specifically, $E_{abs}(\lambda)$ decreased monotonically toward shorter wavelengths in spring, summer, and autumn, while in winter it increased initially before declining in the short-wavelength range. That is, all seasonal and annual $E_{abs}(\lambda)$ values turned downward as wavelengths approached the violet range. The observed decline in $E_{abs}(\lambda)$ is mechanistically explained by the definition of E_{abs} ($E_{abs} = 1 + B_{abs,lensing}/B_{abs,BC}$) given the concurrent decrease in $B_{abs,lensing}$ (Figure 5) and the increase in $B_{abs,BC}$ at shorter wavelengths (Figure 4). This decline in E_{abs} at short wavelengths is thus fundamentally driven by the strengthened blocking effect of BrC coatings, which reduces the net apparent lensing ($B_{abs,lensing} = B_{abs,Tlensing} - B_{abs,blocking}$). The most extreme case occurred in summer at wavelengths below 465 nm, where observed $E_{abs}(\lambda)$ values fell below 1.0, indicating that the net apparent lensing contributed negatively to BC light absorption. This echoes the finding of Luo et al.

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(2018) that $E_{abs}(\lambda)$ can be below 1 in the ultraviolet region for BC with brown coatings due to blocking, which they termed the "sunglasses effect". While the blocking effect of BrC has previously been examined only through numerical simulations (e.g., Luo et al., 2018; Zhang et al., 2021), our study provides the first field-based evidence to demonstrate this phenomenon.

3.3.2 Wavelength-averaged enhancement: insight into pollution-enhancement linkage

Theoretical and experimental studies of lensing-induced absorption enhancement have yielded diverse results. Mie theory for spherical core-shell particles yields enhancement factors up to 3 (Bond et al., 2006; Jacobson, 2001; Schwarz et al., 2008). Such enhancement is supported by laboratory studies (Cappa et al., 2012; Schnaiter, 2005; Shiraiwa et al., 2010; Zhang et al., 2008) and is incorporated into climate models that work out total BC absorption (lensing included) either through simplified mixing-state assumptions (Chung, 2005; Penner et al., 1998) or by fixing the enhancement value at ~1.5 (Flanner et al., 2007; Wang et al., 2014). In contrast, some field measurements have reported negligible absorption enhancement for ambient BC particles (Cappa et al., 2012, 2019; Fierce et al., 2016; Mbengue et al., 2021).

Complementing the wavelength-resolved results, we present the wavelength-averaged enhancement factor (E_{abs}, averaged over 370-880 nm) to assess the overall status of lensing. As shown in Figure 6, the annual average E_{abs} was 1.69 ± 0.10. Seasonal averages varied from 1.09 ± 0.15 (summer) to 2.18 ± 0.08 (winter), with spring and autumn values of 1.81 ± 0.21 and 1.63 ± 0.06, respectively. Given the decisive role of lensing, the considerable seasonal divergence in E_{abs} is attributable to the varying contributions of apparent lensing ($B_{abs,lensing}$) to total absorption (B_{abs_coated}) (e.g., 4.6% in summer versus 52.0% in winter; see Figure 4f). Other researchers have also reported diverse absorption enhancement factors in China, depending on specific conditions influencing lensing, particularly aerosol ageing statuses (Luo et al., 2018). For example, Cui et al. (2016) reported factors of 1.4 ± 0.3 for fresh combustion and ~3 for aged BC

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at a rural North China Plain site. Chen et al. (2017) reported an average of 2.07 ± 0.72 for winter haze in northern China, with diurnal variation from 1.31±0.29 (morning) to 2.23±1.05 (afternoon) and 1.52±0.75 (evening). Xu et al. (2016) even reported E_{abs} values of 2.6–4.0 for Beijing.

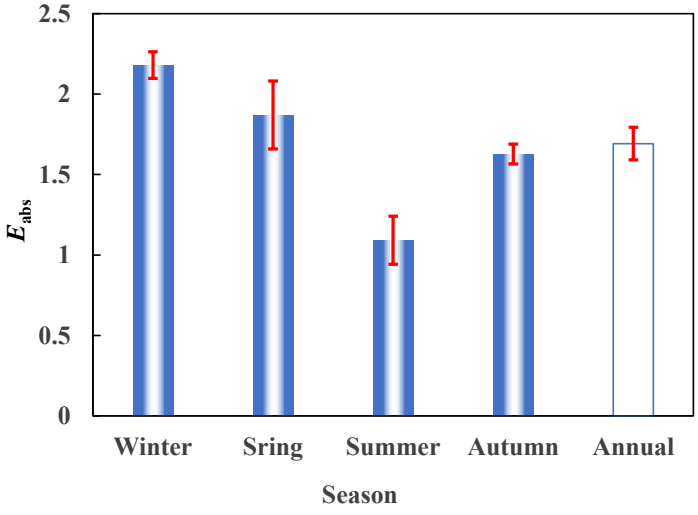

Figure 6. Wavelength-averaged absorption enhancement factors for seasons and the year.

Beyond the comparison with literature values, a notable pattern emerges within our dataset: the seasonal E_{abs} values (Figure 6) vary synchronously with $PM_{2.5}$ concentrations (Figure 2); that is, higher $PM_{2.5}$ levels corresponded to higher absorption enhancement factors. This aligns with our previous findings that increased air pollution leads to greater absorption enhancement (Li et al., 2023b; Zhang et al., 2017, 2020; Zhi et al., 2014). Increased air pollution in northern China usually results from stagnant weather that favors aerosol ageing, leading to the accumulation of primary emissions and the formation of secondary aerosols (Liu et al., 2024; Peng et al., 2021). The increased load of inorganic and organic pollutants enhances the likelihood and thickness of coatings on BC, thereby boosting the lensing effect (Li et al., 2023b).

This positive correlation is supported by the enhanced abundance of low-volatility, oxidised organic components (OC3-OC4, see Figure 2) during the high-$PM_{2.5}$ seasons (winter and autumn). These components are markers of atmospheric ageing and

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secondary formation (Boonpeng et al., 2026; Chow et al., 2007; Tohidi et al., 2022), processes conducive to the development of thicker coatings on BC, thereby promoting a stronger lensing effect.

With sustained improvements in local air quality in China, conflicting climate influences may emerge: the warming effect could weaken due to a continued decline of E_{abs}, while the cooling effect from aerosol scattering could also weaken due to declining $PM_{2.5}$ concentration (Kelesidis et al., 2022; Samset et al., 2025; Xie et al., 2025). Future work should address this potential contradiction.

4 Conclusions

This study developed and applied a novel hybrid measurement method, coupling in situ PAX with filter-based ISS, to quantitatively apportion ambient aerosol light absorption into contributions from BC, BrC, and the lensing effect. The method successfully provided a self-consistent, observationally constrained pathway to separate these three components, addressing a persistent challenge in atmospheric aerosol science.

Key findings from the annual and seasonal analysis of Beijing aerosols in 2023 include:

(1) Successful apportionment: The PAX-ISS method successfully deconvolved the total absorption, confirming BC as the dominant absorber. BrC contributed approximately 10% to the annual wavelength-averaged absorption, highlighting its non-negligible role even in an urban environment.

(2) Spectral Nature and Seasonal Variability of Lensing: The lensing-induced enhancement accounted for an annual average of 40.0% of total absorption but showed profound seasonal variability (winter: 52.0%; summer: 4.6%). Crucially, the apparent lensing effect ($B_{abs,lensing}$) exhibited strong spectral dependence, contracting significantly at shorter wavelengths (UV/blue). This contraction is inconsistent with a purely additive lensing model.

(3) Observational Evidence for the BrC Blocking Effect: The observed spectral contraction, particularly the near-zero or negative apparent lensing at short wavelengths

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in summer, provides compelling field-based evidence for a "blocking effect" caused by absorptive BrC coatings on BC cores. This effect partially offsets the traditional lensing enhancement, leading to the net apparent lensing captured by our method ($B_{abs,lensing} = B_{abs,Tlensing} - B_{abs,blocking}$). Our results align with and provide tangible support for previous theoretical and numerical studies proposing this mechanism.

(4) Absorption Enhancement Factors and Pollution Linkage: The derived wavelength-averaged BC absorption enhancement factor (E_{abs}) varied seasonally from 1.09 ± 0.15 (summer) to 2.18 ± 0.08 (winter), with an annual mean of 1.69 ± 0.10. These values correlated with seasonal $PM_{2.5}$ levels, linking stronger enhancement to more polluted, aged aerosol conditions.

(5) Caveats and limitations: 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. To address this, we have included the aerosol size distribution data, simultaneously measured with a Scanning Mobility Particle Sizer (SMPS), in our accompanying data set (see https://doi.org/10.5281/zenodo.18552344, Shen et al., 2026) for reference by the community. Future research should aim to integrate such size-resolved measurements and multi-component characterizations (e.g., PAX coupled with a thermal denuder (Cappa et al., 2012; Liu et al., 2015; Pokhrel et al., 2017), SP2 (Dahlkötter et al., 2014), or VTDMA (Zhang et al., 2016)) to better constrain the dependence of absorption enhancement on microphysical and compositional variables.

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Data availability. The primary data used in this study can be obtained from https://doi.org/10.5281/zenodo.18552344 (Shen et al., 2026). In addition, we include the aerosol size data we simultaneously observed using an SMPS into above data set for reference. Other data utilized in the present study are available from the corresponding author on request.

Author contributions. Guorui Zhi and Yuzhe Zhang designed the research. Yi Shen and Guorui Zhi wrote the paper. Yi Shen, Wenjing Jin, Yao Kong, and Yuzhe Zhang carried out the field measurements. Yi Shen, Wenjing Jin, Yao Kong, Yuzhe Zhang, Zhengying Li, and Jianzhong Sun analyzed the data. Yuankai Wang and Zhijian Liang helped with the interpretation of the results.

Competing interests. The authors declare that they have no conflict of interest.

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