Supporting Information for

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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Section S1 Quality assurance measures for the sampling filters

A high-volume sampler was used to collect $PM_{2.5}$ samples on 8″ × 10″ quartz filters. Prior to sampling, the quartz filters were pre-baked at 550°C for 6 hours to remove adsorbed organics. Field blanks were also analysed to account for background contamination, and their concentrations were subtracted from the sample results. Filters deposited with $PM_{2.5}$ were stored at -20°C until thermal/optical analysis or integrating sphere analysis of carbonaceous aerosols was conducted.

The mass concentration of $PM_{2.5}$ (μg/m^3) collected on each filter was calculated according to the following formula:

$$PM_{2.5} = \frac{M_{collected} - M_{blank}}{V_{air}}, \quad \text{(S1)}$$

where $M_{\text{collected}}$ and M_{blank} represent the sample- and blank-filter masses (μg), respectively, and V_{air} represents the air volume (m^3) that accommodated the collected particulate matter. An analytical balance with a precision of 0.1 mg was used to obtain highly accurate mass measurements in the laboratory environment.

$PM_{2.5}$ samples were collected daily. Each filter was used for one day, from 9:00 a.m. on the first day to 8:30 a.m. the following day. The eligible days of $PM_{2.5}$ sampling are detailed in Table S1.

38 **Table S1.** Eligible days of $PM_{2.5}$ sampling.

Winter	Spring	Summer	Autumn
2/9/2023	4/7/2023	7/7/2023	10/12/2023
2/10/2023	4/8/2023	7/8/2023	10/13/2023
2/12/2023	4/9/2023	7/9/2023	10/14/2023
2/13/2023	4/10/2023	7/10/2023	10/15/2023
2/14/2023	4/11/2023	7/11/2023	10/16/2023
2/15/2023	4/12/2023	7/12/2023	10/17/2023
2/16/2023	4/13/2023	7/13/2023	10/18/2023
2/17/2023	4/17/2023	7/14/2023	10/19/2023
2/18/2023	4/18/2023	7/15/2023	10/20/2023
2/19/2023	4/19/2023	7/16/2023	10/21/2023
2/20/2023	4/20/2023	7/17/2023	10/22/2023
2/21/2023	4/24/2023	7/18/2023	10/23/2023
2/22/2023	4/25/2023	7/19/2023	10/24/2023
2/23/2023	4/26/2023	7/20/2023	10/25/2023
2/24/2023	4/27/2023	7/21/2023	10/26/2023
	4/28/2023	7/22/2023	10/27/2023
	4/29/2023	7/23/2023	10/28/2023
	4/30/2023	7/24/2023	10/29/2023
	5/1/2023	7/25/2023	10/30/2023
	5/2/2023	7/26/2023	10/31/2023
	5/3/2023	7/27/2023	11/1/2023
	5/5/2023	7/28/2023	11/2/2023
	5/6/2023	7/29/2023	11/3/2023
	5/7/2023	7/30/2023	11/4/2023
	5/8/2023	7/31/2023	11/5/2023
	5/9/2023	8/1/2023	
		8/2/2023	
		8/3/2023	
		8/4/2023	
		8/5/2023	
		8/6/2023	

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Section S2 Measurement of conversion factor

The sample holder inside the integrating sphere chamber (ISS) absorbs light from multiple directions rather than a single direction. As a result, the absorbance measured by the IS system (ABS_{ISS}) differs from that obtained using conventional methods (*ABS*), in which the incident light comes from one direction only. Photoacoustic spectroscopy (PAX) is also considered a conventional method. To convert ABS_{ISS} to *ABS*, a conversion factor (*CF*) was determined by comparing the absorbances of Humic Acid Sodium Salt (HASS) obtained from the ISS system (ABS_{ISS}) and from conventional methods (*ABS*) (e.g., using a cuvette outside the integrating sphere).

$$CF = \frac{ABS_{ISS}}{B_{abs}} \quad (S2)$$

Specifically, a 40 μg/ml aqueous solution of HASS was first prepared in a volumetric flask. Then, 3 ml of the solution was drawn using a pipette and transferred into a 1 cm × 1 cm × 4 cm cuvette. The cuvette was placed into the sample holder inside ISS, and the ABS_{ISS} of the HASS solution was measured. Another 3 ml aliquot of the same HASS solution was transferred into an identical cuvette and placed in the sample holder outside the ISS. The conventional absorbance (*ABS*) was then measured following principles and procedures similar to those of standard spectrophotometers. Both ABS_{ISS} and *ABS* were measured relative to pure water. The *CF* value determined in this study was 2.0.

Section S3 Calculation and correction of absorption values in thermal/optical analysis

The 7-wavelength transmission laser signals from the DRI Model 2015 were used to calculate the light attenuation (*ATN*):

$$ATN(\lambda) = -ln\frac{I_i(\lambda)}{I_f(\lambda)} \ , \quad \text{(S3)}$$

where I_i and I_f are the light intensities transmitted through the filter before (i = initial) and after (f = final) thermal carbon analysis. The absorption coefficients were calculated based on the corrected $ATN(\lambda)$ values, taking into account the filter's multiple scattering and particulate loading (Chen et al., 2015; Chow et al., 2018; Peng et al., 2020). The loading effect refers to the decrease in light attenuation due to "overloading", also known as the shadowing effect (Cheng et al., 2011; Chow et al., 2018; Li et al., 2020; Ram and Sarin, 2009; Virkkula et al., 2007). The multiple-scattering effect arises from the filter matrix itself, which makes the total light attenuation through the filter noticeably higher than the actual absorption coefficient (B_{abs}) (Arnott et al., 2005; Weingartner et al., 2003). We used the same correction method as in Chow et al. (2021), where samples were classified into 14 ranges with increments of 1 to 5 μg/cm^2 of EC filter loadings. $ATN(\lambda)$ values diminish with increased EC loadings. *ATN* at each wavelength was then adjusted upward to compensate for this decrease. The corrected $ATN(\lambda)$ is linearly related to EC loading and, generally, to the light absorption coefficient (B_{abs}). Such adjustment factors for EC loadings at different wavelengths were originally given in Chow et al (2021) and are presented here in Table S2.

Since aerosol absorption decreases exponentially with increasing wavelength, the effective absorption Ångström exponent (AAE_e) can be estimated by fitting all seven wavelengths with a derived coefficient (C):

$$ATN(\lambda) = C \times \lambda^{-AAE_e} \quad \text{(S4)}$$

Table S2 Attenuation adjustment factors for different EC loadings and wavelengths (Chow et al., 2021).

EC range (μg/cm^2)	ATN_{405}	ATN_{445}	ATN_{532}	ATN_{635}	ATN_{780}	ATN_{808}	ATN_{980}
0-3	1	1	1	1	1	1	1
3-4	1.21	1.19	1.17	1.12	1.13	1.12	1.14
4-5	1.32	1.28	1.25	1.18	1.18	1.18	1.2
5-6	1.42	1.36	1.31	1.23	1.22	1.22	1.24
6-7	1.5	1.43	1.36	1.26	1.25	1.24	1.26
7-8	1.58	1.5	1.43	1.3	1.29	1.29	1.3
8-9	1.64	1.55	1.46	1.33	1.31	1.31	1.33
9-10	1.72	1.62	1.51	1.36	1.33	1.34	1.34
10-11	1.79	1.68	1.57	1.41	1.38	1.38	1.39
11-13	1.88	1.75	1.62	1.45	1.41	1.41	1.42
13-16	2.07	1.91	1.75	1.53	1.49	1.48	1.48
16-20	2.36	2.17	1.99	1.71	1.65	1.66	1.66
20-25	2.71	2.53	2.3	1.97	1.91	1.91	1.93
>25	3.6	3.31	3.07	2.62	2.53	2.53	2.51

Section S4 Dual-Wavelength Iterative Calculation for Quantifying BC and BrC Absorbance

Calibration curves were established for Carbon Black (CarB) masses ranging from 3 μg to 21 μg and Humic Acid Sodium Salt (HASS) masses ranging from 5 μg to 72 μg. These curves relate mass to absorbance measured by the instrument at both 650 nm and 420 nm (Figure S2). CarB and HASS served as calibration standards for Black Carbon (BC) and Brown Carbon (BrC), respectively. The use of two wavelengths is essential due to the distinct spectral dependence of BC and BrC absorption. This difference enabled the mathematical separation of BrC absorbance from BC absorbance through an iterative calculation procedure.

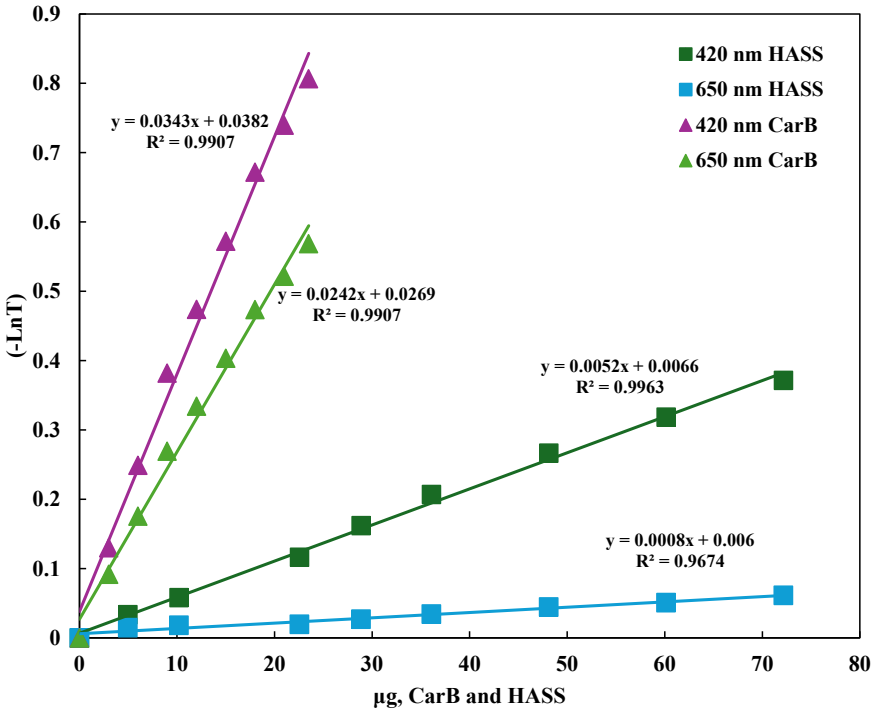

Figure S1 Calibration curves for CarB and HASS at 420 nm and 650nm. T represents the light transmitted through the calibration solution; (-lnT) is absorbance.

The iterative procedure is as follows:

1. Initial BC mass calculation (650 nm): Initiate an iteration cycle by calculating the BC mass. At 650 nm, the total absorbance is initially assumed to originate solely from BC. This mass (m_{BC}) is calculated using the CarB calibration equation at 650

nm (y = 0.0242x + 0.0269).

2. BC absorbance at 420 nm: Substitute the obtained m_{BC} into the CarB calibration equation at 420 nm (y = 0.0343x + 0.0382) to calculate the absorbance attributable to BC at 420 nm ($A_{BC,420}$).
3. BrC absorbance at 420 nm: Subtract $A_{BC,420}$ from the measured total absorbance at 420 nm ($A_{total,420}$). The residual absorbance is attributed to BrC ($A_{BrC,420}$).
4. BrC Mass calculation: Substitute $A_{BrC,420}$ into the HASS calibration equation at 420 nm (y = 0.0052x + 0.0066) to calculate the BrC mass (m_{BrC}).
5. BrC absorbance at 650 nm: Substitute m_{BrC} into the HASS calibration equation at 650 nm (y = 0.0008x + 0.006) to calculate the absorbance attributable to BrC at 650 nm ($A_{BrC,650}$).
6. Refined BC absorbance at 650 nm: Subtract $A_{BrC,650}$ from the measured total absorbance at 650 nm ($A_{total,650}$). The residual absorbance at 650 nm is now attributed to BC ($A_{BC,650}$).
7. Convergence check & next cycle: Use the refined $A_{BC,650}$ to start a new iteration cycle by returning to step (1). Iteration continues until the calculated absorbances for both BC and BrC converge (i.e., when the difference between two consecutive iterative cycles is negligible). The absorbances from the final convergent cycle are accepted as the true absorbances for BC and BrC in the sample.

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