



Size distribution and diurnal variations of brown carbon absorption using DMF and methanol extraction methods: Implications for the origins of extremely low-volatility organics

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ABSTRACT

In this study, paired PM_{2.5} and PM₁₀ samples with low time resolution (11 h) and time-resolved PM_{2.5} samples (2–6 h) were collected from northern Nanjing, China, during summer and winter. The light absorption of organic carbon (OC) in each sample was measured for N,N-dimethylformamide (DMF) and methanol extracts, respectively. The light absorption coefficient of DMF extracts at 365 nm (Abs_{365,d}) showed no significant difference (Student's *t*-test, *p* > 0.05) between PM_{2.5} and PM₁₀ samples, consistent with existing observations for methanol extracts (Abs_{365,m}). However, the mean Abs_{365,d} values for PM_{2.5} and PM₁₀ were significantly higher (*p* < 0.01) than the corresponding Abs_{365,m} values, as extremely low volatility OC (ELVOC) with strong light absorption is more soluble in DMF than in methanol. This difference also applies to other cities in the central Yangtze River Delta, China, based on direct measurements. Diurnal variation analysis using time-resolved PM_{2.5} samples showed that Abs_{365,d} was greater than Abs_{365,m} mainly at 8:00–10:00 and 20:00–2:00 in both summer and winter, indicating that ELVOC from fresh emissions during the morning rush hour and from nighttime oxidation was likely more soluble in DMF than in methanol. Source apportionment of Abs_{365,d} and Abs_{365,m} was conducted using concentration data of organic molecular markers in the same samples. The difference in the contribution of resuspended road dust mixed with combustion emissions to Abs_{365,d} and Abs_{365,m} showed similar diurnal variations to measured Abs_{365,d} minus Abs_{365,m}, suggesting it may be an important source of light-absorbing ELVOC.

1. Introduction

Light-absorbing organic carbon (OC), also known as “brown carbon” (BrC), has been intensively studied over the past two decades because of its significant atmospheric warming effect. It is thought to originate mainly from the combustion of biomass and fossil fuels, as well as associated secondary processes (Laskin et al., 2015; Laskin et al., 2025). Recent studies have identified a fraction of BrC, called “dark BrC” (d-BrC), in biomass burning smoke that absorbs strongly in the visible and near-infrared regions (Chakrabarty et al., 2023; Chelluboyina et al., 2024). This type of BrC is water-insoluble and may exist as viscous, low-volatility tar balls (Hoffer et al., 2017; Adler et al., 2019; Sedlacek

III et al., 2018). Field measurements show that d-BrC absorption accounts for 75% and 50% of total aerosol absorption at short (405/488 nm) and long (664 nm) wavelengths, respectively, in wildfire plumes in the western US (Chakrabarty et al., 2023). Wang et al. (2025) reported a radiative effect of +0.208 W m⁻² (+0.02 – 0.68 W m⁻²) for d-BrC from biomass burning based on a global model simulation, which is comparable to that of black carbon (BC, +0.269 W m⁻²) and much higher than traditional estimates for BrC (<0.15 W m⁻²).

The solvent extraction of ambient or laboratory-generated particulate matter (PM) followed by analysis using UV/Vis spectroscopy and gas or liquid chromatography coupled to various mass spectrometry techniques, has been widely used to investigate the relationship between

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chemical composition and the light-absorbing properties of OC (Lin et al., 2015; Lin et al., 2016; Samburova et al., 2016; Xie et al., 2019a). Water and methanol are the most commonly used solvents for OC extraction (Chen and Bond, 2010; Liu et al., 2013; Zhang et al., 2013), and the water-insoluble fraction dominates BrC absorption (Cheng et al., 2016a; Huang et al., 2020; Soleimanian et al., 2020; Huang et al., 2018). Di Lorenzo and Young (2016) and Di Lorenzo et al. (2017) separated biomass burning OC extracts using size exclusion chromatography and attributed BrC absorption primarily to large molecular weight (MW) components (>500–1000 Da), which were classified as extremely low volatility OC (ELVOC). Online measurements also showed that biomass burning OC absorption is associated mostly with ELVOC (Saleh et al., 2014; Shetty et al., 2023).

By measuring OC concentrations in filter samples before and after solvent extraction, it was found that methanol can dissolve more than 90% of biomass burning OC (Xie et al., 2017; Chen and Bond, 2010), while less than 85% of ambient OC can be extracted using methanol (Xie et al., 2019b; Xie et al., 2022). The small fraction of methanol-insoluble OC may contain ELVOC with strong light absorption (Bai et al., 2020; Atwi et al., 2022). Xu et al. (2022) found that low-volatility OC evaporating at >280 °C from the ambient was more soluble in N, N-dimethylformamide (DMF, ~90%) than in methanol (~80%), resulting in significantly (Student's *t*-test, $p < 0.01$) higher light absorption in DMF extracts than in methanol extracts. These results indicate that, besides biomass burning, other sources contribute to ELVOC. For example, Corbin et al. (2019) identified tar BrC from heavy oil combustion with physical and optical properties similar to biomass burning tar balls. Xie et al. (2017) measured the light absorption of methanol-extractable OC (MEOC) emitted by various motor vehicles, and the mean fraction of MEOC was $75.9\% \pm 9.42\%$. Therefore, increasing the extraction efficiency of ELVOC would reduce the underestimation of BrC absorption when using the methanol extraction method and improve understanding of ELVOC composition.

Evidence shows that BrC absorption in methanol extracts is mainly enriched in fine PM (Liu et al., 2013; Wu et al., 2020). However, coarse PM contains substantial large organic molecules from natural sources (e. g., soil; Samburova et al., 2005; Park and Son, 2016), which may include light-absorbing chromophores that are not soluble in methanol. The diurnal variations of methanol-extractable BrC generally follow those of biomass burning tracers (e.g., levoglucosan; Feng et al., 2023). Since methanol has limited ability to dissolve ELVOC, the diurnal pattern of BrC absorption based on the methanol extraction method may be biased. In this study, paired PM_{2.5} and PM₁₀ samples and time-resolved PM_{2.5} samples were collected during two sampling campaigns in northern Nanjing, China (Feng et al., 2023; Cui et al., 2025), and analyzed for BrC absorption using the DMF extraction method. The light absorption of methanol extracts from the same PM samples was obtained from existing studies for comparison (Feng et al., 2023; Cui et al., 2025), along with concentration data of bulk components and organic molecular markers (OMMs) for source apportionment using positive matrix factorization (PMF). The results reveal the size distribution and diurnal variations of DMF extract absorption and their sources, improving our understanding of the origins of ELVOC in the atmosphere.

2. Methods

2.1. Sample collection

In this study, paired integrated PM_{2.5} and PM₁₀ samples, as well as time-resolved PM_{2.5} samples, were collected during two sampling campaigns in northern Nanjing, China. The sampling site was located on the rooftop of a seven-story library building at Nanjing University of Information Science and Technology (NUIST; 118.71°E, 32.21°N). Details on the sampler setup and sampling protocols are provided in Cui et al. (2025) and Feng et al. (2023). Briefly, 11-h PM_{2.5} and PM₁₀ samples were collected using two identical samplers (Laoying2030.

Longying Environmental, China). The samplers were installed with 2.5 μm and 10 μm cut-point impactors, respectively, and operated at a flow rate of 100 L min⁻¹. Both PM_{2.5} and PM₁₀ were collected on prebaked (550 °C for 4 h) quartz fiber filters during 8:00 – 19:00 (daytime) and 20:00 – 7:00 (overnight, nighttime) in summer 2020 (05/31/2020 – 09/07/2020, 147 pairs) and winter 2020–2021 (12/01/2020 – 01/25/2021, 93 pairs). Time-resolved PM_{2.5} samples were collected using two identical samplers (PM_{2.5}-PUF-300, Mingye Environmental, China) operated alternately at a flow rate of 300 L min⁻¹. Each sampler was equipped with a 2.5 μm cut-point impactor and a prebaked quartz filter. Two-hour PM_{2.5} samples were collected from 8:00 to 20:00 (daytime), and 6-h samples were collected from 20:00 to 8:00 the next day (nighttime). Time-resolved sampling was conducted for eight days in winter 2020 (01/01/2020 – 01/14/2020, $N = 64$) and for twenty-four days in summer 2020 (06/01/2020 – 09/11/2020, $N = 192$).

To demonstrate that the difference in BrC absorption between DMF and methanol extraction methods was not limited to northern Nanjing, 23-h PM_{2.5} samples were collected from downtown Nanjing (118.71°E, 32.06°N), Suzhou (120.64°E, 31.28°N), Wuxi (120.32°E, 31.51°N), Changzhou (120.02°E, 31.78°N), and Zhenjiang (119.44°E, 32.19°N), China in fall 2020 (09/01/2020 – 11/24/2020) during another sampling campaign (Feng et al., 2024). These five cities are located in the center of the Yangtze River Delta (YRD), one of the most developed regions in China. During the sampling period, synchronous PM_{2.5} samples were collected every three days ($N = 29$ for each site) using a four-channel sampler (TH-16a, Wuhan Tianhong, China) at all five sites from 10:00 to 9:00 the next day, with all samplers operating at a flow rate of 16.7 L min⁻¹.

2.2. Light absorption measurement

The measurement of filter extract absorption followed the method described in Xu et al. (2022). In brief, one-quarter of each filter sample was extracted in 10 mL DMF using ultrasonic treatment for 15 min. After filtration, the light absorbance of the DMF extracts ($A_{\lambda,d}$) over 200–900 nm was measured with a UV/Vis spectrometer (UV-1900, Shimadzu Corporation, Japan) and converted to light absorption coefficients ($Abs_{\lambda,d}$, Mm⁻¹) as follows:

$$Abs_{\lambda,d} = (A_{\lambda,d} - A_{700,d}) \times \frac{V_l}{V_a \times L} \ln(10) \quad (1)$$

where $A_{700,d}$ is subtracted to adjust for baseline shift, V_l (m³) is the volume of solvent used for filter extraction, V_a (m³) is the air volume of the extracted filter aliquot, L (0.01 m) is the optical path length in the cuvette, and $\ln(10)$ is used to convert $Abs_{\lambda,d}$ from base-10 to the natural logarithm (Hecobian et al., 2010). The solution mass absorption efficiency of DMF extracts ($MAE_{\lambda,d}$, m² g⁻¹) was calculated as follows:

$$MAE_{\lambda,d} = \frac{Abs_{\lambda,d}}{c_d} \quad (2)$$

where c_d represents the concentration of OC extracted by DMF, calculated by multiplying the OC concentrations of individual PM samples by the mean extraction efficiency of DMF (89.0%) as determined by Xu et al. (2022). OC concentrations for all PM samples in this study were analyzed separately by Feng et al. (2023, 2024) and Cui et al. (2025). Due to the small sample volume, the $A_{\lambda,d}$ values at $\lambda > 400$ nm were mostly close to 0 and subject to large uncertainty. As such, the solution absorption Ångström exponent (Å), typically defined as the absolute value of the regression slope for $\lg(Abs_{\lambda})$ against $\lg(\lambda)$ over 300–550 nm, was not calculated. To allow comparison with previous studies using methanol for filter extraction, $Abs_{\lambda,d}$ and $MAE_{\lambda,d}$ values at 365 nm ($Abs_{365,d}$ and $MAE_{365,d}$) are reported here. The light absorption coefficients and mass absorption efficiency of methanol extracts at 365 nm ($Abs_{365,m}$ and $MAE_{365,m}$) for integrated PM_{2.5} and PM₁₀ samples, time-resolved PM_{2.5} samples, and 23-h PM_{2.5} samples from five cities in

the central YRD region were determined using the same method with different sample aliquots. The measurement results were obtained from Cui et al. (2025), Feng et al. (2023), and Feng et al. (2025), respectively, for comparison. An extraction efficiency of 82.4% for total OC using methanol was determined at the same sampling site (Xie et al., 2022).

2.3. Source analysis

The differences in light absorption between DMF and methanol extracts were due to ELVOC not being soluble in methanol at the sampling site at NUIST (Xu et al., 2022). Comparisons of source distributions between DMF and methanol extract absorption can indicate the origins of methanol-insoluble ELVOC. In this study, compositional data of integrated and time-resolved samples, including bulk components (inorganic ions, OC, and elemental carbon (EC)), nonpolar OMMs (n-alkanes, PAHs, steranes, and hopanes), and polar OMMs (isoprene SOA products, levoglucosan, and sugar polyols), obtained from Cui et al. (2025) and Feng et al. (2023), were combined with $Abs_{365,d}$ and $Abs_{365,m}$ as input for source apportionment using the PMF model (EPA PMF 5.0). Because the number of time-resolved samples in winter is much smaller than in summer and a large number of observations is required for robust PMF analysis, integrated and time-resolved $PM_{2.5}$ samples from northern Nanjing were pooled as the input data set. The selection of input species followed the method described in Feng et al. (2023). Briefly, $PM_{2.5}$ species were prescreened based on their representativeness of emission sources or atmospheric processes, total fraction (<20%) of missing values and measurements below detection limits (BDL), mean signal-to-noise ratio (concentration/uncertainty >3), and the physical meaningfulness of output factor profiles. To minimize the influence of gas-particle partitioning of non-polar OMMs on PMF analysis, non-polar OMMs with vapor pressures ($p^{0,*}_L$) significantly greater than 10^{-9} atm at 298.15 K were excluded. EC was not considered due to its high fraction of missing and BDL values in time-resolved samples during summer. The target polar OMMs were expected to dissolve in aerosol liquid water, with minor fractions present in the gas phase (Feng et al., 2023), so using only particle-phase data may not affect the PMF analysis. The final input data set for PMF analysis contained 479 observations of 35 species ($Abs_{365,d}$, $Abs_{365,m}$, NH_4^+ , NO_3^- , SO_4^{2-} , Ca^{2+} , OC, 21 non-polar OMMs, and 7 polar OMMs; Table S1). The preparation of the uncertainty data set, treatment of missing and BDL values, and robustness evaluation for PMF analysis were performed identically to previous studies (Xie et al., 2022; Feng et al., 2023; Cui et al., 2025) and are described in detail in the supporting information (Text S1 and Table S2).

3. Results and discussion

3.1. Size distribution

Table 1 presents the median and mean Abs_{365} and MAE_{365} values for DMF and methanol extracts of $PM_{2.5}$ and PM_{10} . In both summer and winter, the time series of $Abs_{365,d}$ and $Abs_{365,m}$ for $PM_{2.5}$ samples nearly overlapped with those for PM_{10} samples (Fig. S1). Except for $Abs_{365,m}$ measurements in summer, the mean $Abs_{365,d}$ and $Abs_{365,m}$ in $PM_{2.5}$

samples showed no significant difference ($p > 0.05$) compared to those in PM_{10} samples. As shown in Fig. S1b, the difference in mean $Abs_{365,m}$ between $PM_{2.5}$ and PM_{10} in summer was directly related to a few high $Abs_{365,m}$ values for $PM_{2.5}$ samples and possibly to uncertainties in sampling and light absorbance analysis (Cui et al., 2025). These results indicate that nearly all light-absorbing chromophores are enriched in fine particles, consistent with previous studies (Liu et al., 2013; Wu et al., 2020). Therefore, focusing on the light-absorbing properties of OC in fine particles is unlikely to underestimate BrC absorption, even if the extraction solvent is changed from methanol to DMF. Because mean OC concentrations in PM_{10} were more than 20% higher than those in $PM_{2.5}$ for both summer and winter, and the OC fractions in coarse particles have negligible light absorption, the mean $MAE_{365,d}$ (summer $0.73 \pm 0.38 \text{ m}^2 \text{ g}^{-1}$, winter $1.39 \pm 0.30 \text{ m}^2 \text{ g}^{-1}$) and $MAE_{365,m}$ (summer $0.74 \pm 0.33 \text{ m}^2 \text{ g}^{-1}$, winter $1.23 \pm 0.26 \text{ m}^2 \text{ g}^{-1}$) of $PM_{2.5}$ samples were significantly ($p < 0.01$) higher than those of PM_{10} samples ($MAE_{365,d}$: summer $0.54 \pm 0.20 \text{ m}^2 \text{ g}^{-1}$, winter $1.12 \pm 0.26 \text{ m}^2 \text{ g}^{-1}$; $MAE_{365,m}$: summer $0.49 \pm 0.15 \text{ m}^2 \text{ g}^{-1}$, winter $0.99 \pm 0.25 \text{ m}^2 \text{ g}^{-1}$).

Although $Abs_{365,d}$ and $Abs_{365,m}$ were significantly correlated ($p < 0.01$) in $PM_{2.5}$ and PM_{10} during both summer and winter (Fig. 1a and b), the mean $Abs_{365,d}$ values for $PM_{2.5}$ and PM_{10} were significantly ($p < 0.05$) higher than their mean $Abs_{365,m}$ values (Table 1). Xu et al. (2022) demonstrated that the difference between $Abs_{365,d}$ and $Abs_{365,m}$ of $PM_{2.5}$ from the same sampling site is primarily due to DMF dissolving more

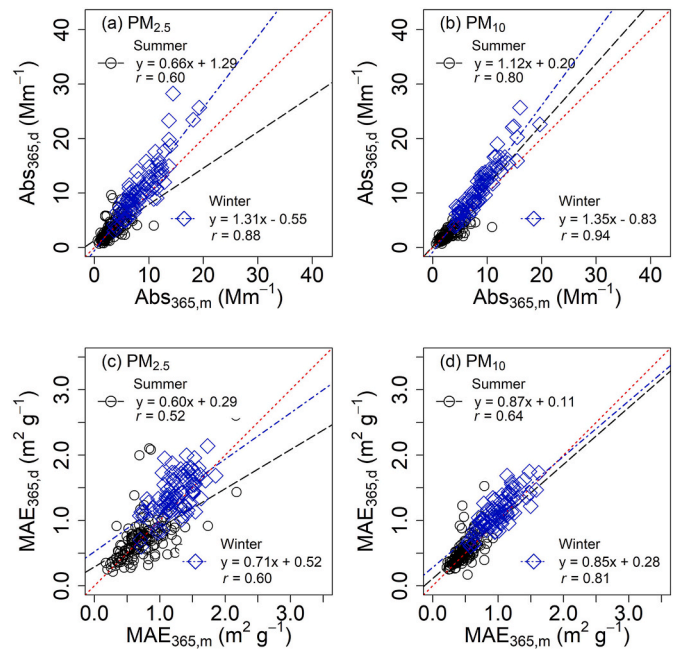


Fig. 1. Comparisons of (a, b) Abs_{365} and (c, d) MAE_{365} values between the DMF and methanol extraction methods for $PM_{2.5}$ and PM_{10} samples in summer and winter.

Table 1

Median and mean Abs_{365} and MAE_{365} values for DMF and methanol extracts of $PM_{2.5}$ and PM_{10} samples in northern Nanjing during summer 2020 and winter 2020–2021.

	PM2.5				PM10			
	Summer (N = 147)		Winter (N = 93)		Summer (N = 147)		Winter (N = 93)	
	Median	Mean	Median	Mean	Median	Mean	Median	Mean
$Abs_{365,d}(\text{Mm}^{-1})$	2.96	3.38 ± 1.79	9.77	10.7 ± 4.67	2.93	3.27 ± 1.66	10.03	10.7 ± 4.52
$MAE_{365,d}(\text{m}^2 \text{ g}^{-1})$	0.70	0.73 ± 0.38	1.34	1.39 ± 0.30	0.51	0.54 ± 0.20	1.08	1.12 ± 0.26
$Abs_{365,m}(\text{Mm}^{-1})$	2.76	3.14 ± 1.62	7.81	8.63 ± 3.13	2.61	2.76 ± 1.19	8.51	8.62 ± 3.16
$MAE_{365,m}(\text{m}^2 \text{ g}^{-1})$	0.69	0.74 ± 0.33	1.22	1.23 ± 0.26	0.47	0.49 ± 0.15	0.97	0.99 ± 0.25
OC ($\mu\text{g m}^{-3}$)	4.95	5.50 ± 2.31	8.58	8.76 ± 3.11	6.34	6.80 ± 2.12	10.77	10.9 ± 4.02

ELVOC than methanol, and excluded the possibility of a “solvent effect”. In northern Nanjing, $MAE_{365,d}$ and $MAE_{365,m}$ were also significantly correlated ($p < 0.01$; Fig. 1c and d), with a stronger correlation consistently observed in winter. However, a significant difference ($p < 0.01$) between $MAE_{365,d}$ and $MAE_{365,m}$ was found only in winter. These results may be attributed to stronger primary emissions from biomass and fossil fuel combustion in winter and more intensive photobleaching of BrC in summer.

As shown in Fig. 2, the mean $Abs_{365,d}$ (3.83 ± 1.53 – 5.09 ± 1.53 Mm^{-1}) and $MAE_{365,d}$ (0.92 ± 0.27 – 1.30 ± 0.52 $m^2 g^{-1}$) values of $PM_{2.5}$ samples from the five urban sites were all greater than their mean $Abs_{365,m}$ (2.49 ± 1.75 – 4.16 ± 1.70 Mm^{-1}) and $MAE_{365,m}$ (0.76 ± 0.27 – 1.27 ± 0.43 $m^2 g^{-1}$) values in fall 2020. Except for urban Nanjing and Changzhou, the differences at the other three urban sites were significant ($p < 0.01$). Therefore, the fact that DMF can dissolve more light-absorbing chromophores applies not only to northern Nanjing but also to at least other cities in the central YRD region. Considering the higher MAE_{365} values for DMF extracts compared to methanol extracts in fall and winter in the central YRD region, the additional ELVOC extracted by DMF should have greater MAE_{365} than that of MEOC in cold seasons.

3.2. Diurnal variations

Fig. 3 shows the mean values of Abs_{365} and MAE_{365} for DEOC and MEOC, as well as the concentrations of OC, levoglucosan, and total PAHs during individual sampling intervals for time-resolved $PM_{2.5}$ samples in winter and summer. The corresponding standard deviations are provided in Table S3. As shown in Fig. 3a–e, the diurnal pattern of mean $Abs_{365,d}$ generally resembles those of levoglucosan and total PAHs—specific tracers for combustion sources such as biomass and coal burning (Simoneit et al., 1999; Yan et al., 2019)—from 8:00 to 20:00 in winter, with high levels in the morning and late afternoon due to increased primary emissions (motor vehicles and domestic heating) and low levels in the middle of the day due to a higher planetary boundary layer height (PBLH) and “photobleaching” (Hecobian et al., 2010; Feng et al., 2023). This indicates combined influences from anthropogenic activities and meteorological changes.

In winter, the mean $Abs_{365,d}$ was significantly ($p < 0.05$) higher than the mean $Abs_{365,m}$ at 8:00–10:00, and the difference diminished during 12:00–14:00 (Fig. 3a). Since light-absorbing ELVOC is recalcitrant BrC and contributes a greater fraction of total absorption as ambient OC ages (Di Lorenzo et al., 2017; Chakrabarty et al., 2023), the ELVOC soluble in DMF only at 8:00–10:00 may gradually become soluble in methanol with daytime $\cdot OH$ oxidation. From 12:00 to 20:00, $Abs_{365,d}$ and $Abs_{365,m}$ varied similarly, and the slightly higher $Abs_{365,d}$ than $Abs_{365,m}$ in the afternoon was likely due to secondarily formed ELVOC with higher

solubility in DMF. Light-absorbing oligomers with low volatility can form from small molecules through gas-phase (Lin et al., 2014), aqueous (Lavi et al., 2017; Kasthuriarachchi et al., 2025), and surface reactions (Ling et al., 2020; Link et al., 2020). The peak $Abs_{365,m}$ value overlapped with that of $Abs_{365,d}$ at 18:00–20:00, when biomass burning and other combustion emissions for domestic heating dominated (Feng et al., 2023), indicating similar solubility of ambient OC in DMF and methanol during this sampling interval. As mentioned in the Introduction, methanol can dissolve more than 90% of biomass burning OC (Chen and Bond, 2010; Xie et al., 2017), which is similar to the extraction efficiency of ambient OC (89.0%) in DMF for northern Nanjing (Xu et al., 2022). After 20:00, high $Abs_{365,d}$ values remained until 2:00, then decreased during 2:00–8:00 (Fig. 3a). Meanwhile, $Abs_{365,m}$ and the concentrations of OC, levoglucosan, and total PAHs all decreased after 20:00 (Fig. 3a–e). This may be because secondarily formed ELVOC at night or primary ELVOC that experienced nighttime oxidation are less soluble in methanol. Evidence indicates that nighttime oxidation of volatile organic compounds is a plausible source of BrC (Chen et al., 2023; Jiang et al., 2019; Kaskaoutis et al., 2026), and nighttime aging (e.g., $\cdot NO_3$ oxidation) of pre-existing ELVOC can enhance its absorption (Kuang et al., 2025; Li et al., 2020), possibly by adding nitrogen-containing chromophores (Chakrabarty et al., 2023).

In summer, differences in diurnal variations between $Abs_{365,d}$ and $Abs_{365,m}$ were mainly observed in the morning (8:00–10:00, $p < 0.01$) and at night (20:00–8:00, $p < 0.05$; Fig. 3b). From 10:00 to 20:00, $Abs_{365,d}$ and $Abs_{365,m}$ showed similar variations with no significant ($p > 0.10$) differences. Due to stronger photooxidation in summer, $Abs_{365,d}$ and $Abs_{365,m}$ converged earlier (10:00–12:00) than in winter. In addition to the nighttime formation and aging of ELVOC, the significant difference between $Abs_{365,d}$ and $Abs_{365,m}$ at 2:00–8:00 in summer may also be due to fresh emissions, as the morning rush hour starts earlier in summer (before 8:00 a.m.) than in winter (Qi et al., 2025). Although the diurnal pattern of $Abs_{365,d}$ in summer was similar to those of levoglucosan and total PAHs (Fig. 3f), the variance magnitude of $Abs_{365,d}$ was much smaller, indicating additional contributing sources beyond biomass and fossil fuel combustion or atmospheric processes. The values of $MAE_{365,d}$ and $MAE_{365,m}$ were similar for most of the day in both winter and summer (Fig. 3c and d). $MAE_{365,d}$ was slightly greater than $MAE_{365,m}$ at 8:00–10:00 (morning) and 20:00–8:00 (nighttime), suggesting that the light-absorbing ELVOC freshly emitted in the morning and experiencing nighttime oxidation is more soluble in DMF and has a larger MAE_{365} than MEOC. However, caution is warranted, as $MAE_{365,d}$ and $MAE_{365,m}$ were calculated based on the mean extraction efficiency of ambient OC in DMF and methanol from existing measurements and are subject to uncertainties. The solubility of OC depends largely on its composition and the measurement technique, and it varies between

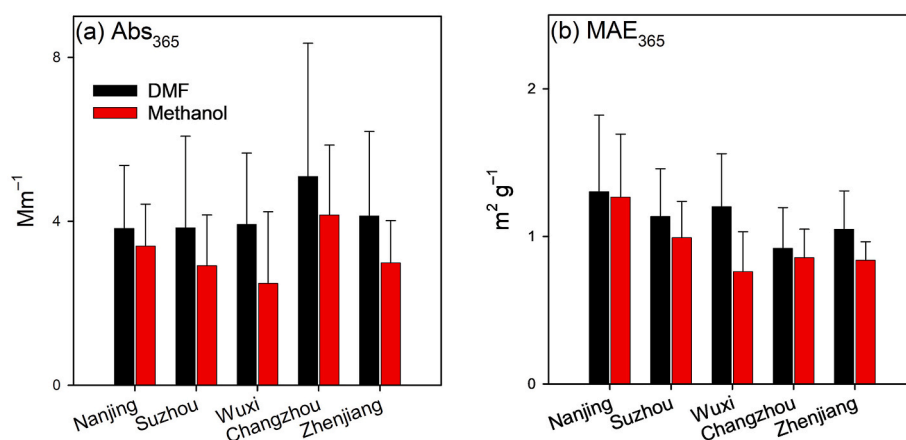


Fig. 2. Comparisons of mean (a) Abs_{365} and (b) MAE_{365} values between DMF and methanol extracts of $PM_{2.5}$ samples from five urban sites in the central YRD region. The whiskers represent one standard deviation.

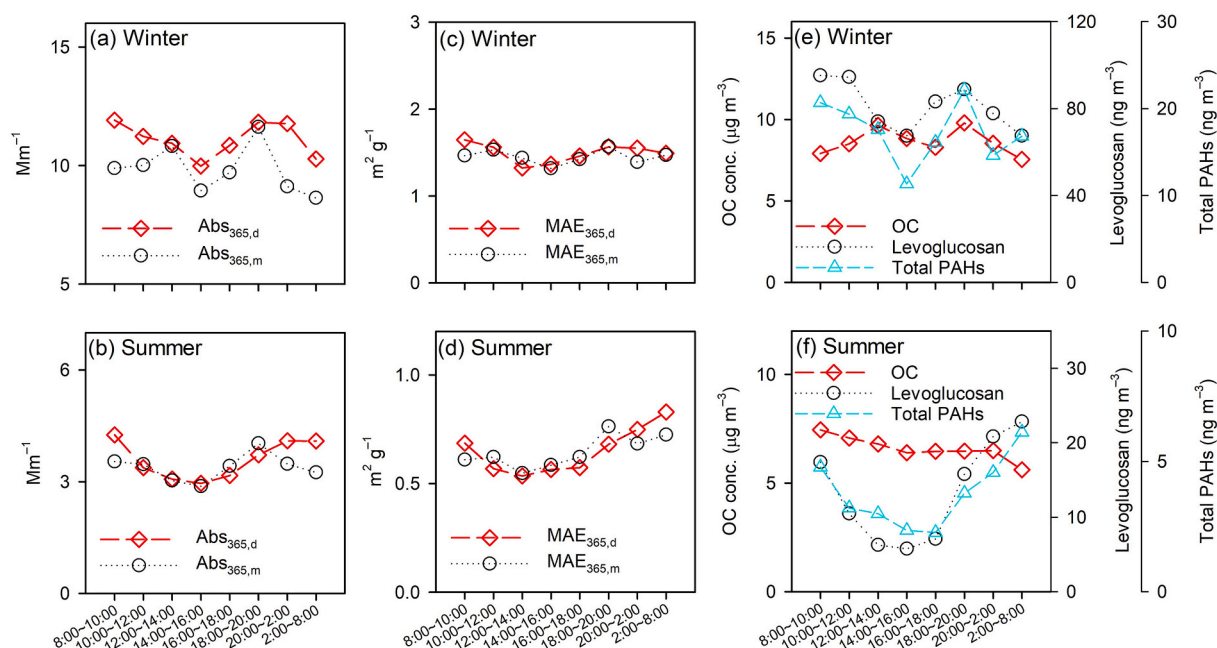


Fig. 3. Diurnal variations of (a, b) $\text{Abs}_{365,d}$ - $\text{Abs}_{365,m}$, (c, d) $\text{MAE}_{365,d}$ - $\text{MAE}_{365,m}$, and (e, f) OC-levoglucosan-total PAHs in winter and summer for northern Nanjing.

samples.

3.3. Source apportionment

An 8-factor PMF solution was finalized after evaluating individual solutions with factor numbers from four to ten. As discussed in Text S1, the 8-factor solution provides the most physically meaningful output factors; Q/Q_{exp} changed less significantly once the factor number exceeded eight; and the error estimation diagnostics of the 8-factor solution suggest robust PMF results (Table S2). Fig. S2 shows the normalized factor profiles used to relate each factor to a specific emission source or atmospheric process. The biomass burning factor is characterized by the highest loading of levoglucosan, and significant fractions of light n-alkanes and PAHs, which are also characteristic compounds from biomass burning (Schauer et al., 2001). Besides high proportions of the four sugar polyols, the biogenic emission factor also shows an odd-to-even carbon number predominance for n-alkanes (n-C26 – n-C35), indicating wax emissions from high plant leaves (Rogge et al., 1993a). PAHs in the study area were primarily associated with fossil fuel (e.g., coal) combustion from industrial activities (Yan et al., 2019; Gong et al., 2022). C5-alkene triols and 2-methyltetrols (C5-ATs and 2-MTs) are typical SOA products from isoprene oxidation (Claeys et al., 2004; Surratt et al., 2006). Steranes and hopanes are specific tracers related to the combustion of engine lubricating oil (Kaplan et al., 2001; Rogge et al., 1997). NH_4^+ , NO_3^- , and SO_4^{2-} are the dominant inorganic species in $\text{PM}_{2.5}$ and are formed through the neutralization of NH_3 with secondarily formed HNO_3 and SO_4^{2-} (Wang et al., 2016; Cheng et al., 2016b). Ca^{2+} indicates crustal soil emissions (Wang et al., 2005). Accordingly, the five factors characterized by these groups of source tracers were identified as industrial activity (high MW PAHs), isoprene SOA (C5-ATs and 2-MTs), lubricating oil combustion (aaa-CHO and abb-MeCHO), secondary inorganics (NH_4^+ , NO_3^- , and SO_4^{2-}), and soil crust (Ca^{2+}). Tire wear debris in road dust particles contain substantial high carbon number n-alkanes with no odd-to-even preference (Rogge et al., 1993b). In Fig. S2f, the resuspended road dust factor also contains some high MW PAHs and substantial levoglucosan, reflecting a mixture of road dust and deposited combustion particles. All these factors were identified in previous OMM-based source apportionment studies and are well explained (Xie et al., 2022; Feng et al., 2023; Feng et al., 2024; Cui

et al., 2025).

3.3.1. Distributions of factor contributions for integrated samples

Figs. S3 and 4 illustrate the averaged distributions of absolute and relative factor contributions to $\text{Abs}_{365,d}$, $\text{Abs}_{365,m}$, and OC for integrated $\text{PM}_{2.5}$ samples in northern Nanjing. Biomass burning ($\text{Abs}_{365,d}$ 5.14 Mm^{-1} , 48.1%; $\text{Abs}_{365,m}$ 4.85 Mm^{-1} , 50.6%) and resuspended road dust ($\text{Abs}_{365,d}$ 3.43 Mm^{-1} , 32.1%; $\text{Abs}_{365,m}$ 2.93 Mm^{-1} , 30.6%) are the two major contributors to BrC absorption in winter. Only the biomass burning factor has a significantly ($p < 0.01$) higher relative contribution to Abs_{365} than to OC concentrations (25.3%), supporting that the light-absorbing OC from biomass burning are strong BrC chromophores. Comparisons of factor contribution distributions between $\text{Abs}_{365,d}$ and $\text{Abs}_{365,m}$ show that industrial activity, lubricating oil combustion, resuspended road dust, and soil crust have higher absolute and relative contributions to $\text{Abs}_{365,d}$ than to $\text{Abs}_{365,m}$. Among the eight factors, the resuspended road dust factor shows the largest difference (0.50 Mm^{-1}) in absolute mean factor contributions between $\text{Abs}_{365,d}$ and $\text{Abs}_{365,m}$, followed by biomass burning (0.29 Mm^{-1}), soil crust (0.20 Mm^{-1}), and industrial activity (0.17 Mm^{-1}). Thus, the light-absorbing ELVOC soluble only in DMF is likely associated with combustion sources in winter and released through direct emissions or road dust resuspension.

In summer, five factors (biomass burning, biogenic emission, isoprene SOA, resuspended road dust, and secondary inorganics) each contributed significantly (>10%) to $\text{Abs}_{365,d}$ and together accounted for 86.9% of the total (Fig. 4). Compared to the distribution of factor contributions to $\text{Abs}_{365,m}$, the isoprene SOA factor showed much higher absolute (0.49 Mm^{-1}) and relative (16.7%) contributions to $\text{Abs}_{365,d}$. Lin et al. (2014) demonstrated the formation of light-absorbing oligomers from the reactive uptake of isoprene epoxydiols. As in winter, the industrial activity, lubricating oil combustion, resuspended road dust, and soil crust factors also had higher absolute and relative contributions to $\text{Abs}_{365,d}$ than to $\text{Abs}_{365,m}$. Therefore, in addition to combustion emissions and road dust resuspension, secondarily formed ELVOC, such as high MW polymers or humic-like substances, could be an additional potential source for the difference between measured $\text{Abs}_{365,d}$ and $\text{Abs}_{365,m}$ in summer.

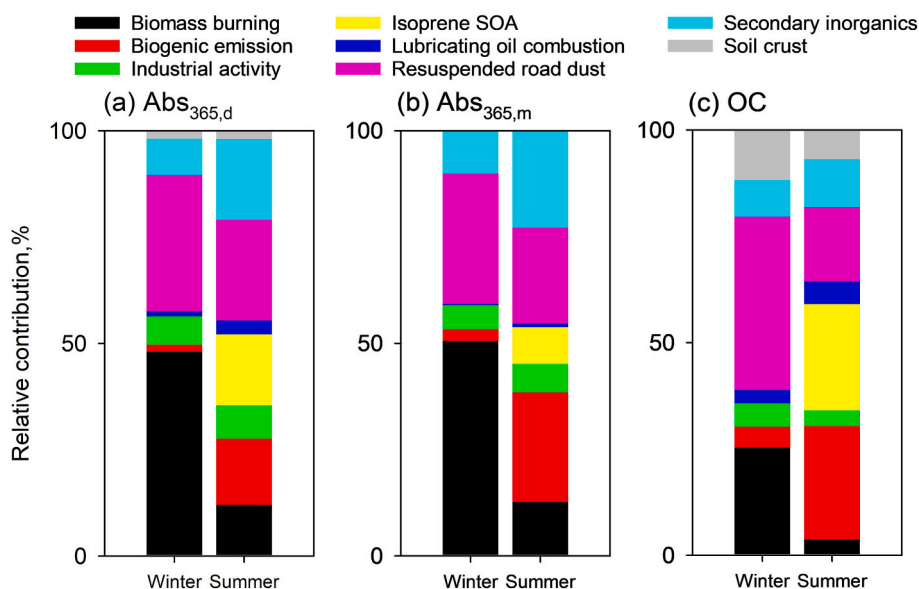


Fig. 4. Distributions of relative factor contributions to (a) $Abs_{365,d}$, (b) $Abs_{365,m}$, and (c) OC for integrated $PM_{2.5}$ samples during winter and summer in northern Nanjing.

3.3.2. Diurnal variations of factor contributions

The diurnal variations of absolute and relative factor contributions are illustrated using stack plots in Figs. S4 and 5. In winter, $Abs_{365,d}$ and $Abs_{365,m}$ displayed similar diurnal patterns in factor contribution

distributions. The biomass burning factor contributed the largest fractions to both $Abs_{365,d}$ ($3.93\text{--}6.08\text{ Mm}^{-1}$, 44.7%–58.4%) and $Abs_{365,m}$ ($3.72\text{--}5.74\text{ Mm}^{-1}$, 45.8%–59.3%). The diurnal patterns of the biomass burning and industrial activity factors were similar, with a significant

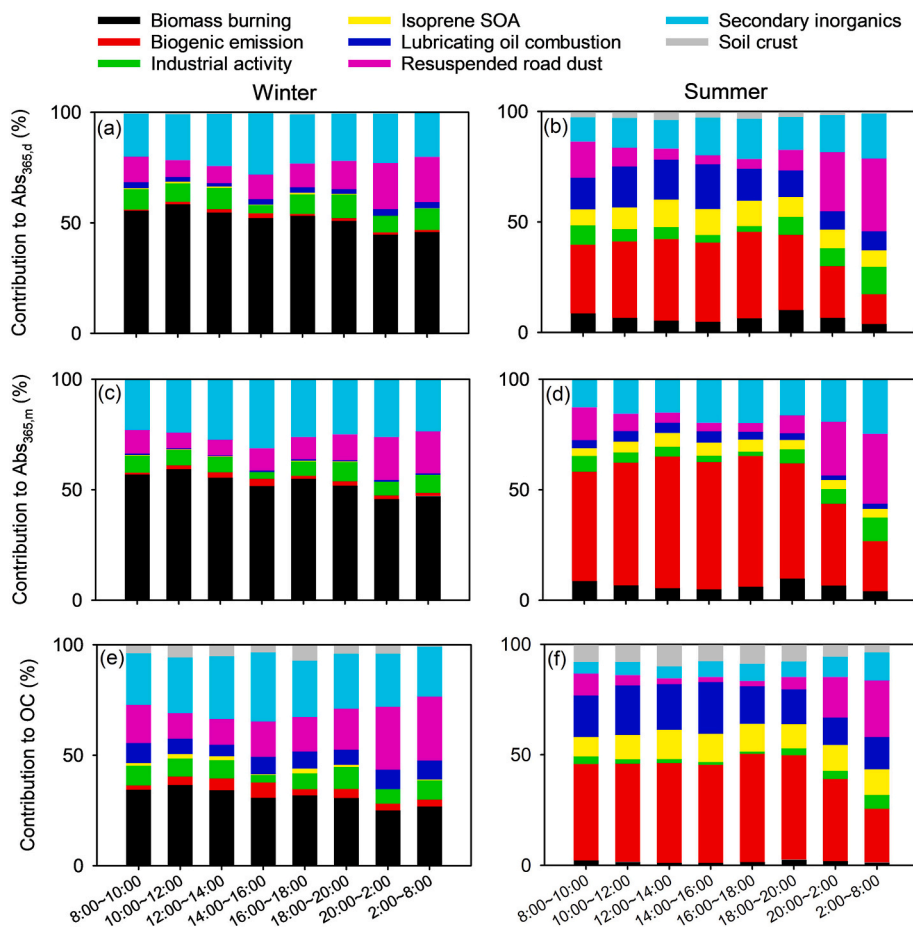


Fig. 5. Diurnal distributions of relative absolute factor contributions to (a, b) $Abs_{365,d}$, (c, d) $Abs_{365,m}$, and (e, f) OC for time-resolved $PM_{2.5}$ samples during winter and summer for northern Nanjing.

decrease after 20:00. The secondary inorganics factor accounted for the second largest fraction of $Abs_{365,d}$ (19.5%–27.6%) and $Abs_{365,m}$ (22.9%–31.2%) between 8:00 and 20:00, peaking at 14:00–16:00 (Fig. 5a, c), which may be related to the formation of secondary BrC through photooxidation and aqueous-phase reactions. Only the resuspended road dust factor showed a marked increase after 20:00, when its contributions became comparable to those of the secondary inorganics factor. Consistent with the results for integrated samples, the biomass burning, industrial activity, lubricating oil combustion, and resuspended road dust factors had higher contributions to $Abs_{365,d}$ than to $Abs_{365,m}$, with a weighted average difference greater than 0.15 Mm^{-1} , supporting that methanol-insoluble ELVOC may originate from combustion and resuspended road dust sources in winter.

In summer, the factor contributions of isoprene SOA (0.26–0.38 Mm^{-1} , 7.21%–12.5%) and lubricating oil combustion (0.28–0.60 Mm^{-1} , 8.26%–20.1%) to $Abs_{365,d}$ became significant (Figs. S4b and 5b) and exceeded their contributions to $Abs_{365,m}$ during all daytime sampling intervals (Figs. S4d and 5d). Due to differences in diurnal variations between $Abs_{365,d}$ and $Abs_{365,m}$, significantly higher fractions ($p < 0.01$) of $Abs_{365,m}$ (22.7%–59.5%) were apportioned to the biogenic emission factor compared to fractions of $Abs_{365,d}$ (13.5%–39.0%; Fig. 5b, d). Considering that the PMF model resolves factor profiles and contributions based on differences in temporal variations between input species (Dall'Osto et al., 2013), and the time series of DEOC absorption reproduced total BrC absorption better than that of MEOC absorption because of the higher solubility of ELVOC in DMF, using measured $Abs_{365,d}$ instead of $Abs_{365,m}$ for source apportionment can yield less biased results.

Fig. S5 shows the diurnal variations in the differences between mean contributions to $Abs_{365,d}$ and $Abs_{365,m}$ for individual factors with higher contributions to $Abs_{365,d}$. The differences between mean contributions to $Abs_{365,d}$ and $Abs_{365,m}$ for the resuspended road dust factor exhibited a diurnal pattern similar to that of mean $Abs_{365,d}$ minus mean $Abs_{365,m}$ (Fig. 3a and b) in both winter and summer, with noticeable values in the morning and at night. Besides photooxidation and aqueous-phase reactions, high MW BrC can also form through polymerization of organics in dust aerosols (Link et al., 2020; Ling et al., 2020). The lubricating oil combustion and industrial activity factors also had higher contributions to $Abs_{365,d}$ than $Abs_{365,m}$ in the morning and at night for winter and summer, respectively. Therefore, resuspended road dust mixed with anthropogenic combustion emissions and associated aging processes might be responsible for the differences in diurnal variations between measured $Abs_{365,d}$ and $Abs_{365,m}$.

4. Conclusions

In this study, the light absorption of DMF- and methanol-extractable OC was measured for integrated paired $PM_{2.5}$ and PM_{10} samples, as well as time-resolved $PM_{2.5}$ samples, in northern Nanjing. Comparison of BrC absorption between $PM_{2.5}$ and PM_{10} shows that nearly all light-absorbing chromophores were enriched in fine particles, regardless of whether DMF or methanol was used as the extraction solvent. Because DMF can extract more ELVOC from ambient particles than methanol, the mean $Abs_{365,d}$ values for $PM_{2.5}$ and PM_{10} were significantly higher than their mean $Abs_{365,m}$ values. This also applies to at least the central area of the YRD region in China. In both winter and summer, differences in diurnal variations between $Abs_{365,d}$ and $Abs_{365,m}$ for $PM_{2.5}$ samples were mainly observed in the morning and at night, likely due to the lower solubility of freshly emitted ELVOC, and secondarily formed ELVOC at night or primary ELVOC that experienced nighttime oxidation in methanol, respectively. OMM-based source apportionment results showed that the light-absorbing ELVOC soluble in DMF but not in methanol is associated with combustion sources and is released as direct emissions or through road dust resuspension in winter, while secondarily formed ELVOC is an additional potential source in summer. The diurnal variations in source contribution distributions suggest that

resuspended road dust mixed with combustion emissions and related aging processes may cause the different diurnal patterns of $Abs_{365,d}$ and $Abs_{365,m}$.

CRedit authorship contribution statement

Yuanyuan Zhang: Writing – original draft, Investigation, Data curation. **Furong Zhao:** Investigation, Data curation. **Chunyu Huang:** Visualization, Investigation. **Zhijuan Shao:** Writing – review & editing. **Wei Feng:** Validation, Investigation. **Hong Liao:** Writing – review & editing, Resources. **Yuhang Wang:** Writing – review & editing, Resources. **Guofeng Shen:** Writing – review & editing. **Mingjie Xie:** Writing – review & editing, Supervision, Project administration, Methodology, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.envpol.2026.128071>.

Data availability

Data will be made available on request.

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