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Characterization and source apportionment for light absorption amplification of black carbon at an urban site in eastern China



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HIGHLIGHTS

- MAE of BC was amplified by internal mixing and non-absorbing coating in eastern China.
- A statistical model was established between coating materials and MAE amplification rate.
- Nitrate and organic matter coating had the strongest effect on MAE amplification.
- The effects of organic matter and nitrate coatings increased with the internal mixing index.
- A useful toolkit for source apportionment of BC MAE amplification was proposed.

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ABSTRACT

The mass absorption efficiency (MAE) of black carbon (BC) could be amplified by both internal mixing and the lensing effect from non-absorbing coating, which could intensify the global warming effect of BC. In this study, a two-year-long continuous campaign with measurements of aerosol optical properties and chemical composition were conducted in Nanjing, a typical polluted city in the Yangtze River Delta (YRD) region. Relatively large MAE values were observed in 2016, and the high BC internal mixing level could be the main cause. The strong positive correlation between the ratio of non-absorbing particulate matter (NAPM) over elemental carbon (EC) and the MAE value indicated that the coating thickness of BC largely promotes its light absorption ability. The impacts of chemical component coating on MAE amplification in autumn and winter were greater than in other seasons. Multiple linear regression was performed to estimate the MAE amplification effect by internal mixing and the coating of different chemical components. Nitrate coating had the strongest impact on MAE amplification, followed by organic matter. The effects of organic matter and nitrate coatings on MAE amplification increased with the internal mixing index (IMI). Based on the positive matrix factorization (PMF) model, it was found that large decrease in the contribution of industrial emissions and coal combustion to $PM_{2.5}$ from 2016 to 2017 was the main cause for MAE reduction. The novel statistical model developed in this study could be a useful tool to separate the impacts of internal mixing and non-absorbing coating.

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1. Introduction

Black carbon (BC) in ambient aerosols exerts pivotal influence on earth's radiation balance and global climate change through light absorption (Bond and Bergstrom, 2006; Jacobson, 2001). Previous studies have estimated that the contribution of BC to global warming is only next to carbon dioxide (Fan et al., 2015; Reinman, 2012; Stocker et al., 2013). BC is measured based on the change in optical attenuation through the filter substrate and aerosol samples (Bond and Bergstrom, 2006; Chow et al., 2001), and the term BC is commonly used in climate models and emission inventories (Owen and Steiner, 2012; Paliwal et al., 2016). Elemental carbon (EC) is the refractory carbon content in aerosols, and usually considered as the massbased proxy for BC when evaluating the mass absorption efficiency (MAE) (Chen et al., 2017a; Cheng and He, 2017; Zhao et al., 2017). EC is determined by thermal-optical technology based on the thermal stability of carbonaceous species at elevated temperatures, and the term EC is commonly used in chemical characteristics of ambient aerosols and receptor models for source apportionment (Hopke et al., 2006; Huang et al., 2014; Watson et al., 2015).

MAE is a critical optical parameter in evaluating the light absorption capability of BC, determined from the light absorption coefficient divided by the EC concentration. MAE typically exhibited distinct spatiotemporal variation. Our previous study showed that the MAE values varied from 3.4 to 19.1 m²/g at a suburban site in Nanjing (Chen et al., 2019). Similar situation was observed in northern India with MAE ranging from 8.3 to 11.1 m²/g at urban, rural and high-altitude sites (Ram and Sarin, 2009). Zanatta et al. (2016) conducted MAE measurements at five sites in Europe, and found MAE values vary from 4.55 to 26.2 m²/g. Aside from the impact of aerosol-filter interactions in the measurements of aerosol optical properties, MAE of individual BC particles could be enhanced by both internal mixing and the lensing effect from non-absorbing coating (Cheng et al., 2020; Jacobson, 2001; Liu et al., 2017; Weingartner et al., 2003).

Generally, an enhancement factor of 2 was used in theoretical model estimation of BC radiative forcing where BC is assumed to be internally mixed (Chung and Seinfeld, 2005; Jacobson, 2001). Wang et al. (2014) found that the enhancement of MAE due to internal mixing was 1.8 times in western China. However, other observation studies (Bond and Bergstrom, 2006; Cappa and Onasch, 2012; Lan et al., 2013) indicated that the factor used in models could be substantially overestimated. Through chamber experiments, Schnaiter (2005) discovered that MAE of internally mixed BC was about 0.8 to 1.1 times larger than that of externally mixed BC. Using the method of single particle soot photometer (SP²), Lan et al. (2013) showed that MAE of internally mixed BC was amplified by only 7 % in summer in southern China. Bergstrom et al. (2007) suggested that MAE enhancement by the mixing state varied in different regions and was largely dependent on coating materials. In addition to BC mixing state, the lensing effect by nonabsorbing coating materials is another important factor for MAE variation (Fierce et al., 2016). Bai et al. (2018) found that non-absorbing coatings could enhance MAE by a factor of 2 at both a polluted urban site and a remote site at the summit of Mt. Tai. Zhang et al. (2018), Cheng et al. (2011), and Chen et al. (2019) suggested that secondary aerosols contributed significantly to MAE enhancement in Paris, Beijing, and Nanjing, respectively. As part of organic aerosols, brown carbon (BrC) coating had lower impact on absorption amplification than purely-scattering coatings such as sulfate and nitrate (Cheng and He, 2017). Cheng et al. (2020) performed laboratory experiments to simulate the aging process of BC particles upon exposure to ammonium nitrate, and their results showed that nitrate coating significantly alters the optical properties of BC. Through intensive observations in the North China Plain in summer, Cui et al. (2016) found that sulfate was the main cause for enhanced BC absorption.

Most existing studies on MAE enhancement of BC particles were based on limited experimental data. Few studies have investigated the relationships between aerosol absorption and chemical coating under different BC mixing states or considered the influence of anthropogenic emissions on BC absorption amplification. In this study, we performed online measurements of aerosol chemical composition and light absorption for two full years in Nanjing, a mega-city in the Yangtze River Delta (YRD) region in eastern China with high level of BC emissions (Zhang et al., 2013). Intensive and complex anthropogenic source emissions make Nanjing a typical case to study the influencing factors for MAE amplification (Chen et al., 2017b; Chen et al., 2019). Statistical models were developed to quantify the contributions of individual chemical components to MAE amplification in different seasons and mixing states, and the contributions of anthropogenic sources to MAE amplification were further investigated. Yields from this study could improve the understanding of the link between complex pollution sources in urban area and aerosol radiative forcing.

2. Methodology

2.1. Monitoring site and study period

As shown in Fig. S1 in the Supplementary Information (SI), the Jiangsu Academy of Environmental Sciences site (JAES, 32.05°N, 118.74°E, 25 m a. s.l.) was a typical urban site in the western area of downtown Nanjing, surrounded by restaurants, business centers, highways, residential area, and schools (Li et al., 2015). With the prevailing east wind, the site was located at the downwind of downtown Nanjing with significant influence from local emissions. Compared with the suburban site in eastern Nanjing in our previous studies (Chen et al., 2017b; Chen et al., 2019), JAES was more affected by aged aerosols.

The observation period at JAES was from January 2016 to December 2017. Up to 16,100 h of valid aerosol optical properties and chemical compositions (inorganic ions and carbonaceous species) were available after screening.

2.2. Methods for online measurements

2.2.1. Measurements of PM2.5 and carbonaceous components

The PM_{2.5} concentration was measured by a continuous particulate matter monitoring instrument (TEOM 1405DF, Thermo Scientific, USA). The monitor was equipped with a Filter Dynamic Measurement System (FDMS) to correct the semi-volatile substances evaporated from PM_{2.5} measurement filter. The airborne particles were first inhaled into a PM₁₀ cyclone at a flow rate of 16.7 L/min, and PM_{2.5} and PM_{2.5-10} were then separated by a round-nozzle virtual impactor. PM_{2.5} was deposited onto the sampling filter as the inlet air passed through. The natural oscillating frequency of the tapered glass element changed, which was converted to mass changes by the vibration theory, and the PM_{2.5} concentration was further calculated based on the changes of the filter mass and the sampling volume.

Hourly averaged thermal organic carbon (OC) and EC concentrations in $PM_{2.5}$ were determined by a semi-continuous thermal-optical transmittance (TOT) carbon analyzer (Sunset, Model-4). The ambient aerosols were inhaled into a $PM_{2.5}$ cyclone for 40 min with a flow rate of 8 L/min, went through a parallel-plate diffusion denuder and got into the instrument. Details on the protocol and the temperature gradient can be referred to our previous study (Chen et al., 2017b).

2.2.2. Measurements of ions on particles

To investigate the impacts of inorganic component coatings on MAE, hourly averaged concentrations of water-soluble ions including NO_3^- , $SO_4^2^-$, Cl^- , NH_4^+ , K^+ , Mg^{2+} , Ca^{2+} and Na^+ were measured using an online Monitor for Aerosols and Gases in ambient Air (MARGA, Model ADI 2080). The sampling system and the analytical system were the two main components of MARGA. The sampling system contained one wet rotating denuder for gas sampling and one steam jet aerosol collector for $PM_{2.5}$ collecting, and the sampling flow rate was set to be 1 m³/h. After sampling, the $PM_{2.5}$ samples were analyzed using ion chromatography. The detection limits of NO_3^- , SO_4^{2-} , Cl^- , NH_4^+ , K^+ , Mg^{2+} , Ca^{2+} , and Na^+ were 0.016, 0.023, 0.011, 0.028, 0.025, 0.020, 0.024, and 0.028 µg/m³, respectively (estimated by three times the standard deviation of blank samples).

2.2.3. Measurements of BC by wavelength

To evaluate the BC mixing state, BC concentrations at different wavelengths were measured by a 7- λ aethalometer (Magee Scientific, Model AE31). The flow rate of the monitor was set to be 5 L/min to ensure the accuracy of PM_{2.5} cyclone. The detection limit for BC was 0.16–0.28 μ g/m³, estimated by three times the standard deviation of blank samples. Unlike the thermal technique for EC measurements, the concentration of BC was determined based on its optical properties. More details on the aethalometer can be found in Weingartner et al. (2003).

2.3. Determination of MAE

The MAE value was estimated based on the relationship between the optical attenuation (ATN) and the EC mass obtained from the Sunset analyzer. However, the aerosol-filter interactions including the shadowing and multiple scattering effects could cause large uncertainties in MAE determination. Therefore, two correction factors (*C* and *R*(*ATN*)) were applied to reduce the uncertainties, which was discussed in detail in our previous study (Chen et al., 2019). The MAE value was calculated using Eq. (1) in this study as follows:

$$MAE \left(m^2/g \right) = \frac{ATN}{ECs \cdot C \cdot R(ATN)} \times 10^3$$
⁽¹⁾

where *ATN* is the light attenuation; *ECs* is the EC mass loading on the filter; *C* is the multiple scatter correction factor; and *R*(*ATN*) is the shadowing effect correction factor. Details on the calculation of *ATN*, *MAE* and *R*(*ATN*) are summarized in Section S1 in the SI. Large uncertainties existed in MAE estimation due to inappropriate values of the two empirical factors in previous studies (Chen et al., 2019; Ram and Sarin, 2009; Shen et al., 2013). By estimating the uncertainties to be ± 7 % for *ATN*, ± 10 % for *ECs* and \pm 41 % for *C*, the propagated error in *MAE* was calculated to be ± 42 %. Details on the uncertainty analysis of MAE estimation are presented in Section S2 in the SI.

2.4. Evaluation of BC mixing state

BC is measured based on the attenuation of light by EC deposited on a filter membrane, and its concentration is highly influenced by additional absorption by the lensing effect from chemical material coatings and light absorbing aerosols such as brown carbon and coarse dust, as well as the mixing state of BC. Weingartner et al. (2003) reported that the absorption coefficient for pure EC particles at 880 nm was almost equal to those for EC particles mixed with (NH₄)₂SO₄ and coated EC particles, indicating that negligible enhancement of an aethalometer absorption at 880 nm was observed for mixed BC with chemical components relative to pure EC. As for brown carbon and coarse dust, existing studies have shown that they exhibit obvious light absorption properties only at the blue and ultraviolet (UV) wavelength of the solar spectrum (Bond and Bergstrom, 2006; Chen et al., 2019; Li et al., 2021). Therefore, the variation of BC at 880 nm was mainly due to the change of the mixing state, while the measurement of EC by Sunset was based on the thermal stability of carbonaceous constituents which was independent of the mixing state. The ratio of optical BC measured at 880 nm to thermal EC mass was thus used as an indicator of the mixing state (internal mixing index, IMI) in this study. A previous study (Jung et al., 2010) also applied this ratio as an indicator of the mixing state of BC to investigate the optical properties of long-range transported Asian dust and polluted aerosols from Northeast Asia. Higher IMI values imply larger proportions of internally mixed BC (Chen et al., 2019; Jung et al., 2010).

2.5. Estimation of secondary organic carbon

The concentration of secondary organic carbon (SOC) at JAES was estimated based on an EC-tracer method (Turpin and Huntzicker, 1991; Lim and Turpin, 2002).

$$[SOC] = [OC] - r_p \cdot [EC] \tag{2}$$

where [OC] and [EC] are the OC and EC concentrations, respectively; r_p is the ratio of primary OC to EC emissions (Castro et al., 1999). In this work, hourly OC and EC concentrations with a correlation coefficient larger than 0.95 were selected to calculate r_p , as suggested by our previous work (Chen et al., 2017a, 2017b).

2.6. Quantification of contributions of chemical components to MAE amplification

Previous studies examined the significance of correlation between MAE amplification and the coating thickness of different chemical components to qualitatively evaluate the contributions of the coating to MAE enhancement (Cui et al., 2016; Zhang et al., 2021). In this study, the absorption amplification effect was defined by a MAE amplification coefficient (φ) as a function of the proxies for individual chemical coating thickness of BC, following Eq. (3):

$$\varphi = \frac{\text{MAE}}{\text{MAE}_{0}}$$

$$= \left(\frac{[\text{OC}]}{[\text{EC}]}\right)^{\alpha_{1} \cdot IMI} \cdot \left(\frac{[\text{SO}_{4}^{2} -]}{[\text{EC}]}\right)^{\alpha_{2} \cdot IMI} \cdot \left(\frac{[\text{NO}_{3}^{-}]}{[\text{EC}]}\right)^{\alpha_{3} \cdot IMI} \cdot \left(\frac{[\text{NH}_{4}^{+}]}{[\text{EC}]}\right)^{\alpha_{4} \cdot IM}$$
(3)

where MAE is the measured mass absorption efficiency; MAE₀ is the background value of MAE. According to our previous study (Chen et al., 2022), when the particle size was near 78 nm, the MAE values of the three monitoring sites were all around 4.59 \pm 0.13 m²/g, which was close to the MAE of the pure BC measured by the aerosol filter dissolution-filtration (AFD) method (Cui et al., 2016). The result indicated that EC particle with size of 78 nm in Nanjing was mostly naked, thus the value 4.59 m²/ g was used as MAE₀ in this study. [OC]/[EC], [SO₄^{2–}]/[EC], [NO₃[–]]/[EC] and [NH₄⁺]/[EC] are the ratios of coating materials over EC by mass concentration, indicating the coating thickness of different chemical components, and α_1 , α_2 , α_3 and α_4 are the influencing coefficients by the coatings of OC, sulfate, nitrate and ammonium, respectively. Eq. (3) was further converted to Eq. (4) by taking the logarithm for multiple linear regression:

$$\begin{split} &\ln \varphi = \ \ln (\text{MAE}) - \ \ln (\text{MAE}_0) \\ &= \alpha_1 \cdot \textit{IMI} \cdot \ \ln \left(\frac{[\text{OC}]}{[\text{EC}]} \right) + \alpha_2 \cdot \textit{IMI} \cdot \ \ln \left(\frac{[\text{SO}_4^2 \]}{[\text{EC}]} \right) + \alpha_3 \cdot \textit{IMI} \\ &\quad \cdot \ \ln \left(\frac{[\text{NO}_3^- \]}{[\text{EC}]} \right) + \alpha_4 \cdot \textit{IMI} \cdot \ \ln \left(\frac{[\text{NH}_4^+]}{[\text{EC}]} \right) \end{split} \tag{4}$$

2.7. Source apportionment of PM_{2.5} with positive matrix factorization model

The potential sources and their respective contributions to the $PM_{2.5}$ mass concentration and its main chemical components were estimated. In total, over 15,000 aerosol samples were analyzed, and the mass concentrations of water-soluble ions and carbonaceous component of aerosols were used for source apportionment. The source contributions of primary particles were obtained using the positive matrix factorization (PMF) model, and those of secondary inorganic aerosol (SIA) and SOC were further determined based on estimates of the nitrogen oxides (NO_x), sulfur dioxide (SO₂) and volatile organic compounds (VOCs) emissions in a local inventory. In PMF, the concentrations of different aerosol chemical components and

their corresponding uncertainties were used as input files. We followed the methods described in the PMF manual and Chen et al. (2020) to calculate the chemical component uncertainties in the dataset. Criteria including the optimum number of factors and the minimization of an objective function Q were determined based on principles described in Tan et al. (2016) and applied to the model to obtain the best solution. More details on the PMF model were described in our previous study (Chen et al., 2020).

2.8. Potential source contribution function (PSCF) model

The potential source contribution function (PSCF) method has been widely used for the identification of potential source regions of air pollution (Ren et al., 2021; Zhang et al., 2022). Based on results of the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model, backward trajectories were matched with BC concentrations at the corresponding time (Tian et al., 2022). In this study, the concentration threshold was set to be the 75th percentile of observed concentration according to previous studies (Jeong et al., 2017; Tian et al., 2022). The gridded *PSCF* values were obtained by comparing the number of polluted trajectory and the number of trajectory endpoints passing through the same grid, following Eq. (5).

$$PSCF_{ij} = \frac{m_{ij}}{n_{ii}} \tag{5}$$

where m_{ij} and n_{ij} are the number of polluted trajectory and the number of trajectory endpoints in the $i \times j$ grid, respectively. To reduce the uncertainty caused by grids with few trajectory endpoints, a weight function (W_{ij}) was applied to the estimation of the weighted *PSCF* (*WPSCF*), the calculation formula is as follows:

$$WPSCF_{ij} = \frac{m_{ij}}{n_{ij}} \times W_{ij} \tag{6}$$

Details on the calculation of W_{ij} can be referred to previous studies (Ren et al., 2021; Tian et al., 2022).

3. Results and discussion

3.1. Seasonal and interannual variations of EC mass concentration

As shown in Fig. 1, similar seasonal pattern of EC concentration (higher in winter and lower in summer) was found in 2016 and 2017. The EC concentration at JAES in summer was over 2 times that in Xuzhou (Liu et al., 2019), while in winter was 2.5 and 6.3 times that in urban Beijing and



Fig. 1. Seasonal variations of EC concentration at JAES in 2016 and 2017. Error bars denote the standard deviation.

that in the Taihang mountain (Ding and Liu, 2022), respectively. These results indicated that Nanjing was in a region with high loadings of BC pollution. Seasonal variation of EC concentration was mainly due to a combination of variations in anthropogenic emission and meteorological condition. According to the Multi-resolution Emission Inventory for China (MEIC, http://www.meicmodel.org/) in 2016 and 2017, BC emissions of Jiangsu Province in winter of 2016 and 2017 were 55 % and 49 % higher than those in summer, respectively. The change of residential emission was the dominant factor for the seasonal variation of BC, which accounted for 45 %–48 % of the total BC emissions in the two winters. In addition, the high EC concentration in the wintertime was partially due to pollutant accumulation caused by air stagnation (Chen et al., 2017b).

The annual average EC concentration in 2017 (2.2 \pm 1.1 µg/m³) was 21 % lower than that in 2016 (2.8 \pm 1.4 µg/m³). The decreased EC concentration in 2017 was caused by the reduction of local anthropogenic emissions and the change in regional air mass transport. Based on the bottom-up inventory model, the annual emission of BC was estimated to decrease by 15 % in Nanjing from 2016 to 2017 (Zhao et al., 2021). Table S1 provides the emissions of BC and the relative changes from 2016 to 2017 by source category. Among all the BC emission sources, the largest reduction of BC emissions occurred in biomass burning (-61 %). As shown in Fig. 1, the EC concentration in winter declined from 3.8 µg/m³ in 2016 to 2.7 µg/m³ in 2017, and in autumn from 2.4 µg/m³ in 2016 to 1.7 µg/m³ in 2017. The change in regional pollution transport was an important factor for the large difference in EC concentrations in autumn and winter of the two years.

In terms of PSCF distribution of EC in winter, higher PSCF values were found in the regions of central and western Henan, eastern Hubei and northern Hunan in 2016 than in 2017 (Fig. S2). Based on the spatial distribution of BC emissions by sector (Fig. S3), high EC emissions from residential coal combustion were identified in the above-mentioned regions. High emissions from industrial sectors and transportation in Henan also contributed significantly to the high EC concentration at JAES in the winter of 2016. Based on comparison between the PSCF distribution (Fig. S2) and the spatial distribution of emission inventories for autumn (Fig. S4), residential, industrial and transportation BC emissions from eastern Hubei and western Shandong were identified as the major sources in 2016 compared with the case in 2017.

3.2. Variation of MAE and its driving factors

Seasonal variations of MAE at JAES during the two years are presented in Fig. 2a. In 2016, relatively high MAE values were found in autumn (8.6 \pm 2.3 m²/g) and winter (9.0 \pm 2.8 m²/g), and the lowest MAE value was found in summer (7.9 \pm 2.1 m²/g). Less significant seasonal variation of MAE was found in 2017. In general, the MAE values of each season in 2016 were higher than those in 2017, and the average annual MAE in 2016 was 6.3 % higher than that in 2017.

The seasonal variation of MAE during the two years was influenced by both internal mixing and the lensing effects of aerosol coating materials. The coating thickness can be indicated by the ratio of non-absorption particulate matter (NAPM) (i.e., the sum of OC, sulfate, nitrate, and ammonium) over EC (Bai et al., 2018; Cui et al., 2016; Zhang et al., 2021). As shown in Fig. 2b, the NAPM/EC ratios for the two years exhibited a similar seasonal variation with low values in summer and high in winter. Similar result was found in Ding et al. (2021) that the bulk coating thickness of BC in winter under more influence from anthropogenic sources was significantly higher than in summer. The seasonal variation of NAPM/EC was consistent with MAE, indicating that the non-absorption coating materials probably had an important impact on the seasonal variation of MAE. Except for spring, the ratios of NO_3^- /EC and OC/EC in 2017 were higher than those in 2016, which was probably due to the significant decrease in precipitation from 2016 to 2017. According to historical amounts of precipitation in Nanjing (data source: http://js.cma.gov.cn/), the precipitation in 2017 decreased by 33 % compared with 2016, especially in spring (40 %), autumn (49 %) and winter (39 %) (Fig. S5). Previous studies found that



Fig. 2. Seasonal variations of (a) MAE and (b) the proxy for coating thickness of EC, defined as the mass ratio of major aerosol components (OC, sulfate, nitrate and ammonium) to EC at JAES from 2016 to 2017.

the average particle scavenging contributions to total wet deposition were estimated to be 28 % for NO₃⁻, 63 % for SO₄⁻⁻ and 70 % for NH₄⁺ (Cheng and He, 2017) while only 0.13 %–6.68 % for EC (Xu, 2014). Less precipitation in 2017 may have limited the scavenging effects for the main chemical species in PM_{2.5} (such as nitrate, sulfate, and OC) and thereby resulted in the higher ratio of NAPM/EC. On the contrary to the annual variation of NAPM/EC, the MAE values for all seasons of 2016 were higher than those in 2017 (Fig. 2a), implying that the BC mixing state was probably a more important factor for the annual MAE variation. According to Fig. S6, the average IMI in 2016 was 8.6 % greater than that in 2017, and significant differences were found in autumn and winter of the two years. For a certain region, the temporal variation of MAE was largely affected by the internal mixing level of BC.

Fig. S7 shows that the MAE value and the NAPM/EC ratio varied dramatically depending on seasons and cities. Poor correlation between MAE and NAPM/EC was found among all the cities ($R^2 = 0.06$), and two outliers were identified (Guangzhou and Ispra). The MAE value in Guangzhou was obtained from absorption efficiency factor which was estimated by a linear regression of BC concentrations at 880 nm from the aethalometer against the aerosol absorption coefficients at 532 nm from a photoacoustic spectrometer, which was different from the estimation method by light attenuation in other studies (Tao et al., 2021). As for Ispra, previous study showed that OC accounted for >62 % of PM_{2.5} (Fig. S8) (Zanatta et al., 2016), which was quite different from the cases in most Chinese cities (e.g., Beijing, Shanghai, Nanjing, Wuxi, Ningbo and Jinan) that sulfate, nitrate, and ammonium dominated the PM_{2.5} concentration (Chen et al., 2018; Du et al., 2017; Zhang et al., 2016). Except for Guangzhou and Ispra, a strong correlation between MAE and NAPM/EC was found ($R^2 = 0.62$) (Fig. S7) indicating that the coating thickness of BC largely affects its light absorption ability.

3.3. Effect of chemical coatings on the MAE amplification

3.3.1. Seasonal contributions of chemical coatings to MAE amplification

The influencing coefficients for each chemical component by season are summarized in Table 1. Good correlations were found between the observed and calculated MAE values for all seasons in the two years ($R^2 > 0.85$), indicating that the fitting function had a good performance.

According to Table 1, the seasonal MAE amplification factors (φ) ranged from 1.59 to 1.82 during the two years. Combined with the mass concentration of the chemical components, the contributions of different chemical components to MAE amplification effect $(\ln(\varphi))$ in different seasons are shown in Fig. 3. The impacts of the chemical component coating on MAE amplification in autumn and winter were greater than those in spring and summer. Among all the chemical components, nitrate coating had the strongest effect on the MAE amplification, followed by OC. Tao et al. (2021) also found that the lensing effect by nitrate and organic matter coatings were the main causes for MAE enhancement in Guangzhou. On the contrary, the influence of sulfate coating on MAE amplification in spring and summer were stronger than in autumn and winter. Cui et al. (2016) found that sulfate coating was an important factor for the enhancement of MAE in Shandong during the summertime. This could be related to the high frequency of new particle formation (NPF) occurrence in spring and summer with sulfuric acid being the key contributor to NPF (Huo et al., 2019). Different from the other chemical components, the influencing coefficients for ammonium on MAE amplification in all seasons were negative,

Table 1

Summary o	of the	infl	uencing	coefficie	nts fo	r in	ndividual	chemical	com	ponents	on	MAE	am	plification	by	season.
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Year	Season	arphi	IMI	OC	SO_4^{2-}	NO ₃	NH_4^+	R^2
				α_1	α ₂	α ₃	$\overline{\alpha_4}$	
2016	Spring	1.67 ± 0.72	1.18 ± 0.41	0.06 ± 0.02	0.08 ± 0.02	0.33 ± 0.05	-0.37 ± 0.07	0.89
	Summer	1.64 ± 0.71	1.08 ± 0.45	0.11 ± 0.03	0.50 ± 0.11	0.12 ± 0.03	-0.65 ± 0.12	0.93
	Autumn	1.71 ± 0.73	1.26 ± 0.48	0.29 ± 0.06	0.01 ± 0.00	0.27 ± 0.05	-0.47 ± 0.09	0.95
	Winter	1.82 ± 0.81	1.14 ± 0.46	0.35 ± 0.05	0.01 ± 0.00	0.31 ± 0.06	-0.45 ± 0.10	0.96
	All year	1.70 ± 0.75	1.15 ± 0.44	0.20 ± 0.03	0.15 ± 0.05	0.25 ± 0.04	-0.46 ± 0.12	0.90
2017	Spring	1.61 ± 0.49	1.08 ± 0.41	0.22 ± 0.04	0.17 ± 0.03	0.29 ± 0.05	-0.62 ± 0.13	0.87
	Summer	1.59 ± 0.76	0.95 ± 0.48	0.08 ± 0.02	0.27 ± 0.05	0.30 ± 0.06	-0.49 ± 0.09	0.95
	Autumn	1.68 ± 0.73	0.96 ± 0.52	0.17 ± 0.03	0.02 ± 0.00	0.41 ± 0.06	-0.49 ± 0.08	0.92
	Winter	1.66 ± 0.72	1.05 ± 0.41	0.18 ± 0.03	0.05 ± 0.01	0.42 ± 0.07	-0.53 ± 0.11	0.94
	All year	1.64 ± 0.70	1.03 ± 0.46	0.16 ± 0.02	0.11 ± 0.02	0.36 ± 0.07	-0.52 ± 0.09	0.90

Notes: the data represent means with corresponding standard deviations, and all the p values for the coefficients were <0.01.

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Fig. 3. The contributions of individual chemical component to $\ln(\varphi)$ in different seasons.

indicating that ammonium coating had a reduction effect on MAE. The comparison of the two years showed that the amplification effect of MAE became weaker from 2016 to 2017, especially in winter. The effects of OC and sulfate coatings decreased, while that of nitrate coating increased significantly.

3.3.2. Contributions of chemical coatings under different BC mixing states

As presented in Fig. S9, the IMI values were mainly distributed in ranges of 0–1.0, 1.0–1.5, 1.5–2.0, 2.0–2.5 and 2.5–4.8, the sum of these intervals accounted for 99 % of the total samples. The five ranges of IMI were thus used to investigate the absorption amplification of individual chemical coatings on MAE amplification under different internal mixing levels. As shown in Table 2, the fitting function had a good performance that the R^2 values for the correlation between the observed and the calculated MAE values under different IMI levels were all above 0.76. Under different IMI levels, the MAE amplification factors ranged from 1.50 to 4.58. With the increase of IMI levels, the effect of MAE amplification showed a significant upward trend.

Combined with the mass concentration of chemical components, the contribution of individual chemical components to MAE amplification effect $(\ln(\varphi))$ under different internal mixing levels is shown in Fig. 4. The effects of OC and nitrate coatings on MAE amplification increased with the increase of IMI, and the contribution of OC and nitrate to $\ln(\varphi)$ at high IMI level (2.5–4.8) was 5.6 and 1.7 times those at low IMI level (0–1), respectively. However, the effect of sulfate coating showed the opposite trend, with the coefficient decreased by 83 % from low to high IMI level. Ammonium coating had a negative effect which was weakened with the increase of IMI.

3.4. Impact of anthropogenic emissions on MAE amplification

To explore the causes of MAE amplification, source apportionment of BC absorption amplification was conducted for different seasons and internal mixing levels by the PMF model, as described in Section 2.6. The PMF model was used to identify the potential sources and quantify their respective contributions to the mass concentration of each chemical species. The summary of the source profiles and their contributions to chemical



Fig. 4. The contributions of individual chemical components to $\ln(\varphi)$ under different internal mixing levels.

components at JAES in 2016 and 2017 are presented in Fig. S10 and Table 3. The main sources identified in this study included coal combustion, industrial pollution, transportation, biomass burning, fugitive dust and secondary sources, and the total contribution of above sources was >70 % in the two years. From 2016 to 2017, the contributions of industrial emissions, coal burning and dust sources decreased significantly, from 33.8 %, 2.3 % and 16.1 % to 22.0 %, 0.7 % and 12.2 %, respectively. The contributions of secondary sources and biomass burning increased from 29.2 %and 7.5 % to 39.8 % and 13.8 %, respectively. Secondary sources contributed significantly to nitrate, sulfate and ammonium. Based on the emission inventory of precursors of SOC (VOCs) and SIA (NOx SO2 and NH3) in Nanjing in 2016 and 2017 (Zhao et al., 2021), the source apportionment for secondary aerosols at JAES were further estimated (Table 3). After reapportionment, the contributions of industrial pollution, transportation and coal combustion increased significantly. The contributions of industrial pollution to primary and secondary PM2.5 decreased from 2016 to 2017, whereas the contributions of transportation and biomass burning increased by 3.7 % and 4.4 %, respectively.

Contributions of emission sources to main chemical components from 2016 to 2017 are shown in Fig. 5. The contributions of industrial emission and coal combustion decreased, while that of transportation increased. Combined with the contributions of chemical species to the MAE amplification (Fig. 3), the result indicated that the decreased in the contributions of industrial emission and coal combustion to the main chemical components in PM_{2.5} from 2016 to 2017 was the main reason for the MAE reduction, while the increase in vehicle emissions was a critical factor that MAE did not show a large decline in 2017.

According to Section 3.3.2, five internal mixing levels with IMI values in the range of 0–4.8 were used to analyze the variation of contributions of anthropogenic emissions to the main chemical components under different internal mixing levels. As shown in Fig. S11, with the increase of BC internal

Table 2

The influencing coefficients of individual chemical components on MAE amplification at different internal mixing levels (IMI).

IMI	arphi	IMI	OC	SO_4^{2-}	NO ₃	NH ₄ ⁺	R^2
			α_1	α_2	α_3	α ₄	
0.0-1.0	1.50 ± 0.21	0.83 ± 0.11	0.06 ± 0.02	0.12 ± 0.03	0.09 ± 0.02	-0.21 ± 0.04	0.76
1.0 - 1.5	1.87 ± 0.84	1.23 ± 0.24	0.15 ± 0.04	0.10 ± 0.03	0.10 ± 0.02	-0.16 ± 0.03	0.81
1.5 - 2.0	2.41 ± 0.39	1.71 ± 0.34	0.18 ± 0.04	0.07 ± 0.02	0.11 ± 0.02	-0.14 ± 0.03	0.86
2.0 - 2.5	3.01 ± 1.62	2.21 ± 0.84	0.29 ± 0.05	0.02 ± 0.00	0.15 ± 0.03	-0.08 ± 0.02	0.88
2.5-4.8	4.58 ± 2.02	3.60 ± 1.17	0.35 ± 0.08	0.02 ± 0.00	0.17 ± 0.04	-0.01 ± 0.00	0.83

Notes: the data represent means with corresponding standard deviations, and all the p values for the coefficients were <0.01.

Table 3

Source apportionments of $\text{PM}_{2.5}$ in Nanjing in 2016 and 2017 (Unit: %).

Year	Source	Primary apportionment	Source	SIA reapportionment	SOC reapportionment
2016	Secondary source	29.2	Industrial pollution	30.0	90.0
			Transportation	23.7	7.9
			Coal combustion	38.3	0.7
			Biomass burning	0.8	1.4
			Agriculture	7.2	0.0
	Industrial pollution	33.8	-	-	-
	Transportation	10.3	-	-	-
	Coal combustion	2.3	-	-	-
	Biomass burning	7.5	-	-	-
	Fugitive dust	16.1	-	-	-
	Others	0.8	-	-	-
2017	Secondary source	39.8	Industrial pollution	25.4	89.2
			Transportation	33.3	9.9
			Coal combustion	33.4	0.4
			Biomass burning	0.8	0.6
			Agriculture	7.2	0.0
	Industrial pollution	22.0	-	-	-
	Transportation	10.1	-	-	-
	Coal combustion	0.7	-	-	-
	Biomass burning	13.8	-	-	-
	Fugitive dust	12.2	-	-	-
	Others	1.5	-	-	-

mixing level, the contributions of industrial emissions to OC and sulfate increased largely, with the maximum increase by 21 % and 7 %, respectively. The contributions of industrial emissions and coal combustion to nitrate increased, with the maximum increase of 3.4 % and 3.5 %, respectively. It is worth noting that the contributions of transportation to the main chemical components of $PM_{2.5}$ (OC, sulfate, nitrate, ammonium) all decreased with the increase of internal mixing level (Fig. S11). The result indicated that industrial emissions and coal combustion enhanced the MAE amplification when IMI increased.

4. Implications and limitations

Based on results from all the methods adopted in this study, sources for light absorption amplification of BC were identified for different seasons in 2016 and 2017. High PSCF values were mainly found on the junction of Anhui, Jiangxi and Zhejiang in spring 2016 (Fig. S2). This region was related to high loads of coal combustion and industrial emissions. Their large contributions to nitrate promoted MAE amplification. In spring 2017, the region with high PSCF values moved towards the junction of Anhui, Jiangsu and Zhejiang with high loadings of agricultural and industrial emissions, contributing significantly to OC, ammonium and nitrate.



Fig. 5. The contributions of anthropogenic emissions to the main chemical components of aerosols from 2016 to 2017.

The negative contribution of ammonium to MAE enhancement led to lower MAE values compared to spring 2016. In the summer of 2016, high PSCF values were found in the central YRD region. Industrial emissions in this region had great contribution to nitrate and sulfate. Higher atmospheric oxidizing capacity and higher contribution of clean air masses from the marine boundary layer accelerated the NPF process, resulting in the prominent contribution of sulfate to MAE amplification. In summer 2017, high PSCF values were found in the region of southern Anhui. The industrial emissions from this region made nitrate a priority. In autumn 2016, high PSCF values were found in the junction of Anhui, Jiangxi and Hubei. With lower negative contribution from ammonium, industrial emissions and biomass burning promoted MAE significantly by contributing more OC and nitrate. In autumn 2017, the industrial pollution from southern Anhui took the lead in contributing OC and nitrate for MAE enhancement. Regional transmission ranges of the two winters were more extensive, the high PSCF values of the two winters were similarly located in the junction of North China Plain, Central China and YRD. MAE amplification in the winter of 2016 was highly affected by industrial emissions from southern Henan and central Anhui and transportation in Nanjing. Local NO_x emissions in urban Nanjing decreased significantly in the winter of 2017 compared with the previous year, leading to a decrease in nitrate and reduced MAE enhancement.

The integrated methods used in this study have provided a useful case study of source apportionment for light absorption amplification of BC. Nevertheless, limitations in this study should be pointed out. The PMF model for source apportionment used in this study still has large uncertainties. The multiple linear regression model (Eq. (4)) established in this study needs verification based on data from other observation sites. In addition, the mechanism for the negative effect of ammonium on MAE amplification unveiled in this work needs to be further explored.

5. Conclusions

A comprehensive study of BC light absorption was conducted at an urban site in a typical polluted city in YRD from January 2016 to December 2017. Relatively low EC concentration was observed in 2017, attributed mainly to the improved BC emission control in Nanjing and variation of regional air mass transport. Compared with MAE in 2017, higher MAE in 2016 was mainly attributed to the high BC internal mixing level. By analyzing the MAEs and NAPM/EC in Nanjing and other cities, it was found that MAE had a strong positive correlation with NAPM/EC. These results indicated that BC internal mixing level was crucial to the seasonal MAE variation in one city. For spatial variation of MAE in cities of China, it is

reasonable to assess MAE ranges by NAPM/EC ratios. MAE amplification by individual chemical components was further estimated by model fitting. Results showed that nitrate coating had a strong positive effect on MAE amplification, followed by organic matter. The effects of organic matter and nitrate coatings on MAE amplification increased with the increase of IMI levels. Ammonium coating had a negative effect on MAE amplification at all IMI levels, and the effect gradually weakened with the increase of IMI. Combined with the source apportionment by the PMF model, it was found that the fact that MAE amplification in 2017 was weaker than in 2016 was mainly due to the significant decrease in the contributions of industrial emission and coal combustion to $PM_{2.5}$ from 2016 to 2017. High internal mixing levels could enhance the MAE amplification effects by nitrate and OC from industrial and combustion sources.

CRediT authorship contribution statement

Dong Chen: Methodology, Investigation, Data curation, Software, Writing – original draft. Wenxin Zhao: Methodology, Investigation, Data curation, Software. Lei Zhang: Conceptualization, Methodology, Resources, Writing – review & editing. Qiuyue Zhao: Resources, Funding acquisition, Writing – review & editing. Jie Zhang: Resources, Data curation. Feng Chen: Resources, Data curation. Huipeng Li: Resources, Data curation. Miao Guan: Resources, Data curation. Yu Zhao: Writing – review & editing.

Data availability

Data will be made available on request.

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

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