



Amplification of light absorption of black carbon associated with air pollution

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Abstract. The impacts of black carbon (BC) aerosols on air quality, boundary layer dynamics and climate depend not only on the BC mass concentration but also on the light absorption capability of BC. It is well known that the light absorption capability of BC depends on the amount of coating materials (namely other species that condense and coagulate on BC). However, the difference of light absorption capability of ambient BC-containing particles under different air pollution conditions (e.g., clean and polluted conditions) remains unclear due to the complex aging process of BC in the atmosphere. In this work, we investigated the evolution of light absorption capability for BC-containing particles with changing pollution levels in urban Beijing, China. During the campaign period (17 to 30 November 2014), with an increase in PM₁ concentration from ~ 10 to $\sim 230 \mu\text{g m}^{-3}$, we found that the mass-weighted averages of the aging degree and theoretical light absorption capability of BC-containing particles increased by $\sim 33\%$ and $\sim 18\%$, respectively, indicating stronger light absorption capability of BC-containing particles under more polluted conditions due to more coating materials on the BC surface. By using an effective emission intensity (EEI) model, we further found that aging during regional transport plays an important role in the difference in the light absorption capability of BC-containing particles under different air pollution levels. During the pollution episode, $\sim 63\%$ of the BC over Beijing originated

from regional sources outside of Beijing. These regionally sourced BC-containing particles were characterized by more coating materials on BC surfaces due to more coating precursors within more polluted air, which contributed $\sim 75\%$ of the increase in theoretical light absorption capability of BC observed in Beijing during the polluted period (PM₁ of $\sim 230 \mu\text{g m}^{-3}$) compared to that in the clean period (PM₁ of $\sim 10 \mu\text{g m}^{-3}$). Due to the increase in theoretical light absorption capability of BC associated with air pollution, the direct radiative forcing of BC was estimated to be increased by $\sim 18\%$ based on a simple radiation transfer model. Our work identified an amplification of theoretical light absorption and direct radiative forcing under a more polluted air environment due to more coating materials on BC. The air pollution control measures may, however, break the amplification effect by reducing emissions of both BC and the coating precursors and achieve co-benefits of both air quality and climate.

1 Introduction

Black carbon (BC) is an important aerosol component that absorbs visible sunlight and contributes to heating of the atmosphere (Menon et al., 2002; Bond and Bergstrom, 2006; Gustafsson and Ramanathan, 2016). Atmospheric BC can

impact climate through radiative effects, which are strongly associated with the optical properties of BC, especially light absorption (Jacobson, 2000; Lesins et al., 2002; Cheng et al., 2006; Ramanathan and Carmichael, 2008). Estimating the climate effects of BC is one of the major challenges in climate change research, partly due to large uncertainties in the light absorption capability of BC-containing particles under ambient conditions (Cappa et al., 2012; Liu et al., 2015; Gustafsson and Ramanathan, 2016; Liu et al., 2017). The light absorption capability of atmospheric BC is complex and poorly quantified, and it changes with the morphology, density and mixing state of the BC-containing particles (Schnaiter et al., 2005; Zhang et al., 2008; Knox et al., 2009; Peng et al., 2016). Previous theoretical (Jacobson, 2001; Moffet et al., 2009; Zhang et al., 2016) and observational (Knox et al., 2009; Cappa et al., 2012; Peng et al., 2016) studies showed a broad range of absorption enhancements (1.05–3.05) of BC-containing particles during the atmospheric aging process. To date, conflicts remain between model- and observation-based studies of the light absorption capability of atmospheric BC-containing particles (Jacobson, 2001; Cappa et al., 2012; Liu et al., 2015; Liu et al., 2017).

The light absorption capability of BC-containing particles depends strongly on the particle mixing state (Liu et al., 2015; Liu et al., 2017), i.e., the degree of internal mixing between BC and other particle species (i.e., non-BC components) by the atmospheric aging process (i.e., condensation, coagulation and heterogeneous oxidation). The non-BC species (i.e., coating materials) on the surface of BC cores can enhance BC light absorption via the lensing effect (namely, the coating materials act as a lens to focus more photons on BC; Fuller et al., 1999; Jacobson, 2001; Bond et al., 2006; Lack and Cappa, 2010). In terms of individual BC-containing particles, more coating materials result in stronger light absorption capability. The coating materials on the BC surface are controlled by secondary processes (e.g., photochemical production) (Metcalf et al., 2013).

The production of secondary aerosols in the atmosphere varies significantly with pollution levels (Cheng, 2008; Yang et al., 2015; Zheng et al., 2016; Mu et al., 2018), indicating that BC-containing particles most likely exert different light absorption capability values under different pollution levels. Compared with clean air conditions, polluted periods feature more secondary aerosols, especially secondary inorganic species such as sulfate (Guo et al., 2014; Sun et al., 2014; Zheng et al., 2015). Whether the changes of secondary aerosols with air pollution will affect the coating materials on the BC is complex, and not only depends on the increase in BC vs. secondary aerosols amount but also controlled by secondary material condensation on BC- vs. non-BC-containing particles. Recent BC aging measurements in Beijing and Houston using an environmental chamber (flowing ambient air fed with lab-generated fresh BC) have revealed that a clear distinction in the light absorption capability of BC-containing particles exists between urban cities in

developed and developing countries (Peng et al., 2016), and this difference is likely due to the differences in air pollution levels.

To date, whether and how the aging degree and light absorption capability of BC-containing particles will change with air pollution development is still unclear. Although the enhancement of BC light absorption due to coating materials on BC surfaces has already been intensively investigated (Schnaiter et al., 2005; Moffet et al., 2009; Shiraiwa et al., 2010; Zhang et al., 2016), there are few studies on the evolution of the light absorption capability of BC-containing particles with changing air pollution levels. The variation in the light absorption capability of BC-containing particles associated with air pollution can lead to different effects of BC aerosols on air quality and climate under different pollution levels. To improve the evaluation of BC-related effects on air quality and climate, some models have considered BC internally mixed with other species (namely coating materials on BC surface), which can affect the light absorption capability of BC-containing particles. However, the difference of coating materials on BC under different air pollution conditions remains unclear.

In this work, we conducted an intensive field measurement campaign in urban Beijing, China, to investigate the difference of the theoretical light absorption capability of atmospheric BC-containing particles under different pollution levels. First, we analyzed the evolution of theoretical light absorption capability of BC with increasing air pollution levels and estimated the relationship between the changing rate in the theoretical light absorption capability of BC and that in the PM₁ or BC mass concentrations. We then explored the cause of the evolution of theoretical light absorption capability of BC with increasing air pollution levels and evaluated the relative importance of regional transport. Finally, we discussed the impact of changes in BC light absorption capability with pollution levels on BC radiative forcing.

2 Methods

2.1 Sampling site and measurements

The in situ measurements were conducted on the campus of Tsinghua University (Tsinghua site, 40°00′17″N, 116°19′34″E) from 17 to 30 November 2014. The Tsinghua site (Fig. S1 in the Supplement) is located in urban Beijing, China. The megacity Beijing is adjacent to Hebei Province and the megacity Tianjin (Fig. S1), in which considerable industrial manufacturing has led to heavy emissions of air pollution, especially in southern Hebei.

Ambient aerosol particles were collected by a PM₁ cyclone and then passed through a diffusion silica gel dryer, and they were finally analyzed using an Aethalometer (AE33, Magee Scientific Corporation), an aerosol chemical speciation monitor (ACSM, Aerodyne Research Inc.) and a single-particle soot photometer (SP2, Droplet Measurement Tech-

nologies Inc.). The AE33 can measure the absorption coefficient (σ_{ab}) of sampled aerosols in seven spectral regions (370, 470, 520, 590, 660, 880 and 950 nm). At a wavelength of 880 nm, the absorption of aerosol particles is dominated by the BC component because the light absorbed by other aerosol components is significantly less (Sandradewi et al., 2008; Drinovec et al., 2015). In this study, the σ_{ab} at 880 nm measured by the AE33 was used to characterize the theoretical light absorption of the BC-containing particles. More details on the AE33 measurement can be found in the work of Drinovec et al. (2015). Considering the filter-loading effect and multiple-scattering effect (Weingartner et al., 2003; Segura et al., 2014; Drinovec et al., 2015), the Aethalometer data were corrected by compensation factors described in the Supplement (Fig. S2 and the associated discussion). The ACSM and SP2 instruments measured the mass concentrations of non-refractory (NR) submicron-scale components (NR-PM₁, i.e., sulfate, nitrate, ammonium, chloride and organics) and refractory BC (rBC), respectively, and the sum of these two measurements was used to estimate the PM₁ mass concentration. The ACSM instrument used in our study was described by H. Li et al. (2017).

The SP2 instrument measures a single BC-containing particle using a 1064 nm Nd:YAG intra-cavity laser beam. As the light-absorbing rBC passes through the laser beam and is heated to its vaporization temperature (~ 4000 K), it will emit incandescent light (i.e., visible thermal radiation), which is linearly proportional to the mass of the rBC (Schwarz et al., 2006; Metcalf et al., 2012; Moteki and Kondo, 2010; Sedlacek et al., 2012). In this study, the calibration curve of rBC mass vs. incandescence signal was obtained from the incandescence signal of size-resolved Aquadag particles (their effective density obtained from Gysel et al., 2011) using a DMA (differential mobility analyzer) and SP2 measurement system. Considering the different sensitivity of the SP2 to different rBC types (Gysel et al., 2011; Laborde et al., 2012), we corrected the SP2 calibration curve by scaling the peak height of incandescence signal for Aquadag particles at each rBC mass based on the relationship between the sensitivity of SP2 to Aquadag and ambient rBC (Laborde et al., 2012). The particle-to-particle mass of ambient rBC can be determined by measuring its incandescence signal and comparing it to the calibration curve. The mass concentration of rBC is calculated from the particle-to-particle mass of rBC and the sampled flow (~ 0.12 L min⁻¹). Note that the SP2 detection efficiency (Fig. S3) has been considered in the calculation of rBC mass concentration. Furthermore, the scattering cross section of a BC-containing particle is obtained from its scattering signal using the leading-edge-only (LEO) fit method (Gao et al., 2007). Zhang et al. (2016) has demonstrated the validity of the LEO fit method for ambient BC-containing particles in China.

2.2 SP2 data analysis

2.2.1 Aging degree of BC-containing particles

Based on the rBC core mass (m_{rBC}) and scattering cross section (C_s) of the BC-containing particle derived from the SP2 measurements, the size of the BC-containing particle (D_p), including the rBC core and the coating materials, was calculated using Mie theory with a shell-and-core model (Zhang et al., 2016). In Mie calculation, the D_p is retrieved from C_s , the size of the rBC core (D_c) and the refractive indices of the non-BC shell (RI_s) and rBC core (RI_c).

The RI_s values used in this study are 1.50-0i based on the chemical compositions of coating materials during the campaign period (Fig. S4 and the associated discussion in the Supplement). In terms of RI_c , we evaluated the sensitivity of D_p values retrieved with Mie mode to the RI_c values (Fig. S5 and the associated discussion in the Supplement). In the following calculation, the RI_c of 2.26-1.26i was used (Taylor et al., 2015).

The D_c is calculated using the m_{rBC} and rBC core density (ρ_c , 1.8 g cm⁻³ used in this study; Cappa et al., 2012) assuming a void-free sphere for the rBC core, as given in Eq. (1). The size distribution of rBC cores under different pollution levels during the campaign period is displayed in Fig. S6.

$$D_c = \left(\frac{6m_{\text{rBC}}}{\pi \rho_c} \right)^{1/3} \quad (1)$$

As in previous studies (Sedlacek et al., 2012; Liu et al., 2013; Zhang et al., 2016), the aging degree of BC-containing particles was characterized by the D_p / D_c ratio in this study. A higher D_p / D_c ratio for BC-containing particles indicates a higher aging degree, i.e., more coating materials on the BC surface.

In terms of the D_p / D_c ratio of BC-containing particles, we focused on the rBC core size (D_c) above detection limit of SP2 incandescence ($D_c > 75$ nm), while the detection limit of SP2 scattering for the whole particle size (D_p) was not considered in this study. If the BC-containing particles with rBC cores larger than ~ 75 nm are large enough to be detected by the SP2 scattering channel, we would calculate their whole particle size (D_p) using the LEO method based on their scattering signal. If not, we would assume that the D_p was equal to the rBC core size (D_c). This assumption might lead to the underestimation of the D_p / D_c ratio of the BC-containing particles with a size above the incandescence limit and below the scattering limit. To evaluate the uncertainty of the D_p / D_c ratio, we calculated the detection efficiency of SP2 scattering (Fig. S7 in the Supplement). In terms of BC-containing particles with a rBC core larger than 75 nm (SP2 size cut for incandescence) observed in our site during the campaign period, most of them (~ 90 – 100 %, Fig. S7) exhibited a particle size (180–500 nm shown in Fig. S8) larger than SP2 size cut for scattering due to a large amount of coating materials on rBC cores. This indicated that the uncertainty of

the D_p / D_c ratio calculated in this study due to mismatch in the SP2 size cut for incandescence vs. scattering is no more than 10 %. High detection efficiency of SP2 scattering for BC-containing particles observed at our site can be attributed to their large size (180–500 nm, Fig. S8).

2.2.2 BC optical properties

Based on the size information on BC-containing particles (i.e., D_c and D_p) obtained from the SP2 measurement (discussed in Sect. 2.2.1), we used Mie theory with a shell-and-core model to retrieve the optical properties of BC-containing particles, including the absorption enhancement (E_{ab}) of rBC, the mass absorption cross section (MAC) of BC-containing particles and bare rBC cores, the mass scattering cross section of bare BC core (MSC_{core}), and the absorption coefficient (σ_{ab}) of BC-containing particles. The calculation of these parameters is described below.

The E_{ab} characterizes the increase in BC light absorption due to the lensing effect of coating materials on the BC surface and is used to quantify the theoretical light absorption capability of BC-containing particles in this study. The E_{ab} is determined by the ratio of the absorption cross section of the entire BC-containing particle ($C_{ab,p}$) to that of the bare BC core ($C_{ab,c}$), as expressed in Eq. (2):

$$E_{ab} = \frac{C_{ab,p}(D_c, D_p, RI_s, RI_c)}{C_{ab,c}(D_c, RI_c)}, \quad (2)$$

where $C_{ab,p}$ is determined by the D_c , D_p , RI_s and RI_c using Mie calculation, and $C_{ab,c}$ is determined by the D_c and RI_c .

The MAC of BC-containing particles (MAC_p) and bare rBC cores (MAC_c) is defined as the $C_{ab,p}$ and $C_{ab,c}$ per unit rBC mass, as Eqs. (3) and (4), respectively:

$$MAC_p = \frac{C_{ab,p}(D_c, D_p, RI_s, RI_c)}{m_{rBC}}, \quad (3)$$

$$MAC_c = \frac{C_{ab,c}(D_c, RI_c)}{m_{rBC}}. \quad (4)$$

MSC_{core} is the scattering cross sections of bare rBC cores ($C_{sca,c}$) per unit of rBC mass, as calculated by Eq. (5):

$$MSC_{core} = \frac{C_{sca,c}(D_c, RI_c)}{m_{rBC}}. \quad (5)$$

The σ_{ab} of BC-containing particles is calculated based on the MAC and the rBC mass concentration (C_{rBC}) measured by the SP2, as expressed in Eq. (6). The uncertainties of σ_{ab} related to the MAC of bare rBC cores from Mie calculation were evaluated in the Supplement (Fig. S9 and the associated discussion).

$$\sigma_{ab,calculated} = MAC_p \times C_{rBC} = MAC_c \times E_{ab} \times C_{rBC} \quad (6)$$

2.3 BC effective emission intensity

To evaluate the impact of regional transport on BC-containing particles, we used a variant of the effective emission intensity (EEI) defined by Lu et al. (2012) to quantify the amounts of BC over the observation site from different source regions. In this study, the spatial origin of the BC observed at our site was divided into local sources in Beijing and regional sources in other areas (i.e., Hebei, Tianjin, Shanxi and Inner Mongolia; Fig. S1). The EEI takes into account emission, transport, hydrophilic-to-hydrophobic conversion and removal processes (i.e., dry and wet deposition) of BC throughout the whole atmospheric transport process from the origin of the BC emission to the receptor site. A novel back-trajectory approach was developed by Lu et al. (2012) to calculate EEI values.

In this study, the back-trajectory analysis was performed by the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPPLIT) model to obtain the transport pathways of BC to the observation site (40°00′17″ N, 116°19′34″ E) during the campaign period (17–30 November 2014). The 72 h back trajectory at 100 m every hour was calculated with the meteorological fields of NCEP Global Data Assimilation System at a 1° × 1° resolution. An anthropogenic BC emission inventory of China in the year 2012 at a resolution of 0.25° × 0.25° was used to support the back-trajectory analysis. The gridded BC emission data are from the Multi-resolution Emission Inventory for China (MEIC) developed by Tsinghua University (<http://www.meicmodel.org>, last access: 20 June 2018).

We calculated the EEI of BC at a resolution of 0.25° × 0.25° based on the algorithm developed by Lu et al. (2012). Following a trajectory l every hour, the fresh BC emitted from a series of spatial grids in sequence (i.e., $l_1, l_2, \dots, l_i, \dots$) is transported to the receptor grid (i.e., l_n). The EEI of trajectory l at the surface grid point i ($EEI_{i,l}$) was determined by Eq. (7):

$$EEI_{i,l} = E_i \times TE_{i,l}, \quad (7)$$

where E_i is the BC emission at surface grid point i , and $TE_{i,l}$ represents the BC transport efficiency of trajectory l from grid point i to the receptor site, as calculated by Eqs. (1)–(4) in Lu et al. (2012).

The total EEI of trajectory l characterizes the total amount of BC transported to the observation site every hour, expressed as Eq. (8):

$$EEI_{total} = \sum_{i=1}^n EEI_{i,l}. \quad (8)$$

2.4 BC radiative efficiency

In this study, we use a parameter of the simple forcing efficiency (SFE) to roughly evaluate the radiative forcing of

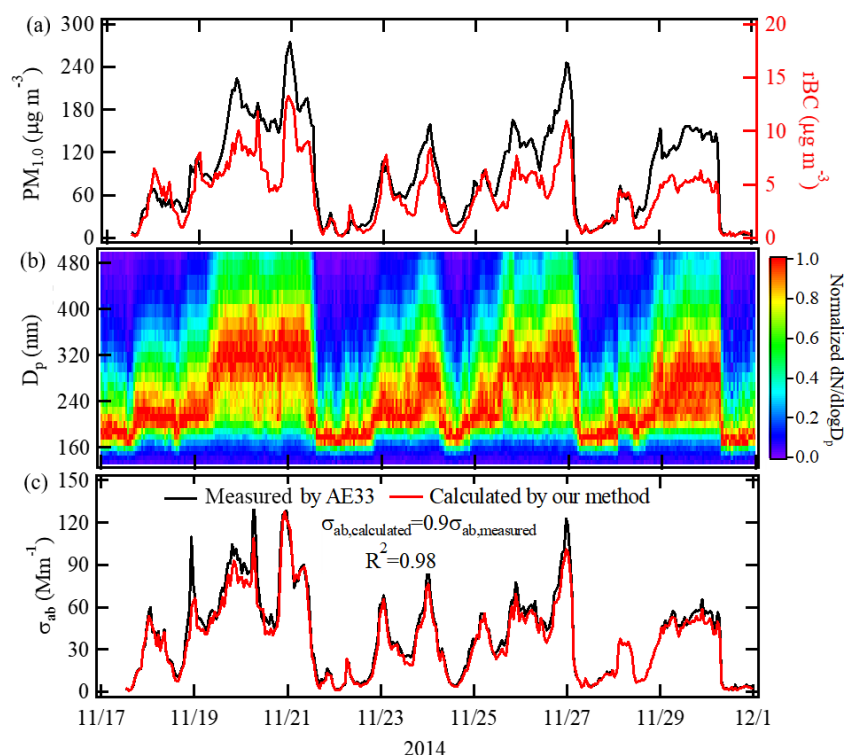


Figure 1. Time series of (a) the $\text{PM}_{1.0}$ and rBC mass concentrations, (b) the diameter of BC-containing particles (D_p), and (c) the light absorption coefficient (σ_{ab}) at 880 nm. The correlation between the calculated σ_{ab} ($\sigma_{\text{ab,calculated}}$) using Mie theory combined with SP2 measurements and the measured σ_{ab} ($\sigma_{\text{ab,measured}}$) from the AE33 is also shown in (c).

BC-containing particles. The SFE is defined as normalized radiative forcing by BC mass, which is wavelength dependent (Chylek and Wong, 1995; Bond and Bergstrom, 2006; Chen and Bond, 2010; Saliba et al., 2016). The wavelength-dependent SFE for BC-containing particles is determined by Eq. (9):

$$\frac{\text{dSFE}}{\text{d}\lambda} = -\frac{1}{4} \frac{\text{d}S(\lambda)}{\text{d}\lambda} \tau_{\text{atm}}^2(\lambda) (1 - F_c) \times \left[2(1 - \alpha_s)^2 \beta(\lambda) \cdot \text{MSC}_{\text{core}}(\lambda) - 4\alpha_s \times \text{MAC}(\lambda) \right], \quad (9)$$

in which a wavelength (λ) of 550 nm is used in this study; $\text{d}S(\lambda)/\text{d}\lambda$ is the spectral solar irradiance, the value of which is from the ASTM G173-03 Reference Spectra ($1.86 \text{ W m}^{-2} \text{ nm}^{-1}$ at 550 nm) and the parameters τ_{atm} , F_c , α_s and β are the atmospheric transmission (0.79), cloud fraction (0.6), urban surface albedo (0.15) and backscatter fraction (0.17), respectively (Chen and Bond, 2010; Park et al., 2011; Jeong et al., 2013; Saliba et al., 2016).

3 Results

3.1 Light absorption of BC-containing particles during the campaign period

Figure 1 shows the time series of the $\text{PM}_{1.0}$ and rBC mass concentration, the diameter of BC-containing particles (D_p) and the measured and calculated light absorption coefficient (σ_{ab}) at 880 nm. During the campaign period, four episodes with different $\text{PM}_{1.0}$ evolution processes (Fig. 1a) were observed: 18–21, 23–24, 25–26 and 28–30 November. The hourly $\text{PM}_{1.0}$ mass concentration ranged from 3.5 to $275 \mu\text{g m}^{-3}$, with an average value of $91 \mu\text{g m}^{-3}$ during the observed period. The rBC mass concentration accounted for $\sim 5\%$ of $\text{PM}_{1.0}$. The D_p including rBC cores and coating materials shown in Fig. 1b exhibited an excellent temporal coherence with the rBC mass concentration. Figure 1b shows that the number distribution of D_p for BC-containing particles exhibited a peak at 180–320 nm, significantly larger than the peak value (D_c of ~ 95 nm) for number size distribution of bare rBC cores (Fig. S5a) due to an abundance of coating materials on BC surfaces. The size information (namely entire particle size and rBC core size) of BC-containing particles observed in our study was consistent with that (D_p of ~ 200 –300 nm and D_c of ~ 70 –100 nm) in previous studies in China (Huang et al., 2013; Wang et al., 2014b; Gong et al., 2016).

Moreover, the D_p exhibited sustained growth from ~ 180 nm to ~ 400 nm during a pollution episode, which could be a consequence of the increase in either D_c or coating materials, or both. Figure S6a shows a slight change in D_c with pollution development. However, the coating thickness of BC-containing particles increased with PM_{10} concentration (Fig. S10a). Therefore, the sustained growth of D_p during a pollution episode was dominated by more coating materials under more polluted conditions. Figure S10 shows the simultaneous increase in the rBC mass concentration and the amount of coating materials on the BC surface, which could significantly enhance the light absorption of BC-containing particles. Figure S11 shows that the measured absorption coefficient ($\sigma_{ab, \text{measured}}$) at 880 nm (dominated by BC component at this wavelength; Drinovec et al., 2015) for the aerosol particles measured with the AE33 exhibited an 18-fold increase in conjunction with an increase in the PM_{10} concentration from ~ 10 to $\sim 230 \mu\text{g m}^{-3}$, larger than the increase (~ 14 times) in rBC mass concentration.

To validate SP2 measurements and Mie calculation used in this study, we compared the calculated light absorption coefficient ($\sigma_{ab, \text{calculated}}$) of BC-containing particles using Eq. (6) with the measured light absorption coefficient ($\sigma_{ab, \text{measured}}$) from AE33, as shown in Fig. 1c. The $\sigma_{ab, \text{calculated}}$ values for BC-containing particles showed an excellent agreement with the $\sigma_{ab, \text{measured}}$ values measured by the AE33, with a difference of $\sim 10\%$ ($R^2 = 0.98$). The difference was dominated by the uncertainties from the compensation algorithm used in the AE33 measurements ($\sim 15\%$; details shown in Fig. S2 and the associated discussion in the Supplement) and Mie calculation (10–20%; Figs. S4, S5 and S9 and the associated discussion in the Supplement). The uncertainty evaluation revealed that the difference between $\sigma_{ab, \text{calculated}}$ and $\sigma_{ab, \text{measured}}$ ($\sim 10\%$) shown in Fig. 1c is reasonable. The comparison between the $\sigma_{ab, \text{calculated}}$ and $\sigma_{ab, \text{measured}}$ values implied that the optical properties (i.e., σ_{ab} , MAC and E_{ab}) of the BC-containing particles derived from Mie calculation combined with SP2 measurements (i.e., rBC concentrations, D_p and D_c) were reliable in our case.

3.2 Enhancement of the theoretical light absorption capability of black carbon associated with air pollution

3.2.1 The D_p / D_c ratio and calculated E_{ab} under different PM_{10} concentrations

Previous theoretical studies reported that the coating materials on the BC surface can significantly enhance the light absorption of BC via the lensing effect (Fuller et al., 1999; Jacobson, 2001; Moffet et al., 2009; Lack and Cappa, 2010). In other words, the aging degree of BC-containing particles (characterized by the D_p / D_c ratio in this study) determines their theoretical light absorption capability (characterized by the calculated MAC and E_{ab} in this study). However, whether

and how the aging degree and light absorption capability of BC-containing particles will change under different pollution levels remain unclear. During the campaign period, we found that the mass-averaged values of the D_p / D_c ratio and calculated E_{ab} of BC-containing particles increased with increasing air pollution levels.

Figure 2a shows the D_p / D_c ratio and calculated E_{ab} of BC-containing particles with rBC cores at 75–300 nm under different PM_{10} concentrations in the range of 1.2–3.5 and 1.3–3.1, respectively. In terms of BC-containing particles with a certain rBC core size, their D_p / D_c ratio and calculated E_{ab} were greater under higher PM_{10} concentrations, which could be attributed to more coating materials on BC surfaces under an environment with more pollution. The increase in both primary and secondary components under more polluted conditions was favorable to BC aging by coagulation and condensation, which happen mostly between BC and non-BC species. On average (i.e., mass-weighted mean across rBC core size larger than ~ 75 nm), the D_p / D_c and calculated E_{ab} for observed BC-containing particles in SP2 under different PM_{10} concentrations during the campaign period varied in the range of 1.6–2.2 and 1.6–2.0, respectively (Fig. 2b). Correspondingly, the mass-averaged values of the D_p / D_c and calculated E_{ab} of BC-containing particles increased by $\sim 33\%$ and $\sim 18\%$, respectively, with increasing PM_{10} concentrations from ~ 10 to $\sim 230 \mu\text{g m}^{-3}$ (compared with rBC mass concentration increasing from ~ 0.7 to $\sim 11 \mu\text{g m}^{-3}$).

Figure 3 shows the increase of the D_p / D_c ratio and calculated E_{ab} (IR_{D_p/D_c} and $IR_{E_{ab}}$, respectively) for BC-containing particles with increasing PM_{10} concentrations from 10 to $230 \mu\text{g m}^{-3}$ as a function of rBC core size. Based on the D_p / D_c ratio and calculated E_{ab} of BC-containing particles with size-resolved rBC cores in SP2 measurement under different PM_{10} concentrations (shown in Fig. 2a), we can obtain the measured IR_{D_p/D_c} and $IR_{E_{ab}}$ as a function of rBC core size. When PM_{10} concentration increases from ~ 10 to $\sim 230 \mu\text{g m}^{-3}$ during the campaign period, the D_p / D_c ratio and calculated E_{ab} of BC-containing particles with rBC cores at 75–200 nm increased by 28–48% and 13–36%, respectively. The size-dependent increase in D_p / D_c ratio and calculated E_{ab} associated with air pollution indicated that the aging process of smaller rBC was relatively more sensitive to air pollution levels. This could be attributed to the fact that the condensational growth associated with air pollution due to the formation of secondary components is more effective for smaller particles in terms of increasing the diameter (Metcalf et al., 2013).

Meanwhile, following a semiquantitative analysis used in Metcalf et al. (2013), we calculated the IR_{D_p/D_c} and $IR_{E_{ab}}$ based on the diffusion-controlled growth law (Seinfeld and Pandis, 2006). The calculated IR_{D_p/D_c} and $IR_{E_{ab}}$ compare with the measured ones from SP2.

According to the diffusion-controlled growth law (Seinfeld and Pandis, 2006), the evolution of the size of BC-

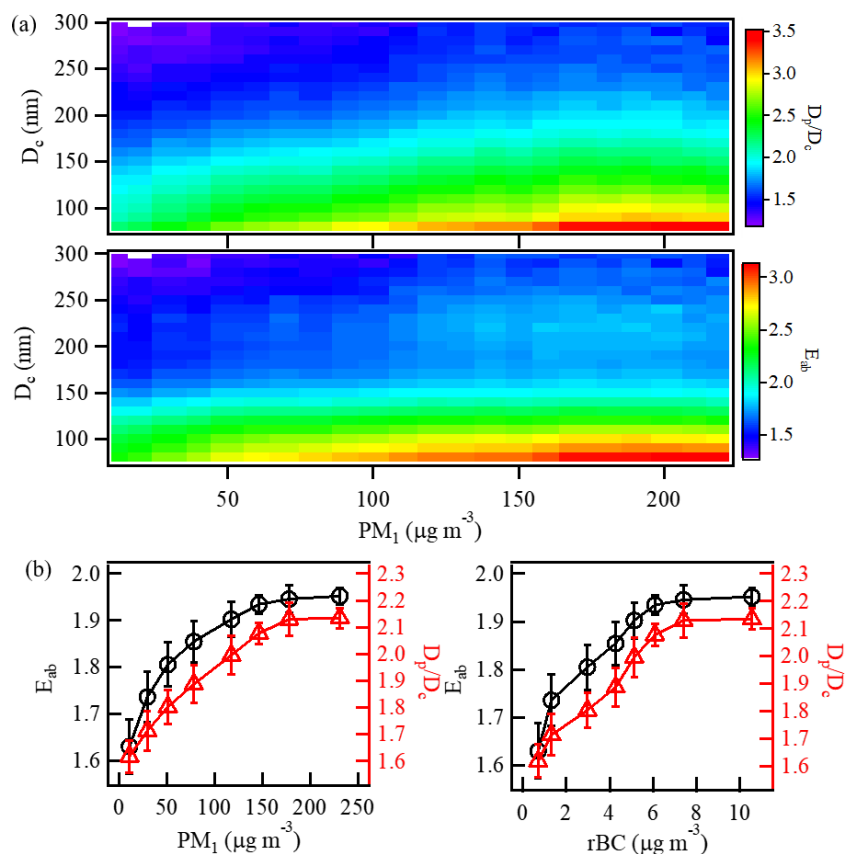


Figure 2. (a) The aging degree (D_p/D_c ratio) and light absorption enhancement (calculated E_{ab}) of BC-containing particles with size-resolved rBC cores (D_c) under different PM_1 concentrations; (b) variations in the mass-averaged values of the D_p/D_c ratio and calculated E_{ab} of BC-containing particles with PM_1 and rBC concentrations.

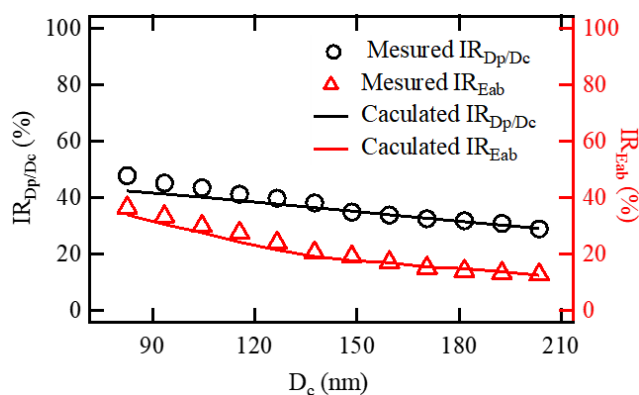


Figure 3. The increase in the D_p/D_c ratio and calculated E_{ab} (IR $_{D_p/D_c}$ and IR $_{E_{ab}}$) for BC-containing particles with PM_1 concentration increasing from 10 to 230 $\mu g m^{-3}$. The calculated IR $_{D_p/D_c}$ and IR $_{E_{ab}}$ values were determined based on Eqs. (13) and (14). The measured IR $_{D_p/D_c}$ and IR $_{E_{ab}}$ values were obtained from SP2 measurements.

containing particles (D_p) is shown:

$$\frac{dD_p}{dt} = \frac{A}{D_p}, \quad (10)$$

in which $\frac{dD_p}{dt}$ represents the diffusion-controlled growth rate; A is the parameter. Integrating Eq. (10) with $D_p(t=0) = D_c$:

$$D_p^2 = D_c^2 + 2At = D_c^2 + B, \quad (11)$$

in which D_c is rBC core diameter and B (i.e., equal to $2At$) is the parameter, varying under different PM_1 concentrations.

Following Eq. (11), the D_p/D_c ratio is given:

$$\frac{D_p}{D_c} = \left(\frac{B}{D_c^2} + 1 \right)^{1/2}, \quad (12)$$

where the parameter B is determined by the value of the measured D_p/D_c ratio with D_c of 160 nm under different PM_1 concentrations.

The IR $_{D_p/D_c}$ for BC-containing particles with PM_1 concentration increasing from 10 to 230 $\mu g m^{-3}$ can be calculated by Eq. (13):

$$IR_{D_p/D_c} = \frac{\left(\frac{D_p}{D_c} \right)_{230} - \left(\frac{D_p}{D_c} \right)_{10}}{\left(\frac{D_p}{D_c} \right)_{10}}$$

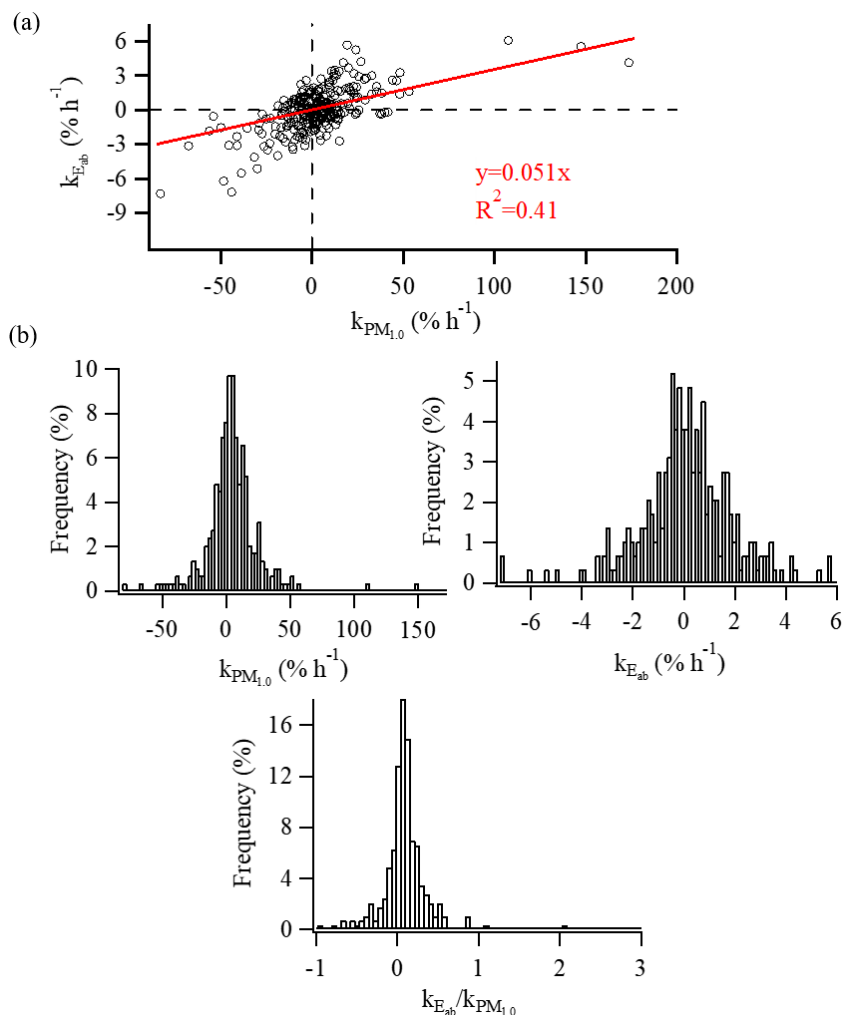


Figure 4. (a) Correlation between the changing rate of calculated E_{ab} ($k_{E_{ab}}$) and the changing rates of PM_1 concentrations (k_{PM_1}) during the campaign period. (b) Frequency distribution of $k_{E_{ab}}$, k_{PM_1} and $k_{E_{ab}}/k_{PM_1}$. The $k_{E_{ab}}$ and k_{PM_1} values represent an apparent changing rate of calculated E_{ab} and PM_1 concentration, respectively, and are from point-by-point differences of hourly E_{ab} and PM_1 concentration, namely $k_{E_{ab}} = (E_{ab,t2} - E_{ab,t1}) / (E_{ab,t1} * (t_2 - t_1))$ and $k_{PM_1} = (PM_{1,t2} - PM_{1,t1}) / (PM_{1,t1} * (t_2 - t_1))$. The sensitivity of the change in calculated E_{ab} with changing PM_1 concentrations was obtained by plotting $k_{E_{ab}}$ vs. k_{PM_1} (i.e., $k_{E_{ab}}/k_{PM_1}$).

$$= \frac{\left(\frac{B_{230}}{D_c^2} + 1\right)^{1/2} - \left(\frac{B_{10}}{D_c^2} + 1\right)^{1/2}}{\left(\frac{B_{10}}{D_c^2} + 1\right)^{1/2}}, \quad (13)$$

$$= \frac{k_{230} \times \left(\frac{B_{230}}{D_c^2} + 1\right)^{1/2} - k_{10} \times \left(\frac{B_{10}}{D_c^2} + 1\right)^{1/2}}{k_{10} \times \left(\frac{B_{10}}{D_c^2} + 1\right)^{1/2}}. \quad (14)$$

where $\left(\frac{D_p}{D_c}\right)_{230}$ and $\left(\frac{D_p}{D_c}\right)_{10}$ represent the D_p/D_c ratio when PM_1 concentrations are 230 and $10 \mu\text{g m}^{-3}$, respectively; B_{230} and B_{10} are parameter B with PM_1 concentrations of 230 and $10 \mu\text{g m}^{-3}$, respectively.

The $IR_{E_{ab}}$ for BC-containing particles with a PM_1 concentration increasing from 10 to $230 \mu\text{g m}^{-3}$ can be derived based on $E_{ab} = k \times (D_p/D_c)$, as expressed in Eq. (5):

$$IR_{E_{ab}} = \frac{k_{230} \times \left(\frac{D_p}{D_c}\right)_{230} - k_{10} \times \left(\frac{D_p}{D_c}\right)_{10}}{k_{10} \times \left(\frac{D_p}{D_c}\right)_{10}}$$

We compared the calculated IR_{D_p/D_c} and $IR_{E_{ab}}$ based on Eqs. (13) and (14) with those from SP2 measurements, as shown in Fig. 3. The agreement indicated that the increase in D_p/D_c and E_{ab} for BC-containing particles with increasing PM_1 concentrations follows the diffusion-controlled growth law.

3.2.2 Changing rate of the theoretical light absorption capability of black carbon

Figure 4a explores the relationship between the changing rate of calculated E_{ab} ($k_{E_{ab}}$) and the changing rates of PM_1 con-

centrations (k_{PM_1}) with pollution development. Linear relationships were estimated, i.e., $k_{E_{\text{ab}}} \approx 0.051 k_{\text{PM}_1}$, revealing that rapid increases in air pollution levels could lead to rapid increases in BC light absorption capability. When the PM_1 concentration exhibited a sharp increase related to an extreme haze episode (Zheng et al., 2015), the increase in BC light absorption capability was dramatic.

Figure 4b shows frequency distribution of $k_{E_{\text{ab}}}$, k_{PM_1} and $k_{E_{\text{ab}}} / k_{\text{PM}_1}$ ratio. During the campaign period, most of $k_{E_{\text{ab}}}$ and k_{PM_1} values were in the range of -50 to 50 and -4 to $4\% \text{ h}^{-1}$, respectively, revealing a lower changing rate for BC aging than that for PM_1 concentration. The peak value of frequency distribution of $k_{E_{\text{ab}}}$ was around zero, indicating the BC-containing particles were shrinking as often as they were growing. The $k_{E_{\text{ab}}} / k_{\text{PM}_1}$ ratio characterized the sensitivity of the change in calculated E_{ab} with changing PM_1 concentrations. The frequency distribution of the $k_{E_{\text{ab}}} / k_{\text{PM}_1}$ ratio showed that $\sim 60\%$ values were in the range of 0 – 1 , with a peak value around 0.05 . Smaller values of the $k_{E_{\text{ab}}} / k_{\text{PM}_1}$ ratio indicated that the change of calculated E_{ab} was not sensitive to variations in PM_1 concentrations. The growth rate of E_{ab} (for BC samples with $k_{E_{\text{ab}}} > 0$: 0.1 – $7.3\% \text{ h}^{-1}$) observed at our study was consistent with the BC aging rate (0.2 – $7.8\% \text{ h}^{-1}$) in previous studies (Moteki et al., 2007; Shiraiwa et al., 2007; Cheng et al., 2012).

Moreover, we found that the growth rate of E_{ab} decreased with increasing PM_1 mass concentrations (Fig. S12a), indicating that the increase in the theoretical light absorption capability of BC-containing particles slowed with further pollution development. This can be explained by larger BC-containing particles when PM_1 concentration is higher (Fig. S12b). The net change in diameter for a given amount of material deposited decreases with increasing particle size due to surface-to-volume scaling, which would expect the growth rate of particles to decrease with increasing PM_1 concentration and thus the $k_{E_{\text{ab}}}$ would also decrease (Fig. S12a).

The evolution of theoretical light absorption of BC with pollution levels depends on the change in both rBC mass concentrations and calculated E_{ab} . Figure S13 shows markedly smaller $k_{E_{\text{ab}}}$ than the changing rate of rBC mass concentrations (k_{rBC}) (i.e., $k_{E_{\text{ab}}} \approx 0.027 k_{\text{rBC}}$), indicating the change of calculated E_{ab} was significantly slower than that of rBC mass concentrations under different pollution levels. Due to less sensitivity of calculated E_{ab} to change in air pollution levels compared with that of rBC mass concentrations, some previous measurements (Andreae et al., 2008; Ram et al., 2009; McMeeking et al., 2011; Wang et al., 2014b) would not have been able to easily discern a difference of E_{ab} among different pollution levels and thus just focus on the change of BC mass concentration. This would lead to uncertainties in estimation of BC light absorption. In our case, Fig. 2b reveals the mass-weighted average of calculated E_{ab} increased by $\sim 18\%$ with the PM_1 concentration increasing from ~ 10 to $\sim 230 \mu\text{g m}^{-3}$ (accompanied by the rBC mass concentration increasing from ~ 0.7 to $\sim 11 \mu\text{g m}^{-3}$). If the increase

Table 1. The average PM_1 mass concentration, rBC mass concentration, normalized $\text{EEI}_{\text{total}}$, $\text{EEI}_{\text{outside}} / \text{EEI}_{\text{total}}$ ratio, mass-averaged values of the $D_{\text{p}} / D_{\text{c}}$ ratio and calculated E_{ab} during clean, slightly polluted and polluted periods. $\text{EEI}_{\text{outside}}$ is the EEI of BC from areas outside Beijing; the $\text{EEI}_{\text{outside}} / \text{EEI}_{\text{total}}$ ratio reflects the amount of BC contributed by regional transport to the total amount of BC observed at our site. The clean ($\text{PM}_{2.5} \leq 35 \mu\text{g m}^{-3}$), slightly polluted ($35 \mu\text{g m}^{-3} < \text{PM}_{2.5} \leq 115 \mu\text{g m}^{-3}$) and polluted ($\text{PM}_{2.5} > 115 \mu\text{g m}^{-3}$) periods were classified according to the Air Quality Index (Zheng et al., 2015).

	Clean	Slightly polluted	Polluted
$\text{PM}_1 (\mu\text{g m}^{-3})$	12.57	54.26	141.93
rBC ($\mu\text{g m}^{-3}$)	0.82	2.89	6.07
Normalized $\text{EEI}_{\text{total}}$	3.68	9.19	16.87
$\text{EEI}_{\text{outside}} / \text{EEI}_{\text{total}}$	0.21	0.39	0.63
$D_{\text{p}} / D_{\text{c}}$	1.62	1.81	2.04
Calculated E_{ab}	1.66	1.81	1.91

in calculated E_{ab} of BC with PM_1 increase was neglected in this study, the theoretical light absorption of BC-containing particles would be underestimated by $\sim 18\%$ under polluted conditions.

3.3 Contribution of regional transport

BC aging in the atmosphere, namely BC internally mixing with other aerosol components, is associated with atmospheric transport (Gustafsson and Ramanathan, 2016). In Beijing, the rapid increase in aerosol particle concentrations during pollution episodes is most likely caused by regional transport of polluted air mass (Yang et al., 2015; Zheng et al., 2015). Therefore, regional transport of pollution may play an important role in the enhancement of BC light absorption capability associated with air pollution. In this study, we used the EEI analysis (Lu et al., 2012) to explore the effects of regional transport on the increase in theoretical light absorption capability of BC with increasing pollution levels.

Figure 5 shows the spatial distribution ($0.25^\circ \times 0.25^\circ$) of the EEI for BC transported to the observation site under different pollution levels (i.e., clear, slightly polluted and polluted periods) during the campaign period. The spatial origin of the BC observed at our site varied significantly among the different pollution periods. In this study, the spatial origin of total BC at the site was classified into local Beijing and other regions (i.e., outside of Beijing is considered to be regional origins in this study). Noted that the local region (i.e., Beijing) defined in this study is smaller than areas outside of Beijing (e.g., Hebei, Tianjin, Shanxi and Inner Mongolia; Fig. S1). Table 1 lists the contribution of BC from regional origins (i.e., $\text{EEI}_{\text{outside}} / \text{EEI}_{\text{total}}$ ratio). During a polluted period, the contributions of BC from regional origins was $\sim 63\%$, larger than that from local Beijing ($\sim 37\%$). This was partly due to comparing the contributions from a

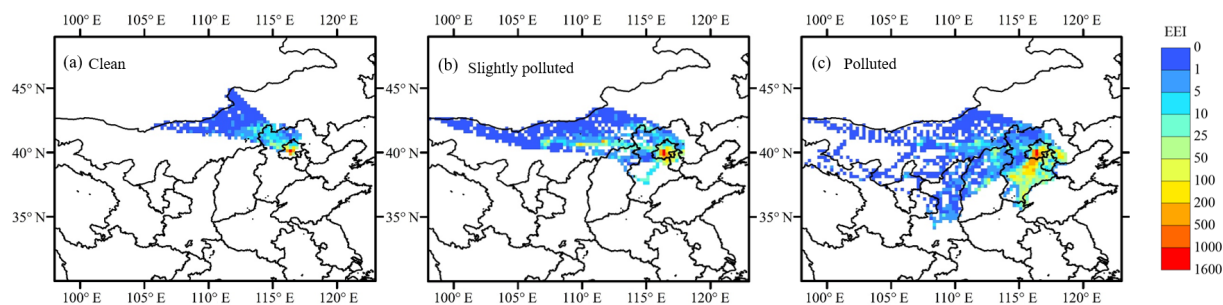


Figure 5. Spatial distribution ($0.25^\circ \times 0.25^\circ$) of the effective emission intensity (EEI, units of tons per grid per year) for BC transported to the observation site ($40^\circ 00' 17''$ N, $116^\circ 19' 34''$ E) during clean, slightly polluted and polluted periods. The clean ($\text{PM}_{2.5} \leq 35 \mu\text{g m}^{-3}$), slightly polluted ($35 \mu\text{g m}^{-3} < \text{PM}_{2.5} \leq 115 \mu\text{g m}^{-3}$) and polluted ($\text{PM}_{2.5} > 115 \mu\text{g m}^{-3}$) periods were classified according to the Air Quality Index (Zheng et al., 2015). The site location and the boundaries of inside the region (i.e., Beijing) vs. outside the region (i.e., other areas such as Tianjin, Hebei, Inner Mongolia, Shanxi and Shandong) are shown in Fig. S1. Note that these regions are defined based on political boundaries.

small region (Beijing) and a large region (outside of Beijing). In this study, we focus on comparing the contributions of BC from outside of Beijing (considered to be regional origins in this study) among different pollution levels (i.e., clean, slight polluted and polluted periods). The BC from regional origins (i.e., outside of Beijing) accounted for ~ 21 , ~ 39 and ~ 63 % of total BC amount at the site during the clean, slightly polluted and polluted periods, respectively. This revealed that the regional contribution to BC over Beijing increased as the air pollution levels increased.

Due to the increase in the regional contribution, the total BC amount transported to the observation site, characterized by the EEI ($\text{EEI}_{\text{total}}$) in this study, increased under more polluted conditions. Table 1 shows that the $\text{EEI}_{\text{total}}$ was 4.6 times higher during the polluted period than during the clean period, revealing that the polluted air mass brought more BC to Beijing. BC concentration at the site strongly depends on both total BC amount (transported from local Beijing and other regions, characterized by $\text{EEI}_{\text{total}}$ in this study) and local meteorology. Table 1 shows that the BC concentrations from the clean period are ~ 7.4 times lower than in the polluted period. The increase in $\text{EEI}_{\text{total}}$ (~ 4.6 times) accounted for an ~ 62 % increase in BC mass concentrations (~ 7.4 times). This indicated that the adverse local meteorology contributed ~ 38 % of the increase in BC mass concentration at the site from the clean period to the polluted period. Compared with regional transport, less of an effect of adverse local meteorology might be attributed to relatively small areas defined as the local region (i.e., Beijing) in this study. Polluted events in China always occur over a large region, e.g., North China Plain (Zheng et al., 2015; Yang et al., 2017). For our case, the adverse meteorology during polluted days in the whole large region including Beijing and other areas can lead to the increase in pollutants and then more transport of pollutants into Beijing. Yang et al. (2017) found that the increases in BC concentration under polluted conditions over the North China Plain (including Beijing and

other adjacent areas) is dominated by its local emissions due to adverse meteorology.

Under different pollution levels, regional transport not only influenced the BC mass concentrations but also the BC aging process and timescale. Table 1 shows that the mass-average value of the D_p/D_c ratio of BC-containing particles at our site was ~ 2.04 during the polluted period, significantly higher than that observed during the clean period (~ 1.62) and slightly polluted period (~ 1.81). On the one hand, under more polluted conditions, more BC-containing particles in Beijing were from regional sources and thus had undergone a longer aging time during the transport than the BC from local sources in Beijing. On the other hand, compared with the BC carried in the clean air mass from the northwest of Beijing during the clean period (Fig. 5a), the BC-containing particles in the polluted air mass underwent regional transport from the region south of Beijing (i.e., Hebei, one of the most polluted provinces in China, with high pollutant emissions) during the polluted periods (Fig. 5c). Peng et al. (2016) pointed out higher BC aging rates under more polluted environments, indicating that BC-containing particles passing through polluted regions would show higher aging rates during atmospheric transport than those from clean regions. The mass-average values of calculated E_{ab} for BC-containing particles observed at our site were ~ 1.66 , ~ 1.81 and ~ 1.91 during the clean, slightly polluted and polluted periods, respectively (Table 1), showing that the theoretical light absorption capability of BC-containing particles observed at our site increased with increasing regional contributions. Our results demonstrated the importance of regional transport in the enhancement of BC light absorption capability associated with air pollution.

To further explore the importance of aging during regional transport, its contributions were compared with those of local chemical processes with respect to increases in D_p/D_c ratio and theoretical light absorption capability of BC-containing particles associated with air pollution. Considering the im-

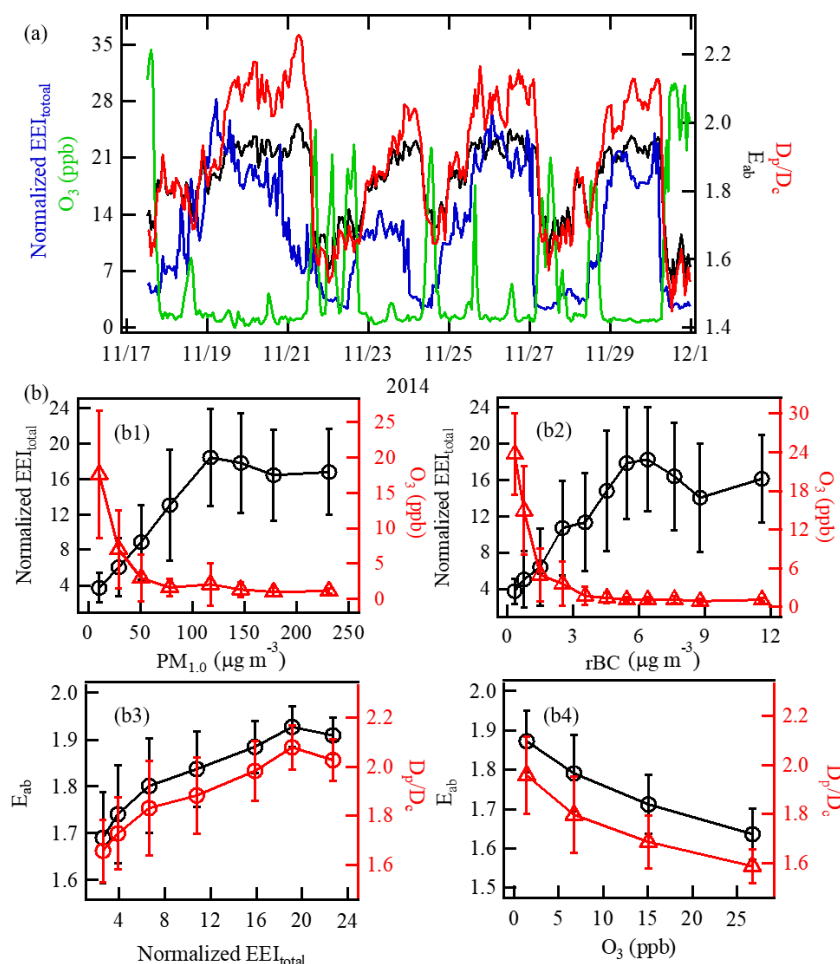


Figure 6. (a) Time series of the normalized EEI_{total} , D_p/D_c ratios and E_{ab} of BC-containing particles and the O_3 concentration during the campaign period. (b) Variations in the D_p/D_c and E_{ab} of BC-containing particles with the normalized EEI_{total} and O_3 concentration. Normalized EEI_{total} ($EEI_{total, normalized}$) was calculated by scaling by a factor of 10^{-3} , namely $EEI_{total, normalized} = EEI_{total}/1000$.

portance of photochemical oxidation in coating formation on BC surface (Metcalf et al., 2013; Peng et al., 2016), we evaluated the capability of local photochemical production by the changes of O_3 concentrations in the atmosphere. Conversely, the changes in the amount of BC from regional transport were characterized by variation in EEI_{total} , which was used to evaluate the contributions of regional transport to BC aging. Figure 6a shows that the EEI_{total} per hour exhibited a temporal coherence with the mass-averaged values of the D_p/D_c ratio and calculated E_{ab} of BC-containing particles. In contrast, the O_3 concentrations showed a different temporal trend. When PM_1 concentrations were higher than $\sim 120 \mu\text{g m}^{-3}$, O_3 concentrations decreased to ~ 2 ppb. Zheng et al. (2015) has demonstrated the weakened importance of photochemistry in the production and aging of secondary aerosols in Beijing under polluted conditions due to a decrease in oxidant concentrations. This indicated that the photochemical processing in BC aging may be weakened under higher polluted levels (i.e., $PM_1 > 120 \mu\text{g m}^{-3}$). Note that

photochemical processing is not the only possible pathway in the BC aging process and other pathways were not discussed in this study. The local aging process of BC might be enhanced by other pathways. For example, high concentrations of aerosols under a polluted environment may compensate for the adverse photochemical conditions for BC aging. In summary, the increases in the aging degree and theoretical light absorption capability of BC-containing particles with increasing air pollution were more likely caused by aging during regional transport than by local photochemical production.

According to the evolution of the EEI_{total} values and O_3 concentrations with increasing air pollution levels (Fig. 6b1 and b2), we separated the pollution levels into two periods. When PM_1 concentrations were lower than $\sim 120 \mu\text{g m}^{-3}$ and rBC mass concentrations were lower than $\sim 6 \mu\text{g m}^{-3}$, the normalized EEI_{total} increased from ~ 3 to ~ 18 with increasing air pollution levels, and the O_3 concentrations decreased from ~ 20 ppb to ~ 2 ppb, indicating

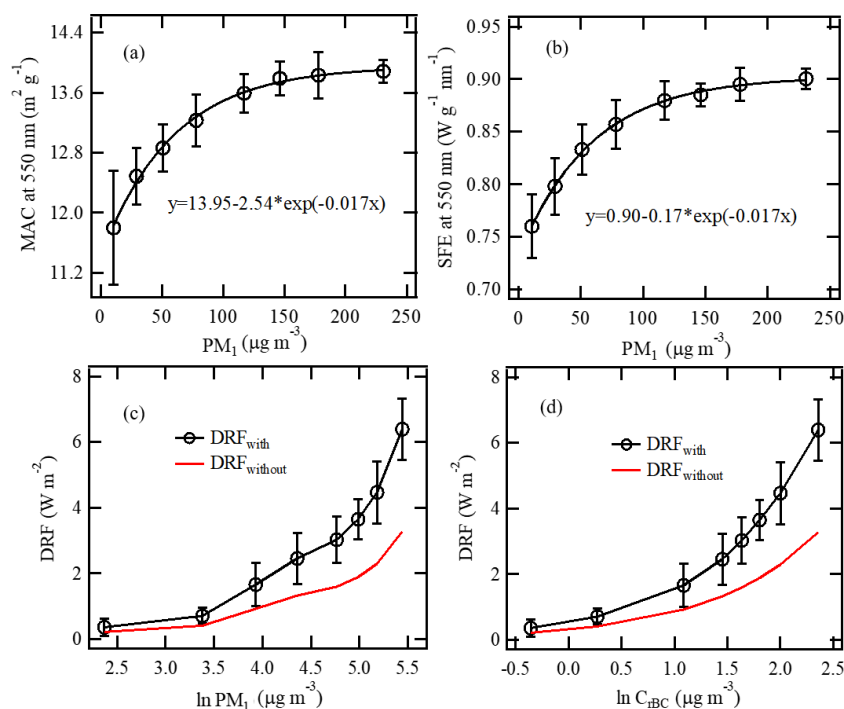


Figure 7. Variations in MAC, SFE and DRF of BC-containing particles under different pollution levels. The DRF values for BC-containing particles were obtained by scaling the average DRF (0.31 W m^{-2} , Table S1) of externally mixed BC from various climate models (Bond et al., 2013). The DRF_{with} and $\text{DRF}_{\text{without}}$ values respectively represent DRF with and without considering the differences of light absorption capability of ambient BC-containing particles under different pollution levels; namely, the DRF_{with} was calculated with scaling factors of the E_{ab} and $r\text{BC}$ mass concentration (C_{rBC}) and the $\text{DRF}_{\text{without}}$ was calculated with a scaling factor of C_{rBC} . Variations in DRF_{with} under different pollution levels were due to the change of both the light absorption capability and mass concentration of BC; variations in $\text{DRF}_{\text{without}}$ were only attributed to the change of $r\text{BC}$ mass concentrations.

enhanced regional contributions and weakened local photochemical production at the observation site. In this period, Fig. 6b3 and b4 show that the mass-averaged values of calculated E_{ab} and the $D_{\text{p}}/D_{\text{c}}$ ratio of BC-containing particles increased from ~ 1.6 to ~ 1.9 and from ~ 1.6 to ~ 2.0 , respectively, with the increase in the normalized $\text{EEI}_{\text{total}}$ (from ~ 3 to ~ 18) and the decrease in the O_3 concentrations (from ~ 20 to ~ 2 ppb). Therefore, in terms of the increase in the BC light absorption capability with increasing air pollution levels, this period (i.e., conditions of $\text{PM}_{10} < 120 \mu\text{g m}^{-3}$ and $r\text{BC} < 6 \mu\text{g m}^{-3}$) represented a regional-transport-controlled period. The increase in calculated E_{ab} of BC-containing particles (~ 1.6 – 1.9) during this regional-transport-controlled period accounted for $\sim 75\%$ of the increase in calculated E_{ab} (~ 1.6 – 2.0) with increasing air pollution during the whole campaign period. Therefore, the aging process during the regional transport dominated the increase in the theoretical light absorption capability of BC-containing particles in Beijing during the campaign period. Another period is defined by PM_{10} concentrations of more than $\sim 120 \mu\text{g m}^{-3}$ and $r\text{BC}$ mass concentrations of more than $\sim 6 \mu\text{g m}^{-3}$, during which both the $\text{EEI}_{\text{total}}$ and O_3 concentrations showed slight changes with increasing air pollution levels. In this period,

the increase in calculated E_{ab} of BC-containing particles (from ~ 1.9 to ~ 2.0) might be attributed to local chemical production (e.g., heterogeneous reaction) in Beijing.

3.4 Implications for BC radiative forcing

The increase in BC light absorption capability with increasing air pollution levels suggests that greater solar absorption (i.e., direct radiative forcing, DRF) by atmospheric BC-containing particles occurs under more polluted conditions. The DRF of atmospheric BC-containing particles depends not only on the BC mass concentrations but also on the BC forcing efficiency, which strongly depends on the light absorption capability of BC-containing particles. In this study, the forcing efficiency of BC-containing particles was estimated based on a simple radiation transfer model (Eq. 9). Figure 7 shows that with increasing pollution levels (i.e., PM_{10} increasing from ~ 10 to $\sim 230 \mu\text{g m}^{-3}$) during the campaign period, the mass-averaged values of calculated MAC at 550 nm for BC-containing particles increased from ~ 11 to $\sim 14 \text{ m}^2 \text{ g}^{-1}$, which resulted in the SFE of BC-containing particles increasing from ~ 0.7 to $\sim 0.9 \text{ m}^2 \text{ g}^{-1} \text{ nm}^{-1}$. This revealed that the enhanced light absorption capability of

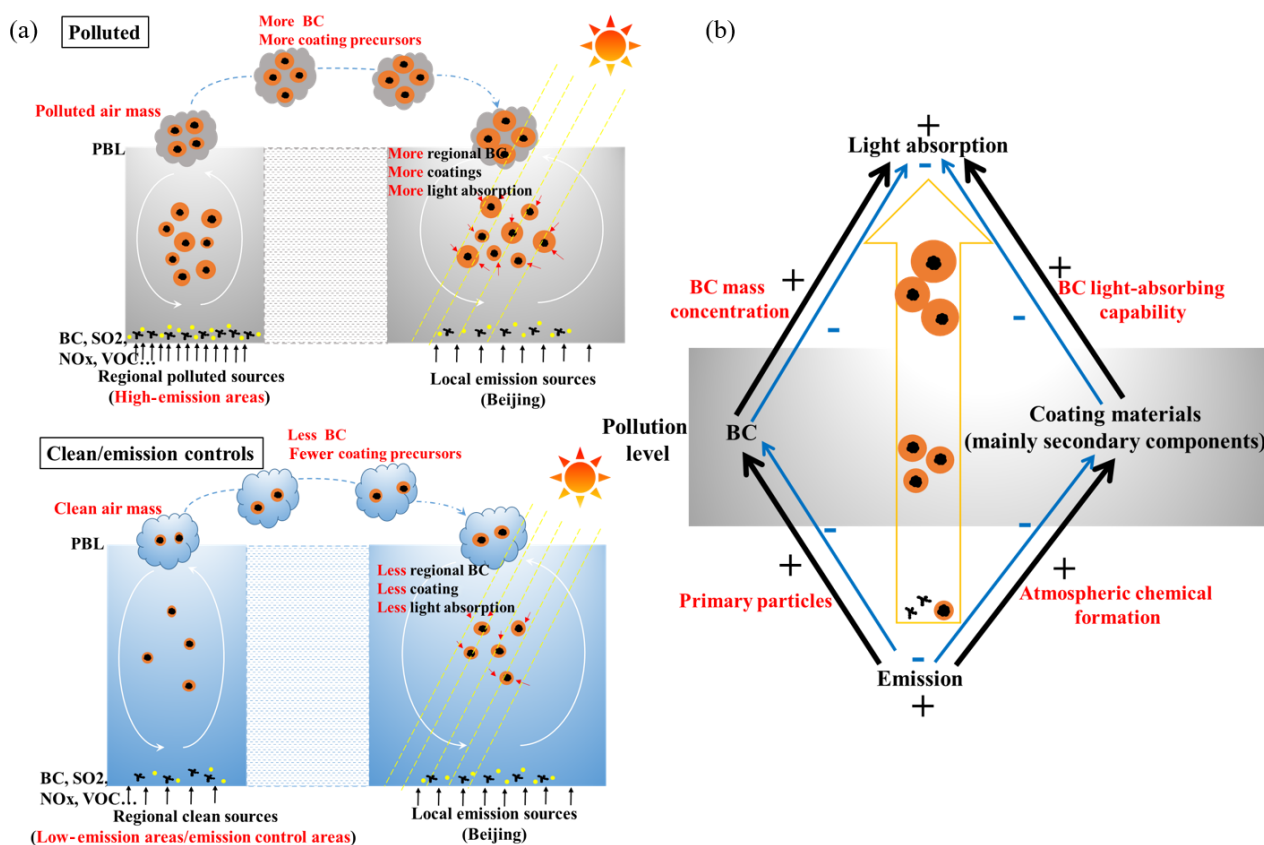


Figure 8. Conceptual scheme of the amplification effect on BC light absorption associated with air pollution.

BC-containing particles under a more polluted environment would increase their radiative forcing.

Considering the increase in both the light absorption capability ($\sim 18\%$) and mass concentrations (from ~ 0.7 to $\sim 11\ \mu\text{g m}^{-3}$) of BC when PM_{10} increased from ~ 10 to $\sim 230\ \mu\text{g m}^{-3}$, the DRF of BC-containing particles increased from ~ 0.3 to $\sim 6.4\ \text{W m}^{-2}$ (i.e., DRF_{with} shown in Fig. 7c, d). This revealed the importance of BC in terms of solar absorption under more polluted conditions. However, when the theoretical DRF of BC-containing particles was derived without considering the light absorption enhancement caused by coating materials (i.e., $\text{DRF}_{\text{without}}$ shown in Fig. 7c, d), it varied in the range of $0.2\text{--}3.3\ \text{W m}^{-2}$, which only depended on the mass concentrations of BC under different pollution levels. The DRF_{with} was significantly higher than $\text{DRF}_{\text{without}}$, identifying the amplification of DRF caused by coating materials on the BC surface. A more significant difference between DRF_{with} and $\text{DRF}_{\text{without}}$ was found under a more polluted environment, which was due to the increase in BC light absorption capability with increasing pollution levels. This indicated that the coating materials played a more important role in the DRF of BC under a more polluted environment.

The enhanced climate effects of BC aerosols in Beijing could be taken to be representative of polluted regions in China. Previous measurements of BC aerosols in China (Andreae et al., 2008; Huang et al., 2013; Wang et al., 2014b; Zhang et al., 2014; Zheng et al., 2015; Gong et al., 2016; Zhao et al., 2017) showed that the BC mass concentrations in different regions (e.g., Beijing, Xi'an, Nanjing, Shanghai and Guangzhou) reached values of $\sim 10\text{--}50\ \mu\text{g m}^{-3}$ during polluted periods (Table S2), similar to our measurements. Therefore, our BC aerosol observations in Beijing were not a special case. In China, high concentrations of BC aerosols under polluted conditions always occur on a regional scale due to intense BC emissions (Zhang et al., 2009; M. Li et al., 2017) and significant regional transport (Zhao et al., 2013; Wang et al., 2014a; Zheng et al., 2015). Our findings in Beijing can provide some implication in the difference of BC radiative forcing in other regions among different air pollution levels.

4 Discussion: breaking the amplification effect by emission control

Our results reveal that under a more polluted environment, the BC-containing particles are characterized by more BC

mass concentrations and more coating materials on the BC surface and therefore a higher light absorption capability. As shown in Fig. 8, this amplification effect on BC light absorption associated with air pollution is caused by increasing BC concentration and at the same time enhanced light absorption capability of BC-containing particles by more coating production in the more polluted air. Variation in both the mass concentration and light absorption capability of BC associated with air pollution strongly depends on the air pollutant emission (e.g., BC, SO₂, NO_x and volatile organic compounds). Under a polluted environment, polluted air mass from high-emission areas not only brings more BC but also more coating materials on the BC surface due to more precursors of secondary components.

Air pollution control measures may, however, break this amplification effect by reducing BC concentration and at the same time lowering the light absorption capability of BC-containing particles by slowing down the coating processes with cleaner air (Fig. 8). Take air pollution controls during the 2014 Asia-Pacific Economic Cooperation (APEC) meeting in Beijing, China, as an example; we found that as a result of emission controls on local Beijing and areas adjacent to Beijing (i.e., Hebei, Tianjin, Shanxi, Henan, Shandong and Inner Mongolia), light absorption of BC-containing particles decreased significantly during APEC compared to before APEC (Zhang et al., 2018). This is not only contributed by a reduction of BC mass concentration but also by lower light absorption capability of BC-containing particles with less coating materials on the BC surface in cleaner atmosphere conditions, indicating that synergetic emission reduction of multi-pollutants could achieve co-benefits for both air quality and climate.

5 Conclusions

The light absorption of BC-containing particles depends not only on the BC mass concentration but also on their light absorption capability (characterized by the calculated MAC and E_{ab} in this study). In this work, we investigated the difference of theoretical light absorption capability of BC under different air pollution conditions. During an intensive field measurement campaign in Beijing, China, we found that with increasing pollution levels, the increase in BC mass concentration was always accompanied by an increase in the theoretical light absorption capability of BC, resulting in an amplification effect on the light absorption of the ambient BC-containing particles. During the campaign period, the hourly values of mass-weighted averages of the D_p/D_c ratio and calculated E_{ab} for BC-containing particles was in the range of 1.5–2.3 and 1.5–2.0, respectively. When PM₁ concentration increased from ~ 10 to $\sim 230 \mu\text{g m}^{-3}$ accompanied with the rBC mass concentration in the range of ~ 0.7 – $11 \mu\text{g m}^{-3}$, the mass-weighted averages of the D_p/D_c ratio and calculated E_{ab} values increased by $\sim 33\%$ and $\sim 18\%$, respectively. The increase in BC light absorption capability asso-

ciated with increasing air pollution can be explained by the increase in coating materials on the BC surface under more polluted conditions. Moreover, the increase in the D_p/D_c ratio and calculated E_{ab} with increasing air pollution levels was size dependent; namely, more increase was exhibited for smaller rBC cores. This indicated that the aging degree and light absorption capability of smaller rBC was more sensitive to air pollution levels. Using a semiquantitative analysis method based on the diffusion-controlled growth law (Seinfeld and Pandis, 2006), we also calculated the theoretical increase in the D_p/D_c ratio and calculated E_{ab} with increasing pollution levels for BC-containing particles with size-resolved rBC cores. The agreement between the measured and theoretical increase in the D_p/D_c ratio and E_{ab} indicated the increase in coating materials on the BC surface with increasing PM₁ concentrations following the diffusion-controlled growth law.

The relationships between the changing rate of calculated E_{ab} ($k_{E_{ab}}$) with air pollution development and that of the PM₁ and rBC concentrations (k_{PM_1} and k_{rBC} , respectively) were estimated: $k_{E_{ab}} \approx 0.051 k_{\text{PM}_1}$ and $k_{E_{ab}} \approx 0.027 k_{\text{rBC}}$. During the campaign period, $k_{E_{ab}}$ values were in the range of -4 to 4 h^{-1} , with a peak of frequency distribution around zero, indicating that the BC-containing particles were shrinking as often as they were growing. The frequency distribution of the $k_{E_{ab}}/k_{\text{PM}_1}$ ratio showed a peak value around 0.05, revealing that the change of calculated E_{ab} was not sensitive to variations in PM₁ concentrations. Although the changing rate of the BC light absorption capability was significantly lower than that of the BC mass concentration, the effect of enhanced BC light absorption capability on the light absorption of ambient BC-containing particles under polluted conditions is not negligible. In our case, if we had not considered the increase in the theoretical light absorption capability of BC with increasing air pollution during the campaign period, the theoretical light absorption of BC-containing particles under polluted conditions would have been underestimated by $\sim 18\%$.

The increase in BC light absorption capability with increasing pollution levels in Beijing was controlled by aging during regional transport. The EEI analysis showed that $\sim 63\%$ of the BC observed at our site was transported from regional sources (i.e., areas outside of Beijing) during the polluted period, whereas the regional contributions were significantly lower ($\sim 21\%$) during the clean period. More BC in more polluted air from regional transport could lead to a higher local BC concentration in Beijing. Not only more BC but also more coatings are carried into Beijing by more polluted regional air mass (Fig. 8a), which can be explained by more coating precursors (e.g. SO₂, NO_x and volatile organic compounds) in more polluted air. Moreover, we separated the change of the D_p/D_c ratio and theoretical light absorption capability of BC associated with air pollution into regional-transport-controlled period (i.e., PM₁ $< 120 \mu\text{g m}^{-3}$ and BC $< 6 \mu\text{g m}^{-3}$) and local-chemistry-controlled periods

(i.e., $\text{PM}_{10} > 120 \mu\text{g m}^{-3}$ and $\text{BC} > 6 \mu\text{g m}^{-3}$). In the regional-transport-controlled period, the mass-averaged values of D_p/D_c and calculated E_{ab} of BC-containing particles in Beijing increased from ~ 1.6 to ~ 2.2 and from ~ 1.6 to ~ 2.0 , respectively, with increasing pollution levels. The further increase in mass-averaged values of D_p/D_c (~ 2.0 to ~ 2.2) and calculated E_{ab} (~ 1.9 to ~ 2.0) associated with air pollution is slower and is most likely attributed to local chemical production. Therefore, we attributed $\sim 75\%$ of the increase in theoretical light absorption capability of BC with increasing air pollution during the campaign period to aging during regional transport, demonstrating regional transport has an important influence on the variations in light absorption capability of BC-containing particles in Beijing under different pollution levels.

Due to the increase in BC light absorption capability with increasing air pollution levels, stronger forcing efficiency of the BC-containing particles was found under more polluted conditions. During the campaign period, the BC forcing efficiency increased by $\sim 18\%$ with PM_{10} increasing from ~ 10 to $\sim 230 \mu\text{g m}^{-3}$. Considering the increase in both BC forcing efficiency and BC mass concentration, the DRF values of ambient BC-containing particles could increase from $\sim 0.3 \text{ W m}^{-2}$ during the clean periods to $\sim 6.4 \text{ W m}^{-2}$ during the polluted periods. The results identified that BC in a more polluted environment exhibited a larger DRF, which was caused not only by the increase in BC mass concentrations but also by the enhancement of BC light absorption capability.

The amplification effect on BC DRF due to the increase in BC light absorption capability introduced in this work not only concerns Beijing but is also likely to operate in other polluted regions in China. The amplification effect could not only increase the direct contribution of BC to air pollution and climate change due to more light absorption but would also enhance the indirect contribution by stronger aerosol–meteorology and aerosol–climate feedbacks. Our finds in this work can provide some implication in the difference of BC-related effect on air quality and climate under different air pollution conditions (e.g., clean air and polluted environment) due to change in BC light absorption capability associated with air pollution.

The air pollution control may break the amplification effect by reducing BC concentration and at the same time lowering the light absorption capability of BC-containing particles by slowing down the coating processes with cleaner air. Therefore, breaking the amplification effect through emission controls would achieve a co-benefit effect with simultaneous mitigation of air pollution and climate change. Further study will focus on if and how emission reduction of BC and other pollutants in China will break the amplification effect.

Data availability. The observational data used in this study can be provided upon request to Yuxuan Zhang and Qiang Zhang (yuxuan.zhang@mpic.de and qiangzhang@tsinghua.edu.cn).

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Author contributions. YZ and QZ designed the research. YZ and HL performed the field measurements. YZ analyzed the data. YZ, QZ, YC and HS interpreted the data. YZ and QZ wrote the manuscript with input from all co-authors.

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

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