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Supplement of

Amplification of light absorption of black carbon associated with air pollution

Yuxuan Zhang et al.

Correspondence to: Qiang Zhang (qiangzhang@tsinghua.edu.cn)

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Figure S1. Location of the observation site (red star).

Figure S1 shows the geographic location of our observation site (namely red star marked in the Fig. S4). The site (40°00′17″ N, 116°19′34″ E) is located in megacity Beijing, the capital of China. The air pollution levels in our site can be influence by the air mass from adjacent regions (i.e., Tianjin, Hebei, Inner Mongolia, Shanxi, Shandong).
Figure S2. (a) The correlation between the absorption coefficient from AE33 at 660 nm ($\sigma_{ab,AE}$) and MAAP at 670 nm ($\sigma_{ab,MAAP}$). (b) Variety of multiple-scattering compensation factor $C$ with different Aethalometer measurements.

Aethalometer artefacts are mainly from the loading effect and multiple-scattering effect (Weingartner et al., 2003; Segura et al., 2014). In terms of the loading effect, the compensation algorithm has been incorporated into Aethalometer model AE33 (Drinovec et al., 2015). In this study, we focused on the multiple-scattering compensation, which was characterized by enhancement parameter $C$. The factor $C$ for our sites was determined by comparing the absorption coefficient derived from AE33 ($\sigma_{ab,AE}$) with the ones from MAAP ($\sigma_{ab,MAAP}$). Noted that the AE33 and MAAP measurements used to calculate the factor $C$ were at different wavelengths, namely 660 nm and 670 nm, respectively. Considering that the absorption is inversely proportional to wavelength (Bond and Bergstrom, 2006), the difference in wavelength would lead to an uncertainty of $\sim1.5\%$ for the corrected absorption coefficients in AE
measurement. As shown in Fig. S2a, the slope 2.6 was taken as the value of factor $C$ to compensate the Aethalometer data. The specifically site-calculated values of the factor $C$ varies in the range of 1.9-4 in this work (Fig. S2b), consistent with previous studies (Drinovec et al., 2015; Weingartner et al., 2003; Segura et al., 2014). In this study, the uncertainty in the factor $C$ was dominated by the uncertainty in MAAP measurements. We corrected the MAAP data using the algorithm reported by Hyvärinen et al. (2013). They estimated that the uncertainty in absorption coefficients derived from MAAP based on the developed algorithm was ~15% by comparing the results from a PAS against those derived from the MAAP in Beijing. This indicated that the factor $C$ used in our study (~2.6) would exhibit an uncertainty of ~15% from the uncertainty in MAAP measurements. Considering the uncertainty on the AE33 measurements was mainly from the factor $C$, the absorption coefficient from AE33 was estimated to have an uncertainty of ~15%.

Figure S3. SP2 detection efficiency of particle ($\eta$) in each rBC size-bin.

Figure S3 shows the SP2 detection efficiency concentration ($\eta$) in each rBC size-bin. In our study, the SP2 detection efficiency was determined with a DMA-SP2/CPC system. Monodispersed Aquadag particles generated by DMA were simultaneously measured by SP2 and CPC. The size-resolved $\eta$ was calculated by dividing the particle number concentration from SP2 measurement by that from CPC measurement. The SP2 detection efficiency (Fig. S3) have been considered in the
calculation of rBC mass concentration.

The $RI_s$ value used in this study are 1.50-0i based on the chemical compositions of coating materials during the campaign period. The components of coating materials was similar to non-refractory compositions in PM$_1$ particles (Peng et al., 2016). Figure S4 reveals that the fraction of inorganic and organic components in coating materials of BC-containing particles are ~51% and ~49%, respectively. It is known from the literature (Schkolnik et al., 2007; Mallet et al., 2003; Marley et al., 2001) that major inorganic components of ambient aerosol from urban emission (nitrate, sulfate, mineral dust, sea salt and trace metal) have a refractory of (1.5-1.6)-0i and there is a range of (1.4-1.5)-0i for the refractory of organic components. In this study, we used the values of 1.55-0i and 1.45-0i as refractive indexes of inorganic and organic components of coating materials. The refractive index of a mixture particle can be calculated as the volume weighted average of the refractive indices of all components (Hänel, et al. 1968; Marley et al., 2001; Bond and Bergstrom, 2006; Schkolnik et al., 2007), as $\bar{m} = \sum_i \bar{m}_i c_i$, where $\bar{m}$ is the refractive index of a mixture particle; $\bar{m}_i$ is the refractive index of particle species; $c$ is the volume ratio of particle species. Based on the equation, the refractive index of coating materials of BC-containing particles ($RI_s$) was ~1.50-0i during the campaign period.
Figure S5. The relative difference between the sizes of BC-containing particles ($D_p$) derived from Mie calculation with $RI_c$ of 2.26-1.26i and 1.95-0.79i. Various values (e.g., 2.26-1.26i, 1.95-0.79i) of refractive index of BC core ($RI_c$) have been used in literature (Bond and Bergstrom, 2006; Cappa et al., 2012; Taylor et al., 2015). Figure S5 shows the relative difference between the sizes of BC-containing particles ($D_p$) derived from Mie calculation with $RI_c$ of 2.26-1.26i and 1.95-0.79i. For BC-containing particles with 75-300 nm rBC cores, the relative difference is 3-10%, indicating that the $D_p$ values were not sensitive to $RI_c$ values in our study. This could be attributed to significantly larger in volume of coating materials than that of rBC cores.
Figure S6. Size distribution of refractory BC (rBC) as a function of the PM$_1$ concentration: (a) number size distribution and (b) mass size distribution.

Figure S6 shows the size distribution of rBC as a function of the PM$_1$ concentration. Above the detection limit of SP2 incandescence (rBC with size larger than ~75 nm), the number size distribution of rBC cores shows a peak at ~95 nm under different PM$_1$ concentration (Fig. S6 (a)), and there are about 95% of rBC particles in number concentration lower than 200 nm. As shown in Fig. S6 (b), the mass size distribution of rBC cores shows a wide mode at ~95-200 nm under different PM concentration.
Figure S7. The detect efficiency of SP2 scattering for BC-containing particles with size-resolved rBC cores (75-200 nm) under different PM$_1$ concentration. In this study, the detect efficiency of SP2 scattering in terms of BC-containing particles at a certain rBC core size is defined as the ratio of the number concentration of particles above the detection limit of SP2 scattering and total particles.

Figure S7 shows the detect efficiency of SP2 scattering for BC-containing particles with size-resolved rBC cores (75-200 nm) under different PM$_1$ concentration. For BC-containing particles above the detection limit of our SP2 incandescence (rBC cores larger than ~75 nm), the detect efficiency of SP2 scattering is defined as the ratio of the number concentration of particles above the detection limit of SP2 scattering and total particles. The SP2 scattering exhibited a high detection efficiency (90-100%) for observed BC-containing particles with rBC cores more than 75 nm, which could be attributed to large BC-containing particles (~180-500 nm shown in Fig. S8) in our site due to atmospheric aging. High detection efficiency of SP2 scattering is favor to retrieve the thickness of coating materials on rBC cores (>75 nm size studied in this work) based on scattering signal.
Figure S8. Frequency of the $D_p/D_c$ ratio of BC-containing particles with size-resolved rBC cores as a function of PM$_1$ concentrations.

Figure S8 shows frequency distribution of the $D_p/D_c$ ratio of BC-containing particles with size-resolved rBC cores under different PM$_1$ concentrations. For BC-containing particles with 75-300 nm rBC cores, their particle size was in the range of 180-500 nm. The particle size ($D_p$) of BC-containing particles with rBC cores at a certain size significantly increased with increasing PM$_1$ concentration, revealing more coating materials on BC surface under more polluted environment.

Figure S9. The time series of MAC derived from Mie calculation for BC cores (i.e., bare BC) at 880 nm.

In this study, we used Mie mode to calculate optical properties of all
BC-containing particles including bare BC and aged BC. Note that Mie theory is fundamentally ill-suited to calculation of optical properties for bare BC particles, which would lead to an uncertainty of their light absorption. Based on Mie calculation, we obtained the $MAC$ of rBC core ($MAC_c$) at 880 nm in the range of 3.8-4.5 m$^2$/g with an average of $\sim$4.3 m$^2$/g during the campaign period (Fig. S9). Bond and Bergstrom (2006) suggested a value of 7.5 m$^2$/g for the MAC of bare BC at 550 nm. Considering that the absorption is inversely proportional to wavelength (Bond and Bergstrom, 2006), the MAC of bare rBC at 880 nm is estimated to be $\sim$4.7 m$^2$/g, which was slightly greater than that ($\sim$4.3 m$^2$/g) obtained from Mie calculation in our study. This indicated the uncertainty of MAC for bare rBC from Mie calculation was $\sim$8%. We estimated that the uncertainties of calculated BC light absorption related to MAC of bare rBC from Mie calculation was $\sim$8%.

Figure S10. Variations in the coating thickness of BC-containing particles with the (a) PM$_1$ and (b) rBC mass concentrations.

Figure S10 shows the coating thickness of BC-containing particles increased with PM$_1$ and rBC concentration. The simultaneous increase in the rBC mass concentration and the amount of coating materials on the BC surface could significantly enhance the light absorption of BC-containing particles.
Figure S11. Changes of the light absorption coefficient at 880 nm ($\sigma_{ab,880nm}$) with PM$_1$ mass concentrations.

Figure S11 shows the changes of the light absorption coefficient at 880 nm ($\sigma_{ab,880nm}$) with PM$_1$ mass concentrations. The $\sigma_{ab,880nm}$ and rBC mass concentrations increased with increasing PM$_1$ mass concentrations. The simultaneous increase in the rBC mass concentration and the amount of coating materials shown in Fig. S10 revealed that the increase of $\sigma_{ab,880nm}$ (~18 fold from ~10 $\mu$g m$^{-3}$ of PM$_1$ to ~230 $\mu$g m$^{-3}$ of PM$_1$) could be attributed to simultaneous increase in the rBC mass concentration and the amount of coating materials on the BC surface.
Figure S12. (a) Changes of growth rate of calculated $E_{ab}$ ($k_{Eab}$) with PM$_1$ mass concentration. (b) Variations in the diameter of BC-containing particles ($D_p$) with the normalized PM$_1$ concentrations.

As shown in Figure S12, the changes of growth rate of $E_{ab}$ ($k_{Eab}$) decreased with increasing PM$_1$ mass concentration. During the campaign period, the $k_{Eab}$ of BC-containing particles was in the 0.5-1.5% h$^{-1}$. The decrease of $k_{Eab}$ associated with air pollution indicated the enhancement of light absorption capability of BC-containing particles slowed with further air pollution, because BC aging process by condensational growth was less effective for more-aged BC particles with larger size under more pollution environment (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 $k_{Eab}$ would also decrease.

Figure S13. Correlation between the growth rate of $E_{ab}$ ($k_{Eab}$) and the growth rates of rBC mass concentrations ($k_{rBC}$) during the campaign period.

Figure S13 shows the relationship between the change rate of calculated $E_{ab}$ ($k_{Eab}$) and the change rates of rBC mass concentrations ($k_{rBC}$) with pollution development. Linear relationships were estimated, i.e., $k_{Eab} = 0.027 \ k_{rBC}$. Compared with the values of $k_{rBC}$, the significantly smaller $k_{Eab}$ value indicated that the light absorption capability of BC increased more slowly than rBC mass concentrations.

Table S1 The DRF of externally mixed BC from global climate models. The modeled values were taken from Bond et al. (2013).

<table>
<thead>
<tr>
<th>Global climate Model</th>
<th>Mixing state</th>
<th>Modeled MAC (m$^2$ g$^{-1}$)</th>
<th>Modeled DRF (W m$^{-2}$)</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>AeroCom models</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>GISS</td>
<td>External</td>
<td>8.4</td>
<td>0.22</td>
<td>Schulz et al. (2006)</td>
</tr>
<tr>
<td>LOA</td>
<td>External</td>
<td>8.0</td>
<td>0.32</td>
<td>Schulz et al. (2006)</td>
</tr>
<tr>
<td>LSCE</td>
<td>External</td>
<td>4.4</td>
<td>0.30</td>
<td>Schulz et al. (2006)</td>
</tr>
<tr>
<td>SPRINTARS</td>
<td>External</td>
<td>9.8</td>
<td>0.32</td>
<td>Schulz et al. (2006)</td>
</tr>
<tr>
<td>UIO-CTM</td>
<td>External</td>
<td>7.2</td>
<td>0.22</td>
<td>Schulz et al. (2006)</td>
</tr>
<tr>
<td>UMI</td>
<td>External</td>
<td>6.8</td>
<td>0.25</td>
<td>Schulz et al. (2006)</td>
</tr>
<tr>
<td>Other models</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>BCC_AGCM</td>
<td>External</td>
<td>4.3</td>
<td>0.10</td>
<td>Zhang et al. (2012)</td>
</tr>
<tr>
<td>CAM3 ECA</td>
<td>External</td>
<td>10.6</td>
<td>0.57</td>
<td>Kim et al. (2008)</td>
</tr>
<tr>
<td>GISS-GCM II</td>
<td>External</td>
<td>7.8</td>
<td>0.51</td>
<td>Chung and Seinfeld (2002)</td>
</tr>
<tr>
<td>Average values</td>
<td></td>
<td>7.5</td>
<td>0.31</td>
<td></td>
</tr>
</tbody>
</table>

The DRF values for BC-containing particles at different pollution levels were
obtained by scaling the average DRF (0.31 W m$^{-2}$) of externally mixed BC from various climate models (Bond et al. 2013) with a scaling factor of the calculated $E_{ab}$ under different PM$_1$ concentrations (Fig. 2b). The DRF (0.31 W m$^{-2}$) of externally mixed BC was the global averages from the global climate models listed in Table S1. In order to point out the effect of BC light-absorption capability on DRF under different PM$_1$ concentrations, we did not consider the changes of BC amount for DRF calculation.
Table S2. Previous studies on the BC and PM (PM$_{1}$ or PM$_{2.5}$) mass concentrations in China.

<table>
<thead>
<tr>
<th>Site</th>
<th>Measurement Period</th>
<th>PM (μg m$^{-3}$)</th>
<th>BC (μg m$^{-3}$)</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Beijing (urban site)</td>
<td>1 to 31 January 2013</td>
<td>PM$_{2.5}$: $\sim$4.4-855 (mean: 162)</td>
<td>$\sim$0.2-25</td>
<td>Zheng et al., 2015</td>
</tr>
<tr>
<td>Xi’an, Shaanxi (urban site)</td>
<td>23 December 2012 to 18 January 2013</td>
<td>PM$_{2.5}$: $\sim$10-600 (mean: 8.8)</td>
<td>$\sim$0.3-44.5</td>
<td>Wang et al., 2014</td>
</tr>
<tr>
<td>Nanjing, Jiangsu (urban site)</td>
<td>1 January to 31 December 2015</td>
<td>PM$_{1}$: $\sim$10-250 (mean: 48)</td>
<td>$\sim$0.5-20</td>
<td>Zhao et al., 2017</td>
</tr>
<tr>
<td>Shanghai (urban site)</td>
<td>5 to 10 December 2013</td>
<td>PM$_{2.5}$: $\sim$40-636 (mean: 221)</td>
<td>$\sim$0.6-12.1</td>
<td>Gong et al., 2016</td>
</tr>
<tr>
<td>Jiaxing, Zhejiang (suburban site)</td>
<td>29 June to 15 July 2010</td>
<td>PM$_{1}$: Summer $\sim$4.6-104 (mean: 32.9)</td>
<td>Summer $\sim$0.4-11.7 (mean: 3.0)</td>
<td>Huang et al., 2013</td>
</tr>
<tr>
<td></td>
<td>11 to 23 December 2010</td>
<td>Winter 5.8-160 (mean: 41.9)</td>
<td>Winter $\sim$0.52-49.5 (mean: 7.1)</td>
<td></td>
</tr>
<tr>
<td>Guangzhou, Guangdong (urban site)</td>
<td>5 October to 5 November 2004</td>
<td>PM$_{2.5}$: $\sim$63-152 (mean:103)</td>
<td>$\sim$3-20</td>
<td>Andreae et al., 2008</td>
</tr>
<tr>
<td>Heshan, Guangdong (suburban site)</td>
<td>From 21 November to 1 December 2010</td>
<td>PM$_{2.5}$: 23.5-145.2 (mean: 74.6)</td>
<td>2.9-13.8</td>
<td>Zhang et al., 2014</td>
</tr>
</tbody>
</table>
Table S2 lists the BC and PM (PM\(_1\) or PM\(_{2.5}\)) mass concentrations in China in previous study. In this study, the PM\(_1\) and rBC concentrations were 10-230 \(\mu g \, m^{-3}\) and 0.7-11 \(\mu g \, m^{-3}\) in Beijing during the campaign period (17 to 30 November 2014), which was consistent with previous studies in other polluted regions in China (Table S2). The consistency indicated that the enhancement of the light absorption capability of BC-containing particles associated with air pollution not only occurred in Beijing but also might be observed in other polluted regions in China.

**References**


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