



## Radiative absorption enhancement from coatings on black carbon aerosols



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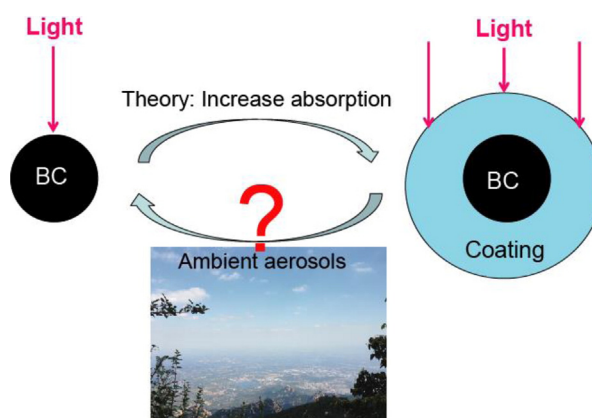
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### HIGHLIGHTS

- A method was developed to remove coatings surrounding BC in ambient aerosols.
- The MAC of decoated BC of 4.4 was enhanced to  $9.6 \text{ m}^2 \text{ g}^{-1}$  for ambient BC aerosols.
- BC radiative forcing in the ambient atmosphere was enhanced by a factor of ~2.
- BC absorption enhancement peaked in day time driven by secondary sulfate

### GRAPHICAL ABSTRACT



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### ABSTRACT

The radiative absorption enhancement of ambient black carbon (BC), by light-refractive coatings of atmospheric aerosols, constitutes a large uncertainty in estimates of climate forcing. The direct measurements of radiative absorption enhancement require the experimentally-removing the coating materials in ambient BC-containing aerosols, which remains a challenge. Here, the absorption enhancement of the BC core by non-absorbing aerosol coatings was quantified using a two-step removal of both inorganic and organic matter coatings of ambient aerosols. The mass absorption cross-section (MAC) of decoated/pure atmospheric BC aerosols of  $4.4 \pm 0.8 \text{ m}^2 \text{ g}^{-1}$  was enhanced to  $9.6 \pm 1.8 \text{ m}^2 \text{ g}^{-1}$  at 678-nm wavelength for ambiently-coated BC aerosols at a rural Northern China site. The enhancement of MAC ( $E_{MAC}$ ) rises from  $1.4 \pm 0.3$  in fresh combustion emissions to ~3 for aged ambient China aerosols. The three-week high-intensity campaign observed an average  $E_{MAC}$  of  $2.25 \pm 0.55$ , and sulfates were primary drivers of the enhanced BC absorption.

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## 1. Introduction

Black carbon (BC) is emitted from incomplete combustion of biomass and fossil fuel. The atmospheric aerosol BC contributes to one of the largest uncertainties in climate models with estimates of its global mean radiative absorption varying from a lower range of  $0.1\text{--}0.6\text{ W m}^{-2}$  (Allen et al., 2012; Boucher et al., 2013; Myhre et al., 2013; Schulz et al., 2006) to a strong effect of  $0.9\text{--}1.2\text{ W m}^{-2}$  (Ramanathan and Carmichael, 2008; Sato et al., 2003). Accurate assessments of BC radiative absorption require observational constraints of BC emissions, BC mass absorption cross-section (MAC), and enhancement of BC absorption after mixing with non-BC aerosol components in the atmosphere (Bond et al., 2013; Schulz et al., 2006). Non-absorbing matter co-emitted with BC or formed in the atmosphere may surround a BC core (as coating or internal mixing). Such non-absorbing coatings of BC potentially lead to an enhancement of MAC ( $E_{MAC}$ ) and thus the radiative absorption (Bond et al., 2006; Jacobson, 2001; Zhang et al., 2008).

At present, climate models vary widely in describing BC aerosols to be either externally or internally mixed, which leads to large uncertainties of  $E_{MAC}$ , spanning over the range 1–3 (Chung and Seinfeld, 2005; Jacobson, 2012; Schulz et al., 2006). To better constrain the current large diversities in description of BC radiative absorption in climate models, improved measurements of BC  $E_{MAC}$  in ambient aerosols are critical (Boucher et al., 2013). The direct measurement of  $E_{MAC}$  requires experimentally-removing the coating materials in ambient BC-containing aerosols (Bond et al., 2013). It is documented that BC coating materials consist mainly of sulfate, nitrate, ammonium and organic carbon (Moffet and Prather, 2009). Pioneering efforts have developed a heating denuder to evaporate high-volatility and semi-volatile coatings. A recent report using such a heating denuder method suggested negligible absorption enhancement of BC in ambient aerosols

in North America (Cappa et al., 2012), which contrasts to theoretical estimations of a factor of  $\sim 2$  (Jacobson, 2001). However, another research observed the absorption enhancement of a factor of 1.4 in ambient aerosols in U.K. using such a heating denuder method (Liu et al., 2015). These variations in observations underline the urgent need of independent methods/experiments to quantify  $E_{MAC}$ . The removal of coating matter from ambient BC aerosols remains the central challenge for the quantification of  $E_{MAC}$  and thus for an accurate modeling of the total BC-related aerosol warming in assessments of climate change (Boucher et al., 2013; Myhre et al., 2013).

We have developed an aerosol filter dissolution-filtration (AFD) system for a two-step removal of both inorganic and organic matter coatings of ambient aerosols. Samples of aerosol fine particulate matter ( $PM_{2.5}$ ) in North China were analyzed for MAC of BC before and after the coating removal to establish  $E_{MAC}$  values. The observations showed strong absorption enhancements by a factor of 2–3 for BC in North China  $PM_{2.5}$ . The  $E_{MAC}$  in fresh emissions of fossil-fuel combustion was a factor of  $\sim 1.4$ , increasing rapidly with sulfate abundance.

## 2. Materials and methods

### 2.1. Intensive campaign

The high-intensity sampling and monitoring campaign was achieved at Yucheng ( $36.83^\circ\text{N}$ ,  $116.57^\circ\text{E}$ ), a regional rural site in the center of the densely populated North China Plain (Fig. 1).  $PM_{2.5}$  aerosols were collected on pre-combusted quartz filters (90 mm in diameter, Pallflex, Tissuquartz 2500 QAT-UP) mounted on mid-volume ( $100\text{ L min}^{-1}$ ) atmospheric samplers (TH-150C-III, Wuhan Tianhong Instrument Co., Ltd, China). The Yucheng campaign ran sample collections during daytime (9:00 AM–17:00 PM) and night (17:30 PM–8:30 AM). In total 36  $PM_{2.5}$  samples were collected during three

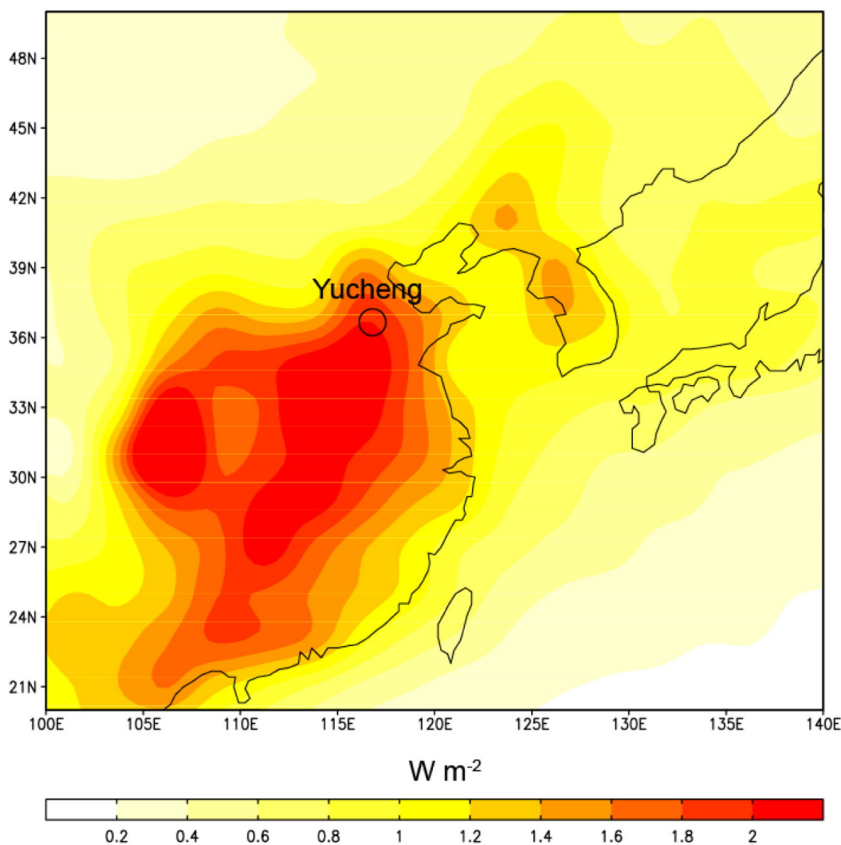


Fig. 1. Map showing the rural sampling site at Yucheng in Northern China Plain. BC radiative forcing in CMIP5 GISS simulation is color shaded (Schmidt et al., 2014).

weeks of June and July 2014. Three samples were collected in relative humidity of 100%. The high humidity wetted the original sample filter, and part of the coating materials may thereby have been removed before AFD solvent treatment. We therefore excluded these three samples from the statistical analysis.

The concentrations of water soluble inorganic ions ( $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$ ,  $\text{NH}_4^+$ ) in  $\text{PM}_{2.5}$  were measured by a model ADI 2080 online analyzer for Monitoring for Aerosols and Gases (MARGA, Applikon Analytical B.V., the Netherlands) at 1-h time resolution (Du et al., 2011). The ion concentrations were used to calculate the ratios:  $\text{SO}_4^{2-}/\text{EC}$ ,  $\text{NO}_3^-/\text{EC}$ ,  $\text{NH}_4^+/\text{EC}$ , to infer BC coatings and shell thickness. The sum of those ratios and OC/EC of bulk concentrations represent the total non-absorption particulate matter (NA-PM) coatings.

The effectiveness of the AFD method was tested by measurement of fresh emissions in diesel vehicle exhaust and tunnels. Five  $\text{PM}_{2.5}$  samples were directly collected at a diesel vehicle exhaust, and six samples were collected in the center of a 1.5 km long road tunnel.

## 2.2. Water and organic solvent treatment with AFD

The aerosol filter filtration-dissolution (AFD) method removes non-BC substances from the sample filters by two stages of dissolution mediated by water and organic solvents. Water dissolves sulfate, nitrate, ammonium, and water soluble organic carbon in filter aerosol samples. The remaining OC on the filter is removed with dichloromethane (DCM) and acetone mixtures (Rajput and Sarin, 2014).

A punch of 47 mm in diameter of filter sample was cut and fixed in a sand core funnel. A rubber tube was attached to the funnel outlet and blocked by water pressure. A volume of 50–200 ml (depended on the sample loadings of filters) distilled water was slowly injected into the funnel. The filter was immersed in water in the funnel for 30 min. Then, the rubber tube at the funnel outlet was slowly released of pressure, and water in the funnel gently passed through the sample filter and drained out. Another volume of 50–200 ml water was added in the funnel, and filtered out slowly. A small amount of ethanol was applied to remove remaining water in filter and funnel, and the AFD system was ready for next stage of organic solvent treatment.

The volume of 60 ml mixtures of dichloromethane (DCM) and acetone (1:1, v:v) was injected into the funnel, and held for 30 min before being filtered out. Small amounts of ethanol, then water, were employed to clean the funnel. To ensure slow and gentle filtration, care was taken during the release of water pressure at the funnel outlet rubber tube. Through the dissolution of organic and inorganic coatings, the solvent-decoated, and therefore pure BC remained on the sample filter. Finally, the treated filter in the funnel was dried at 60 °C. The solvent treatment and original sample filters were then stored in a desiccator before further analysis.

## 2.3. Determination of BC mass and optical properties

A thermal-optical transmittance (TOT) carbon analyzer (Sunset Laboratory, Tigard, OR, USA) was used to determine the elemental carbon (EC, as the mass-based proxy for optical BC) and organic carbon (OC) loadings on the filter. The analysis protocol was based on NIOSH870 (Panteliadis et al., 2015) that is similar to our earlier study (Chen et al., 2013) and others' works (Schauer et al., 2003; Subramanian et al., 2006).

The ECOC analyzer is equipped with a laser beam at wavelength 678 nm to measure the light-absorption of aerosols on the filter. The mass absorption cross-section (MAC) of samples before and after the AFD solvent treatment was determined following early studies (Cheng et al., 2011; Li et al., 2016; Ram and Sarin, 2009). The replicate analysis of original samples ( $n = 19$ ) showed uncertainties of EC and MAC were ~13% and ~10%, respectively.

The AFD treatment may change the BC/soot morphology. Changes in absorption upon morphological changes of BC are zero in the Rayleigh–

Debye–Gans (RDG) approximation (Radney et al., 2014). Light-absorbing OC – brown carbon (BrC) – may potentially interfere with the light-absorption by BC. However, BrC particles typically have a more pronounced wavelength-dependency, with significant light-absorption at shorter wavelengths (typically <400 nm). Thus, BrC is typically weakly absorbing (~5% of total aerosol absorption) or non-absorbing at the 678 nm used in the ECOC analyzer (Lack et al., 2012; Massabò et al., 2015; Yang et al., 2009).

## 2.4. Enhancement of mass absorption cross-section

The BC absorption enhancement ( $E_{\text{MAC}}$ ) in ambient aerosols was measured as the ratio between MAC of original ambient samples and that after AFD solvent-decoating treatment.

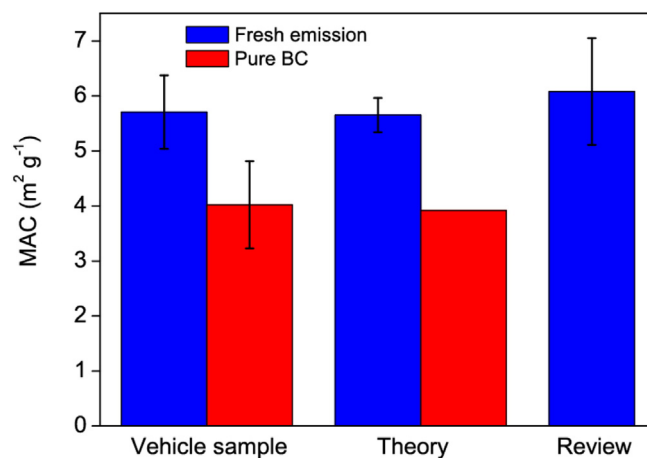
$$E_{\text{MAC}} = \frac{\text{MAC}_{\text{original}}}{\text{MAC}_{\text{treated}}} \quad (1)$$

Where,  $\text{MAC}_{\text{original}}$  and  $\text{MAC}_{\text{treated}}$  represent the MAC measurements of original ambient aerosol samples and subsequent solvent decoated pure BC, respectively.

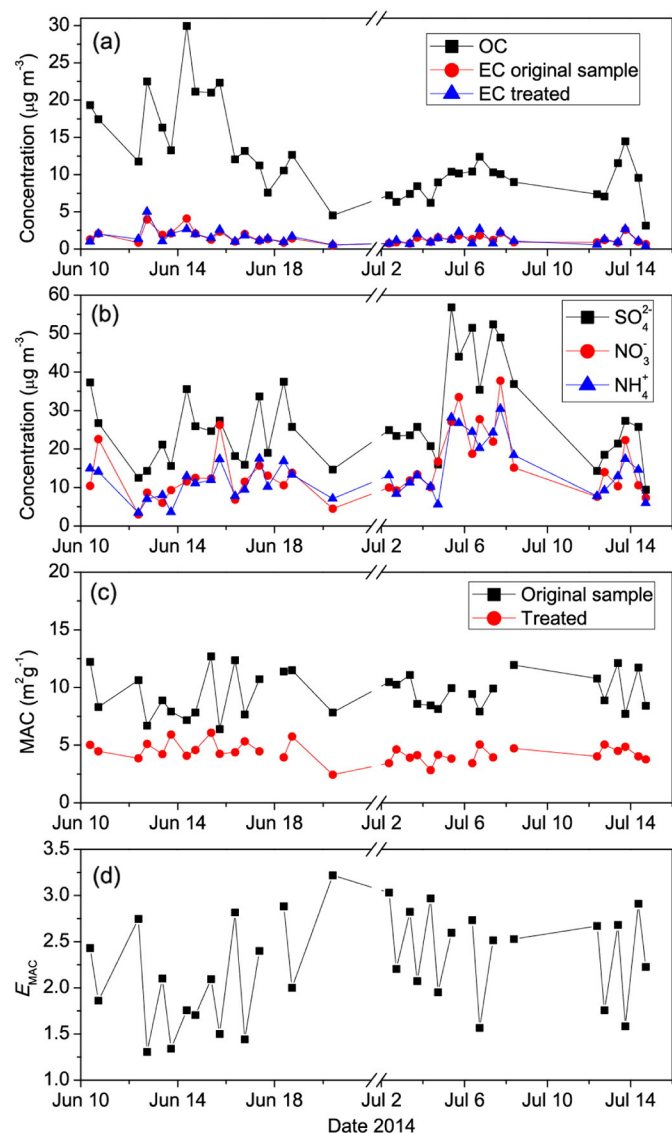
## 3. Results

### 3.1. Measurement of vehicle aerosols

The  $E_{\text{MAC}}$  measurements were tested by comparing with theoretical calculations for vehicle-emission aerosols. The vehicle samples (including diesel vehicle exhaust and road tunnel emissions) yielded MAC values of  $5.70 \pm 0.67 \text{ m}^2\text{g}^{-1}$  (average and standard deviation of 11 measurements) at 678-nm wavelength. This measurement agrees with theoretical calculations of MAC for fresh diesel soot ( $5.65 \pm 0.31 \text{ m}^2\text{g}^{-1}$ , Fig. 2) (Adler et al., 2010), also overlapping with measured ranges reported in literature reviews (Bond and Bergstrom, 2006; Bond et al., 2013). The subsequent AFD solvent treatments of the same vehicle samples lowered their MAC to  $4.02 \pm 0.79 \text{ m}^2\text{g}^{-1}$ . Such a MAC of vehicular BC, when stripped of associated coatings, is close to a classic



**Fig. 2.** Mass absorption cross-section (MAC) of vehicle samples for measurement in this study, theoretical calculations, and literature review. Left columns: the measured MAC of vehicle samples (Fresh emission) and their solvent/decoated pure BC isolated in this study. Central columns: the calculations of MAC of diesel soot using classic Rayleigh–Debye–Gans (RDG) theory on clusters of coated spherules (models 4 and 5 in Adler et al.) (Fresh emission), and on an aggregate of total radius  $R_g$  containing pure uncoated BC spherules of 10, 15, or 25 nm radius (model 2 in Adler et al.) (Adler et al., 2010). Right column: the measured values of freshly emitted fossil-fuel combustion suggested in the literature review (Bond and Bergstrom, 2006; Bond et al., 2013). The MAC of wavelength 532-nm in the theoretical calculation (Adler et al., 2010) and 550-nm in the literature review are adjusted to 678 nm using an Absorption Angstrom Exponent (AAE) of 1 following (Cheng et al., 2011).



**Fig. 3.** Temporal trends of aerosol concentrations and light absorption enhancement at the rural site in Northern China Plain. (a) Concentrations of OC and EC of original aerosol samples, and EC of AFD/solvent treatment. (b) Sulfate, nitrate, and ammonium concentrations. (c) Mass absorption cross-section (MAC) of original aerosol samples and after solvent treatment. (d) The enhancement of MAC ( $E_{\text{MAC}}$ ) from coating effect.

theoretical prediction of pure uncoated BC ( $3.92 \text{ m}^2 \text{g}^{-1}$ , Fig. 2) (Adler et al., 2010).

### 3.2. MAC for ambient samples before and after decoating treatment

The two-stage solvent treatments dissolved all water-soluble salts and ~90% organic carbon in the aerosol samples. The AFD system thus enables the decoating of BC, with the agreement of EC mass before and after the AFD treatment (Fig. 3a). The MAC for ambient untreated samples were  $9.58 \pm 1.83 \text{ m}^2 \text{g}^{-1}$  at 678 nm (Fig. 3c). In contrast, the AFD/solvent treatment, removing this ambient coating to leave the pure BC residue on the filter, decreased the MAC of the stripped/uncoated BC to a narrow range of  $4.38 \pm 0.79 \text{ m}^2 \text{g}^{-1}$ .

### 3.3. BC absorption enhancement

The AFD/solvent treatment thus offers a means to measure the MAC of ambient aerosol samples and their decoated pure BC, with the ratio of the two measurements representing the absorption enhancement

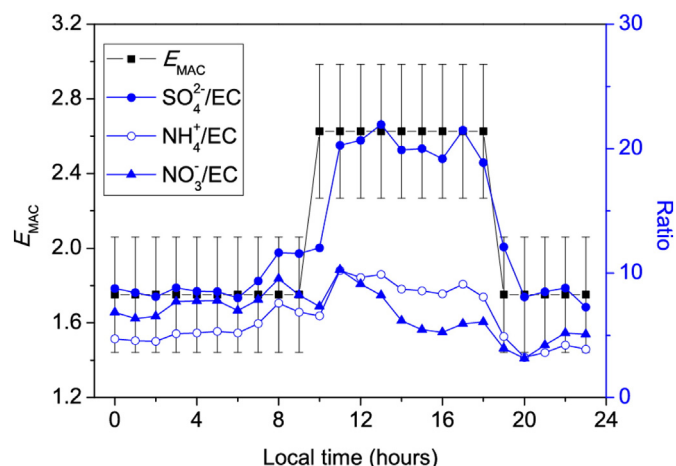
( $E_{\text{MAC}}$ ) of BC due to coatings in ambient aerosols. The  $\text{PM}_{2.5}$  samples at the rural North China site showed  $E_{\text{MAC}}$  factors of  $2.25 \pm 0.55$  (Fig. 3d). The MAC of decoated BC ( $4.38 \pm 0.79 \text{ m}^2 \text{g}^{-1}$ ) from solvent treatments is also lower than those for fresh emissions from fossil-fuel combustion ( $6.08 \pm 0.97 \text{ m}^2 \text{g}^{-1}$  adjusted to 678 nm) reported in the literature (Bond and Bergstrom, 2006; Bond et al., 2013), suggesting an enhancement of BC absorption in fresh emissions from co-emitted matter. This suggests that the  $E_{\text{MAC}}$  of BC due to co-emitted matter in the fresh combustion emissions is in the range of  $1.44 \pm 0.30$ . This range is also in agreement with the measured absorption enhancement ( $1.44 \pm 0.19$ ) from the fresh emissions of diesel vehicle exhaust and the tunnel samples of this study.

## 4. Discussion and conclusion

The changes in  $E_{\text{MAC}}$  primarily reflect the formation of coating aerosols. The increase in aerosol coating can be inferred from the ratio of non-absorbing particulate matter (NA-PM) relative to BC (or EC). The  $E_{\text{MAC}}$  showed high values in day time during which photochemical production of secondary aerosols such as sulfate and ammonium was enhanced (Fig. 4). An increase of  $E_{\text{MAC}}$  correlates well with the ratios  $\text{SO}_4^{2-}/\text{EC}$  ( $R^2 = 0.64$ ),  $\text{NH}_4^+/\text{EC}$  ( $R^2 = 0.52$ ), and  $\text{NA-PM}/\text{EC}$  ( $R^2 = 0.56$ ) (Fig. 5). The production of secondary sulfates and organic substances (Park et al., 2016; Wang et al., 2014) contributes to the BC absorption enhancement.  $E_{\text{MAC}}$  increases rapidly with increasing  $\text{SO}_4^{2-}/\text{EC}$  ratio up to a ratio of about 20, then the  $E_{\text{MAC}}$  approaches a flat trend at large  $\text{SO}_4^{2-}/\text{EC}$  ratios of 30–50 (Fig. 5a). This suggests that coatings on aged aerosols can increase the  $E_{\text{MAC}}$  to about a factor of three, which is twice that in fresh emissions from incomplete fossil fuel combustion.

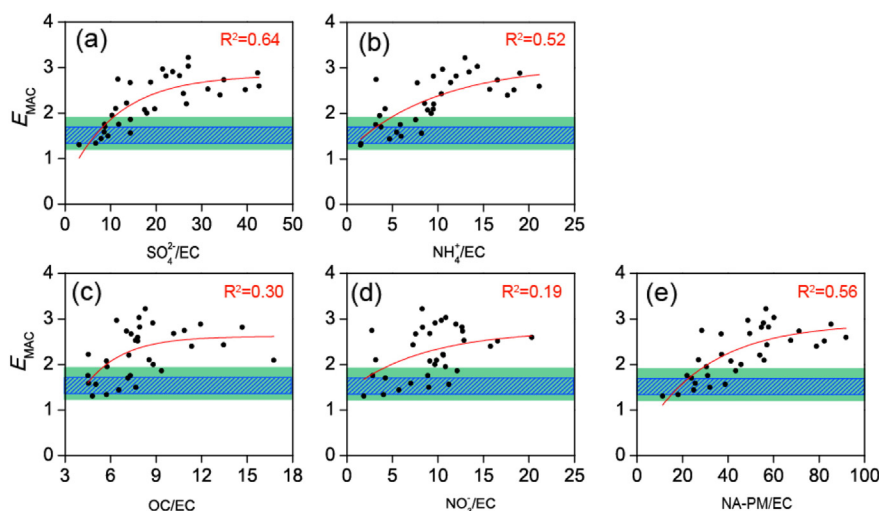
The result  $E_{\text{MAC}}$  of a factor of ~2 contrasts with a recent suggestion of BC absorption enhancements of only 6% in ambient aerosols in California (Cappa et al., 2012), and it is also larger than the enhancement factor (1.4) of U.K. observations (Liu et al., 2015). Previously, the direct measurements of  $E_{\text{MAC}}$  had relied on a thermal denuder (TD) of ~200 °C to evaporate volatile aerosols that may be ambient coatings on BC core. However, low-volatile coatings that remained after the TD heating could lead to biases in those previous efforts for ambient determination of BC radiative absorption enhancement.

The high levels of ambient BC in the atmosphere of East Asia have been apportioned to predominantly fossil fuel combustion (Chen et al., 2013). The strong emission of  $\text{SO}_2$  in fuel combustion in industrially-active regions (Kurokawa et al., 2013) causes BC mixing with abundant sulfate products of thick coatings (Yao et al., 2016), explaining the large absorption enhancement in populated and developed areas such as North China. The accounting for the factor of two



**Fig. 4.** Diurnal pattern of  $E_{\text{MAC}}$  compared with ratios between non-absorbing aerosols and EC.





**Fig. 5.** BC absorption enhancement ( $E_{MAC}$ ) as a function of ratios in non-absorbing matter vs. EC for PM<sub>2.5</sub> samples in Northern China Yucheng Campaign.  $E_{MAC}$  change with increase in  $SO_4^{2-}/EC$  (a),  $NH_4^+/EC$  (b),  $OC/EC$  (c),  $NO_3^-/EC$  (d), and total non-absorbing particulate matter (NA-PM)/EC (e). Green shading and blue pattern show the  $E_{MAC}$  of fresh emission of fossil fuel combustion and vehicles, respectively.

absorption enhancement of ambient BC may improve the climate model description of BC absorption of solar radiation. Such improvement in BC radiative estimates may have implications for several climate effects such as the currently underestimated rates of tropical expansion that is primarily driven by BC radiative forcing (Kovilakam and Mahajan, 2015).

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