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Radiative absorption enhancement from coatings on black carbon aerosols

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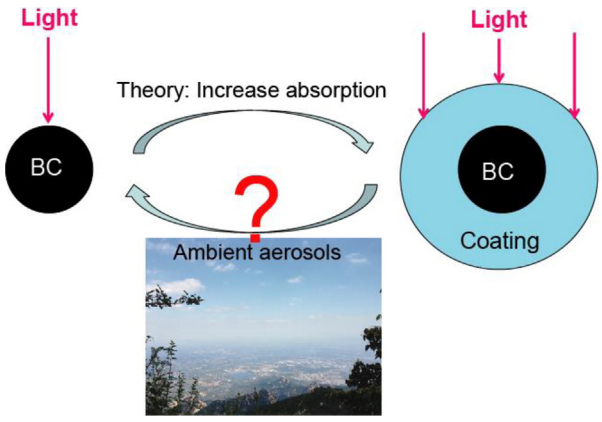
HIGHLIGHTS

- 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 m^2g^{-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 ± 0.8 m^2g^{-1} was enhanced to 9.6 ± 1.8 m^2g^{-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 ± 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 ± 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 0.1–0.6 W m^{-2} (Allen et al., 2012; Boucher et al., 2013; Myhre et al., 2013; Schulz et al., 2006) to a strong effect of 0.9–1.2 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 ~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 ~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°N, 116.57°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 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

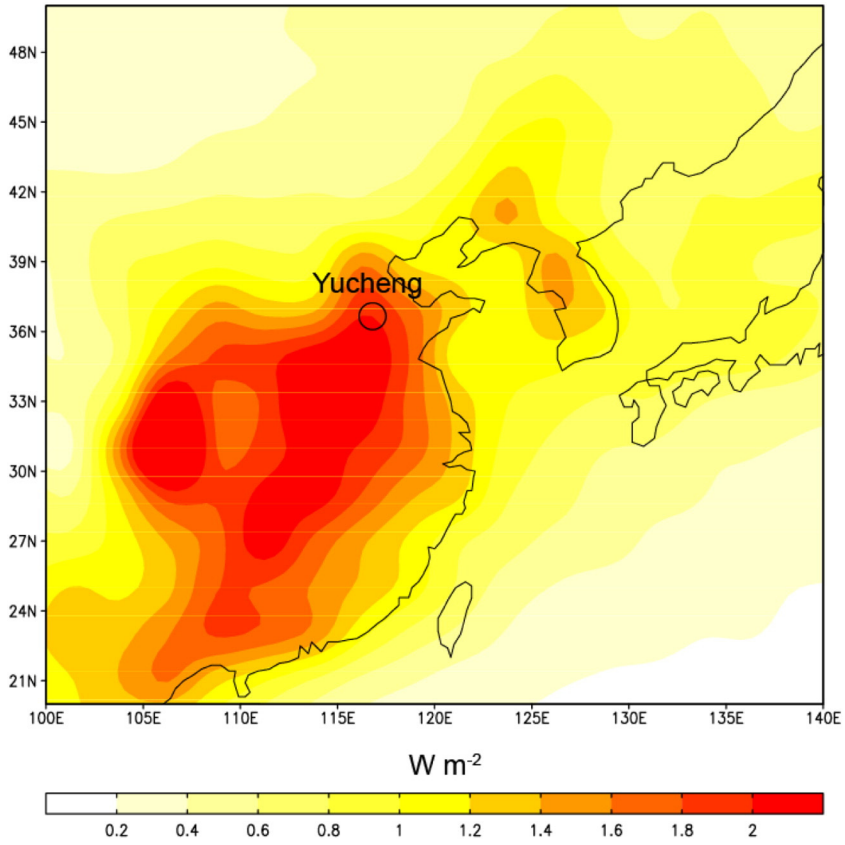

Fig. 1. Map showing the rural sampling site at Yucheng in Northern China Plain. BC radiative forcing in CMIP5 GISS simulation is color shaded (Schmidt et al., 2014).