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Light absorption enhancement of black carbon from urban haze in Northern China winter☆

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ABSTRACT

Atmospheric black carbon (BC) is an important pollutant for both air quality and Earth's energy balance. Estimates of BC climate forcing remain highly uncertain, e.g., due to the mixing with non-absorbing components. Non-absorbing aerosols create a coating on BC and may thereby act as a lens which may enhance the light absorption. However, this absorption enhancement is poorly constrained. To this end a two-step solvent dissolution protocol was employed to remove both organic and inorganic coatings, and then investigate their effects on BC light absorption. Samples were collected at a severely polluted urban area, Jinan, in the North China Plain (NCP) during February 2014. The BC mass absorption cross-section (MAC) was measured for the aerosol samples before and after the solvent-decoating treatment, and the enhancement of MAC (E_{MAC}) from the coating effect was defined as the ratio. A distinct diurnal pattern for the enhancement was observed, with E_{MAC} 1.3 ± 0.3 (1 S.D.) in the morning, increasing to 2.2 ± 1.0 in the afternoon, after that dropping to 1.5 ± 0.8 in the evening-night. The BC absorption enhancement primarily was associated with urban-scale photochemical production of nitrate and sulfate aerosols. In addition to that, regional-scale haze plume with increasing sulfate levels strengthened the absorption enhancement. These observations offer direct evidence for an increased absorption enhancement of BC due to severe air pollution in China.

1. Introduction

Atmospheric black carbon (BC) or soot in fine particulate matter ($PM_{2.5}$) is emitted from incomplete combustion of fossil fuel or biomass/bio-fuel. The scientific confidence of BC health risks recently has advanced; BC and organic carbon may be the top toxic pollutants in $PM_{2.5}$ that leads to annually ~3 million premature deaths (Apte et al., 2015; Lelieveld et al., 2015; Lim et al., 2012b). On the other hand, BC-induced climate warming, including regional effects, remains highly uncertain, particularly in the key emission regions such as China and India (Andreae and Ramanathan, 2013; Boucher et al., 2013). China aims at reducing the $PM_{2.5}$ mass concentrations up to 25% by 2017 (Chinese-State-Council, 2013). These $PM_{2.5}$ reduction measures will also reduce BC emissions from fossil fuel combustion and residential energy use such as heating and cooking (Chen et al., 2016b; Ding et al., 2016). However, an accurate prediction of BC radiative forcing is required to assess environmental and climate benefits of the China $PM_{2.5}$ reduction.

It is estimated that BC might be second only to CO_2 regarding climate warming effects, although the uncertainties for BC radiative forcing are much larger (Bond et al., 2013; Ramanathan and Carmichael, 2008; Sato et al., 2003). Meanwhile, the model simulations of BC absorption are generally lower than ground and satellite-based observations. Poorly constrained effects for BC absorption enhancement due to aerosol-mixing states in ambient aerosols have been proposed as a potential key factor for the model-observation discrepancy (Cappa et al., 2012; Fierce et al., 2016; Peng et al., 2016). Sensitivity tests suggest that mixing state and particle size of BC aerosols may be the most important factor

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governing climate model estimates (Boucher et al., 2013; Ma et al., 2012). Climate models predict that a non-absorbing coating on a BC core could enhance the light absorption of BC (E_{MAC}) by a factor of 2 (Jacobson, 2001). Such model predictions have yet to be clearly verified from ambient observations (Cappa et al., 2012). It is called for the establishment of methods that directly determine the BC E_{MAC} of ambient aerosols (Bond et al., 2013; Gustafsson and Ramanathan, 2016).

To estimate coating effects, a Thermodenuder (TD) based method was recently developed, which may remove coating materials through evaporating high-volatility and semi-volatile components from the aerosol phase. The absorption enhancement of BC is then directly determined by comparing the light absorption before and after the TD heating on ambient aerosols. Several field campaigns using the TD decoating method suggest that the BC absorption enhancement from ambient coatings considerably varied from 1 to 1.5 (Cappa et al., 2012; Lack et al., 2012a, 2012b; Liu et al., 2015). However, the TD decoating method might not be efficient in removing, e.g., low-volatile organic carbon and salts (Thornberry et al., 2010).

Meanwhile, an aerosol filter dissolution-filtration (AFD) system has recently been established, including a two-step removal of both inorganic and organic matter coatings for ambient aerosols (Cui et al., 2016). The enhancement of mass absorption cross-section (MAC) of BC is defined as the ratio of the MACs before and after the AFD decoating treatment, herein E_{MAC}. In this study, we employed the AFD method to $PM_{2.5}$ filter samples collected, along with online aerosol and gas measurements, at an urban area, Jinan, in Northern China during February 2014.

2. Methods

2.1. Study area

The study area, Jinan, is located in the middle of the Northern China Plain (NCP) where severe air pollution shows a regional coverage (Fig. 1). The $PM_{2.5}$ monitoring in 190 cities in China suggests that the top worst 20 cities are located in the Shandong Province (nine cities), the Hebei Province (eight cities), and the Henan Province (three cities). All these urban hot spots are located in the middle of NCP (Zhang and Cao, 2015). The $PM_{2.5}$ pollution, which is intensified during winter season, may at least in part be explained by strong emissions of gaseous and particulate pollutants such as SO_2, NO_x, VOC, and BC from industrialization and urbanization in this densely populated area (Fig. S1).

Jinan, the capital of Shandong Province, is a megacity with a population of 7.0 million. The annual mean $PM_{2.5}$ in Jinan is 85 μg m^{-3} in 2014/2015 (Zhang and Cao, 2015), which is higher than "the national ambient air quality standards" (NAAQS): 15 μg m^{-3} and 35 μg m^{-3} for Category I and II zones, respectively. The concentrations of sulfate, nitrate, and ammonium observed in Jinan are comparable to those of Beijing, Tianjin, Shanghai, and Guangzhou (Fig. S2) (Cao et al., 2016; Wu et al., 2016).

2.2. $PM_{2.5}$ samples

A two-week intensive campaign in 16–28 February 2014 was conducted in the urban area of Jinan. $PM_{2.5}$ was sampled at the Shandong University Monitoring Station (SDUMS, 36.67°N, 117.06°E), about 30 m above ground level at the urban center. Three samples per day were collected: during morning (7:00 a.m. - 13:00 p.m.), afternoon (13:30–17:30 p.m.), and evening-night (18:00 p.m. - 6:30 a.m.). The particles were collected on pre-combusted quartz filters (90 mm in diameter, Pallflex, Tissuquartz 2500 QAT-UP) mounted on a mid-volume (100 L min^{-1}) atmospheric sampler.

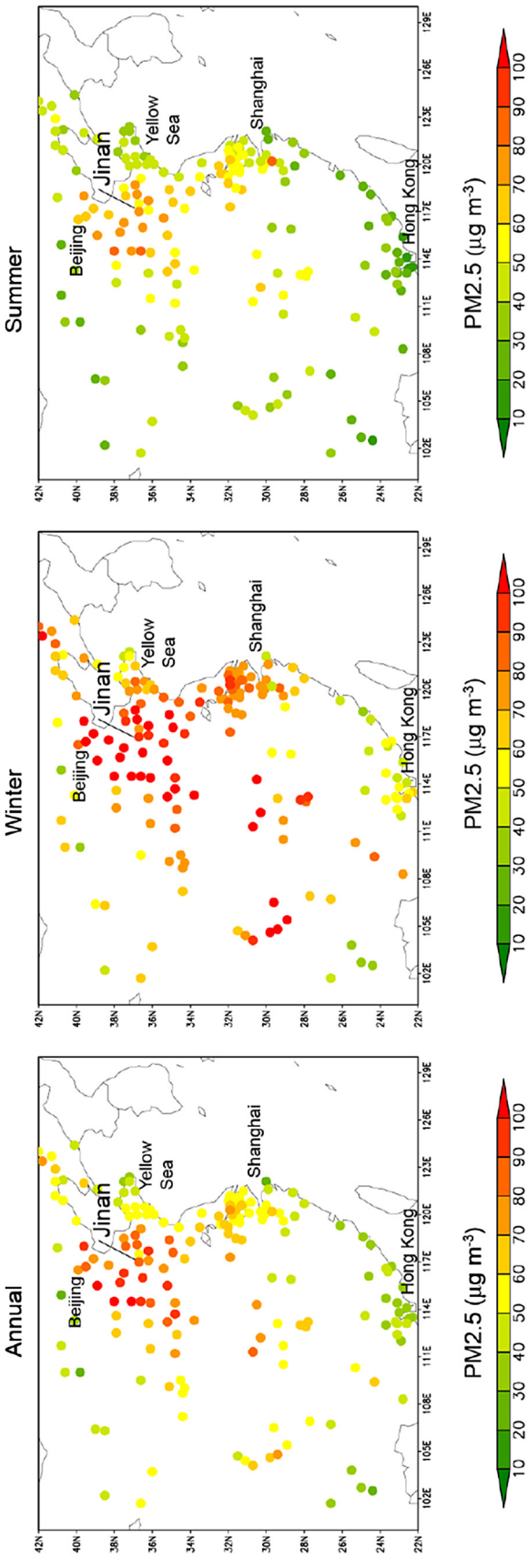

Fig. 1. The sampling site at an urban area, Jinan, in the Northern China Plain (NCP). $PM_{2.5}$ concentrations in China are shown in color plots. The annual, winter, and summer averages of monitored $PM_{2.5}$ (hourly resolution) in 2014/2015 are extracted for total 190 cities (Zhang and Cao, 2015).