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## Impact of size distributions of major chemical components in fine particles on light extinction in urban Guangzhou

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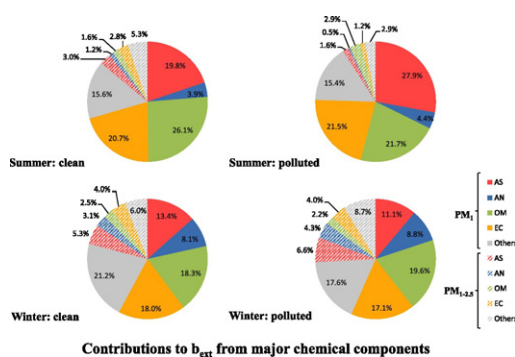
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### HIGHLIGHTS

- Aerosol chemical components in PM<sub>1</sub> and PM<sub>1–2.5</sub> were compared in two seasons.
- The size distributions of chemical components varied with pollution levels.
- Contributions of chemical components in PM<sub>1</sub> and PM<sub>1–2.5</sub> to light extinction were quantified.

### GRAPHICAL ABSTRACT



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### ABSTRACT

To evaluate the impact of fine particulate matter (PM<sub>2.5</sub>) size distribution on aerosol chemical and optical properties, dominant chemical components including water-soluble inorganic ions (WSII), organic carbon (OC) and elemental carbon (EC) in PM<sub>1</sub> and PM<sub>2.5</sub>, aerosol scattering coefficient ( $b_{sp}$ ), and aerosol absorption coefficient ( $b_{ap}$ ) were collected synchronously at an urban site in Guangzhou, south China during a typical summer month in 2009 and a winter month in 2010. PM<sub>1</sub> (sizes smaller than 1  $\mu\text{m}$ ) constituted 77% and 63% of PM<sub>2.5</sub> in summer and winter, respectively. From the reconstructed mass concentrations, the sum of  $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$  and  $\text{NH}_4^+$  (SNA) distributed more in PM<sub>1</sub> than in PM<sub>1–2.5</sub> (PM<sub>2.5</sub> minus PM<sub>1</sub>) in summer and the opposite was found in winter, while carbonaceous aerosols distributed more in PM<sub>1</sub> in both summer and winter. With the aggravation of PM<sub>2.5</sub> pollution, the mass fraction of PM<sub>1</sub>/PM<sub>2.5</sub> increased for  $(\text{NH}_4)_2\text{SO}_4$  (AS),  $\text{NH}_4\text{NO}_3$  (AN) and EC but decreased for organic matter (OM) in summer, and the opposite was found in winter.  $B_{sp}$  of PM<sub>1</sub> and PM<sub>1–2.5</sub> was estimated from the mass extinction efficiencies (MSEs) of the dominant chemical components, which showed good correlations ( $R^2 = 0.99$ ) with measured ones and those estimated

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using the IMPROVE formula. The fractional contributions of dominant chemical components to extinction coefficient ( $b_{\text{ext}}$ ) were consistent with their respective mass size distributions, indicating the importance of chemically-resolved aerosol size distributions on aerosol optical properties and haze formation.

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## 1. Introduction

High levels of particulate matter, especially  $\text{PM}_{2.5}$  (aerodynamic diameter  $< 2.5 \mu\text{m}$ ) have detrimental impacts on air quality and visibility, and thus human health, besides their roles on climate impact and ecosystem health (Samet et al., 2000; Malm et al., 2003; Ramanathan and Feng, 2009). The frequent hazy days occurred in recent years across China have spurred numerous investigations (Cheung et al., 2005; Zhang et al., 2013; Huang et al., 2014; Che et al., 2015a). Haze intensity can be characterized by the sum of aerosol light scattering and absorption (Watson, 2002), both strongly dependent on aerosol chemical components and associated size-distributions (Cozic et al., 2008; Lin et al., 2013). Multivariable regression models and empirical formulas are commonly used for investigating the relationships between aerosol optical properties and  $\text{PM}_{2.5}$  components. Secondary inorganic aerosols (SIA), OM and EC in  $\text{PM}_{2.5}$  have been identified as the dominant contributors to light extinction (Hand and Malm, 2007). However, these chemical species are mainly distributed in  $\text{PM}_1$  (Aiken et al., 2009; Sun et al., 2011), and the MSEs of submicron particles with diameters close to wavelength of visual light are much larger than those of the coarse particles (Bergin et al., 2001; Cheng et al., 2008; Che et al., 2015b). It is important to quantify the contributions of  $\text{PM}_1$  on the total aerosol optical properties and its role on haze formation.

The Pearl River Delta (PRD) region is one of the three megacity clusters in China, where have been frequently suffering haze weather in recent years (Wang et al., 2003; Andreae et al., 2008; Deng et al., 2008). Field experiments have been conducted in PRD to identify the causes of haze (Hu et al., 2008; Zhang et al., 2010; Yue et al., 2010; Tao et al., 2017). In some studies, the Mie Model and the IMPROVE formula were used to identify major chemical components contributing to light extinction (Liu et al., 2009b; Tao et al., 2014). Most of these studies focused on the physical and chemical characteristics of aerosols and their relationships with optical properties. Studies explored the size fractions of PM have also been conducted in PRD (Cheng et al., 2008; Jung et al., 2009); however, few studies have explored the relative contributions of  $\text{PM}_1$  and  $\text{PM}_{1-2.5}$  on haze formation.

To fill this knowledge gap, chemically-resolved  $\text{PM}_1$  and  $\text{PM}_{2.5}$ ,  $b_{\text{sp}}$ , and  $b_{\text{ap}}$  were collected at an urban site in Guangzhou during a summer month in 2009 and a winter month in 2010. The size distributions of major chemical components were first briefly characterized in Section 3.1, followed by discussions on their differences under different pollution levels (Section 3.2). Chemical factors contributing to  $b_{\text{ext}}$  were then quantified in Section 3.3.

## 2. Methodology

### 2.1. Site description

Measurements were collected at the monitoring station of the South China Institute of Environmental Science (SCIES) (23.12° N, 113.35° E) located in an urban area of Guangzhou. All instruments used in this study were installed on the roof of a building 50 m above the ground (Tao et al., 2014). The land is generally flat and there is no obvious industrial pollution source around the station. The data observed at this site largely represented air pollution levels in Guangzhou.

### 2.2. Sampling and chemical analysis

Daily (24 h from 10:00 am to 9:30 am the next day, local time)  $\text{PM}_1$  and  $\text{PM}_{2.5}$  samples were synchronously collected using particulate samplers (BGI Incorporated, Model PQ200) in a summer (July 1st to July 31st, 2009) and a winter (January 1st to January 31st, 2010) months. Samples were collected on 47 mm quartz filter (Whatman OM-A) with a flow rate of  $16.7 \text{ L min}^{-1}$ , baked at  $800 \text{ }^\circ\text{C}$  for at least 3 h and equilibrated in desiccators for 24 h in advance. The exposed filter was stored in a freezer at minus  $20 \text{ }^\circ\text{C}$  to prevent particle volatilization.

A punch of  $0.5 \text{ cm}^2$  from each quartz filter was analyzed for eight carbon fractions following the IMPROVE TOR protocol by Desert Research Institute (DIR) model 2001 carbon analyzer (Atmoslytic Inc. Calabasas, CA) (Chow et al., 2004, 2007; Cao et al., 2007). Eight inorganic ions ( $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$ ,  $\text{Cl}^-$ ,  $\text{Na}^+$ ,  $\text{NH}_4^+$ ,  $\text{K}^+$ ,  $\text{Mg}^{2+}$  and  $\text{Ca}^{2+}$ ) in aqueous extracts of the filter were determined by an ion chromatography (Dionex Corp, Sunnyvale, CA, Model Dionex 600).

### 2.3. Measurements and corrections of aerosol optical properties

$b_{\text{sp}}$  was measured at three wavelengths (450, 550 and 700 nm) with an integrating nephelometer (TSI Performance Measurement Tools, Model 3563) equipped with a conventional total suspended particulate (TSP) cyclone. The flow rate was set at  $20 \text{ L min}^{-1}$  for drawing air through a temperature controlled inlet, which controlled the relative humidity (RH) of inflow air to be  $< 70\%$  to minimize the influence of water vapor. Nephelometer calibration was carried out by carbon dioxide ( $\text{CO}_2$ ) as a high span gas and by filtered air as a low span gas. The output data were set to be 1 min average, and the baseline data were measured hourly. The raw data of  $b_{\text{sp}}$  have been corrected for truncation and non-Lambertian illumination errors according to Anderson and Ogren (1998).

$b_{\text{ap}}$  was measured using a 7-channel aethalometer (Magee Scientific Company, US, Model AE31) which is used for real time measurement of black carbon (BC). The aethalometer was equipped with a conventional total suspended particulate (TSP) cyclone and the flow rate was set at  $5 \text{ L min}^{-1}$ . The aethalometer was calibrated to zero by replacing the filter in the canister inlet with a clean filter every week. In this study, a direct method proposed by Weingartner et al. (2003) was used to obtain  $b_{\text{ap}}$  with correction parameters based on Wu et al. (2012, 2013).

### 2.4. Data analysis methods

#### 2.4.1. Reconstruction of PM mass

To evaluate whether the determined chemical components represent the measured  $\text{PM}_1$  and  $\text{PM}_{1-2.5}$ , the measured  $\text{PM}_1$  and  $\text{PM}_{2.5}$  masses were reconstructed based on AS ( $1.375 \text{ SO}_4^{2-}$ ), AN ( $1.29 \text{ NO}_3^-$ ), OM, and EC. The converting factor between OM and OC was chosen as 1.6 (Feng et al., 2009; Zhang et al., 2013). Unidentified chemical species, termed as the "Other" here, can be estimated by subtracting the sum of the determined chemical species mentioned above from the measured  $\text{PM}_1$  or  $\text{PM}_{2.5}$  mass concentrations.  $\text{PM}_{1-2.5}$  was obtained by subtracting  $\text{PM}_1$  from  $\text{PM}_{2.5}$ .

#### 2.4.2. Estimation of $b_{\text{sp}}$ of $\text{PM}_1$ and $\text{PM}_{1-2.5}$

The reconstructed chemical components in Section 2.4.1, which were also the major contributors to  $b_{\text{sp}}$ , were used to estimate  $b_{\text{sp}}$  of  $\text{PM}_1$  and  $\text{PM}_{1-2.5}$ . An external mixing of individual species was assumed in the analysis. Due to the lack of information on the size distributions of the chemical components, the densities and MSE curves of the major

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