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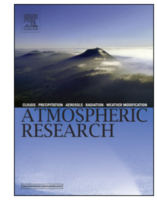

Characterization of chemical compositions in size-segregated atmospheric particles during severe haze episodes in three mega-cities of China

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ABSTRACT

To investigate the characterization of chemical compositions in size-segregated particles during severe haze pollution episodes in different regions of China, a campaign was conducted in Tianjin, Hangzhou and Chengdu. Size-segregated particles were collected with eight-stage Anderson cascade impactor in these cities in winter respectively. Ten major compositions of particles including (Na^+, NH_4^+, K^+, Mg^{2+}, Ca^{2+}, Cl^-, NO_3^-, SO_4^{2-}, OC and EC) were analyzed. A similar bimodal distribution of particles was found between northern and southern cities peaked at 0.7–2.1 and 9.0–10.0 μm. OC, EC, SO_4^{2-}, NO_3^-, Cl^- and NH_4^+ were the major chemical compositions of fine-mode particles, whereas OC, EC, SO_4^{2-}, NO_3^- and Ca^{2+} were the major compositions of coarse-mode particles. In the three cities, Cl^-, SO_4^{2-}, NO_3^-, NH_4^+ and K^+ of all compositions were unimodal distributions peaked at 0.7–2.1 μm. Different sources to particles in the three cities were inferred based on the size distribution characteristics of chemical compositions. For Tianjin, the influence of sea salt was greater to Hangzhou and Chengdu based on the concentrations and distributions of Na^+ and the Cl^-. Fine-mode Cl^- and SO_4^{2-} were highest in Tianjin, meaning the greater contribution of coal burning to particles during severe pollution. For Hangzhou, the NO_3^- concentration was higher than Tianjin and Chengdu. Contribution of nitrate to PM was higher than that of sulfate. For Chengdu, carbonaceous species contributed mostly to fine particles. However, sulfate and nitrate contributed mostly to fine particles in Tianjin and Hangzhou. The contributions of EC and Ca^{2+} to coarse-mode particles was much higher than that in other cities, implying the greater influence of soil particles, construction dust or falling dust to PM in Chengdu. In addition, there were greater emission source of NH_4^+ in Chengdu. Northwest, west and southeast were the major transport pathways of air masses for Tianjin, Hangzhou and Chengdu.

1. Introduction

Severe haze pollution has been recognized as one of the most serious environmental problems in Chinese megacities over the past decade (Wang et al., 2015a, 2015b; Wu et al., 2005; Tian et al., 2016; Fu et al., 2013). Frequent occurrences of haze have a detrimental effect on human health, traffic safety, and climate change (Pope and Dockery, 2006; Tie et al., 2009; Yang et al., 2015; Zhang et al., 2015a, 2015b; Yu et al., 2002). Atmospheric particulate matter (PM) plays an important role in the formation of haze which strongly depends on the concentration, chemical composition, size distribution and mixed state (Xiang et al., 2017; Contini et al., 2014; Tan et al., 2016; Zhao et al., 2011). The formation, transformation and removal mechanisms of particles are different in different mode (Whitby, 1978; Zhao et al., 2011; Hering and Friedlander, 1982). An observation of size-segregated PM and its chemical compositions is necessary for knowing the detailed information on the formation and sources of particles (Wu et al., 2015; Du et al., 2015; Yao et al., 2003; Ondov and Wexler, 1998). Reasonable measures for controlling air pollution can be applied according to the size distribution characteristics of the particle and chemical compositions, especially during severe haze episodes (Zhao et al., 2011; Hering et al., 1997; Han et al., 2015b; Deng et al., 2015).

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Beijing–Tianjin–Hebei, Yangtze River Delta, Pearl River Delta, and Sichuan Basin are the most polluted regions in China (Chen and Xie, 2012, 2014b; Wu et al., 2016; Brauer et al., 2012; Huang et al., 2012; Cao et al., 2009). Sources and diffusion conditions of PM in these regions may vary because of the varying energy consumption levels, industrial structures, car ownership levels, meteorological conditions, and topographic features (Fu et al., 2014; Deng et al., 2012; Li et al., 2015; Chen et al., 2014). A series of studies about haze pollution have been conducted in regions. Characterization of carbonaceous aerosols (Cao et al., 2007; Li and Bai, 2009), source apportionment of fine particles and volatile organic compounds (Wu et al., 2016; Liu et al., 2015), vertical characteristics of pollutant (Han et al., 2015a, 2015b) have been conducted in Tianjin. Studies about the chemical composition and source apportionment of $PM_{2.5}$ and PM_{10} (Liu et al., 2015; Cao et al., 2009) and the analysis of factors influencing aerosol light extinction (Wang et al., 2016; Xiao et al., 2011) were done in Hangzhou. Most studies made in Chengdu focused on the influence of dust storms and biomass burning to particles (Tao et al., 2013; Chen and Xie, 2014a and Chen et al., 2015; Yang et al., 2012). However, no study has reported the characterization of chemical compositions in different mode during severe haze pollution episodes in the three cities. In addition, most studies on air pollution have only been conducted in one city or region (Du et al., 2011; Tan et al., 2014; Hu et al., 2016). It is crucial to investigate and compare the characteristics of chemical compositions in size-segregated atmospheric particles during severe haze episodes in different polluted regions of China.

In this study, Tianjin, Hangzhou, and Chengdu, which are the central cities of Beijing–Tianjin–Hebei, Yangtze River Delta, and Sichuan Basin, respectively, were selected as the study areas. Size-segregated atmospheric particle samples were collected using eight-stage Anderson cascade impactors in these cities during winter, a season in which severe haze episodes are frequent in China. Ten major compositions of particles, namely carbonaceous species [elemental carbon (EC) and organic carbon (OC)], and water-soluble inorganic ions (Na^+, NH_4^+, K^+, Mg^{2+}, Ca^{2+}, Cl^-, NO_3^-, and SO_4^{2-}) were analyzed. The primary objective of this study was to 1) investigate the size distribution of chemical compositions in particles in three megacities during severe haze pollution episodes and 2) compare and analyze the differences in the size distributions of chemical compositions during these episodes among the cities.

2. Experimental techniques and methods

2.1. Sampling site descriptions

The locations of Tianjin (117°26′E, 39°00′N), Hangzhou (120°12′E, 30°13′N) and Chengdu (104°06′E, 31°06′N) are provided in Fig. 1. There are significant differences in the features of topographic, climate, energy consumption and industrial structure for these three cities.

Tianjin is a northern city of China, which has a sub-humid warm monsoon climate. Hangzhou and Chengdu are southern cities, which experience subtropical monsoon climate. Tianjin and Hangzhou are located on the coasts, while Chengdu lies on the west of the Sichuan Basin and midstream of Minjiang River. The average temperature of Tianjin (1.4 °C) in winter is lower than that of Hangzhou (8 °C) and Chengdu (7 °C). The relative humidity of Chengdu (80%) in winter is obviously higher than that of Hangzhou (65%) and Tianjin (56%).

These three cities are all the center of politics, economy, culture and business in the local regions. Tianjin is a municipality directly under the central government, while Hangzhou and Chengdu are the provincial capitals of Zhejiang and Sichuan respectively. The population of Hangzhou (9.0 million) is least with the biggest area (16,596 km^2), compared with Tianjin (15.5 million people in 11,946 km^2) and Chengdu (14.7 million people in 14,605 km^2). The GDP of Tianjin, Hangzhou and Chengdu is 1654, 1005, 1080 billion RMB respectively, while the possession of private vehicles is 2.35, 1.85 and 3.31 million in 2015. The industry structure of Tianjin, Hangzhou and Chengdu are 1.3: 46.7: 52.0, 2.9: 38.9: 58.2 and 3.5: 43.7: 52.8 (primary industry: secondary industry: tertiary industry) in 2015. A high proportion of secondary industry in Tianjin means that coal-related industrial combustion and incineration are the important contributors to air pollution (Lai et al., 2007; Zhao et al., 2011; Kaneyasu et al., 1999).

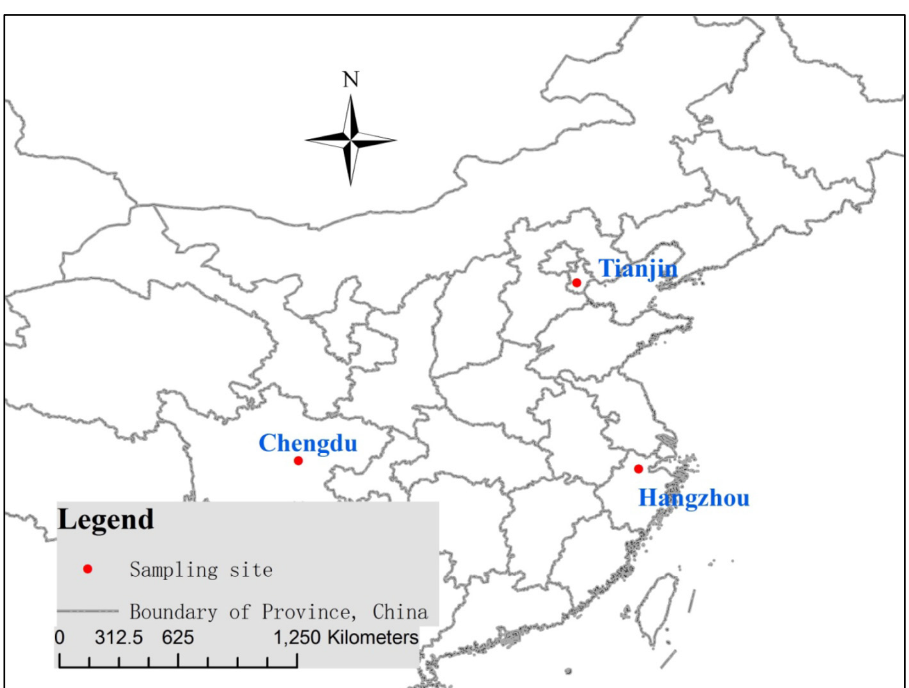

Fig. 1. Locations of three sampling sites.