



Characterization of PM_{2.5} and the major chemical components during a 1-year campaign in rural Guangzhou, Southern China



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ABSTRACT

A 1-year campaign was conducted in the rural area of Guangzhou, a megacity in southern China, to collect fine particulate matter (PM_{2.5}) from March 2012 to February 2013. The mass concentrations of PM_{2.5} and the major chemical components including 6 water-soluble ions, organic carbon (OC), elemental carbon (EC), and 13 additional elements were measured. The annual average concentration of PM_{2.5} was $44.2 \pm 25.8 \mu\text{g}/\text{m}^3$. Sulfate was the most dominant component, accounting for 28.6% of PM_{2.5}, followed by organic matter (21.9%). Both sea salt and crustal material accounted for only a small fraction of PM_{2.5} (<5%). Seasonal enhancement of PM_{2.5} was observed in autumn 2012, especially with high-PM_{2.5} events (more than $100 \mu\text{g}/\text{m}^3$) in October. The backward Lagrangian particle dispersion modeling (LPDM) and the cluster analysis of the back-trajectories indicate that the northern area is an important source region of long-range transport. An enhancement of PM_{2.5} as well as sulfate, OC, and EC was observed in the samples with the influence of northern air masses. However, the footprint retroplume of the samples shows that the sources in the Pearl River Delta Region should also be considered, especially secondary aerosol formation and biomass/biofuel burning. Two high-PM_{2.5} case studies show that both local and long-range transport can play important roles in the PM_{2.5} elevation episode.

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1. Introduction

Atmospheric aerosol is one of the variable components in the atmosphere. The physical and chemical properties as well as the environmental and health effects of atmospheric aerosols have been widely studied in past decades (Pöschl, 2005). It has been shown that aerosols can play an important role in regional and global climate by directly and indirectly impacting solar radiation (Ding et al., 2013b; Pöschl, 2005). Light extinction (both scattering and absorbing) can cause visibility degradation on a limited scale to a regional scale. The ability of light extinction depends not only on particle size but also on chemical composition and hygroscopic properties, as well as the mixing state of some light extinction-related species such as black carbon and ammonium sulfate (Koike et al., 2014; Robinson et al., 2013). Atmospheric aerosol can also affect human health by the penetration of fine and ultra-fine particles into the human respiratory system (Pope et al., 2002). Toxic chemical components and pathogenic organisms on particles can

cause further harm to the human body with acute and chronic effects (Lepeule et al., 2012).

In recent years, the frequent outbreaks of severe haze episodes over a vast area of China have drawn the attention of the public. The episodes normally had the feature of dramatic visibility degradation and PM_{2.5} elevation. The haze can disturb local and regional transport systems, intervene in urban activities, and cause an increase in morbidity and mortality due to chronic and cardiovascular diseases (Hua et al., 2014; Li and Zhang, 2014). As the key pollutant in haze episodes, PM_{2.5} was for the first time included as one of the targeted pollutants in the National Ambient Air Quality Standard of China in 2012 (NAAQS-China, GB3095-2012). Guangzhou, located in the Pearl River Delta region (PRDR), is the third largest city in China, with a population of more than 12 million. The fast regional economic development across the PRDR drove the fine particulate matter pollution from the cities to the whole region (Yuan et al., 2013). In addition to local emissions, sources outside of the PRDR contribute a significant fraction of the PM_{2.5}, which is also related to the seasonal change in the prevailing wind direction in the region (Hagler et al., 2006).

Here, we present the results of PM_{2.5} and the major chemical components from a 1-year campaign (March 2012–February 2013) at a rural site

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in Guangzhou. We intend (1) to investigate the characteristics of PM_{2.5} and the major chemical components at this site; (2) to understand the contribution of PM_{2.5} sources; and (3) to reveal the contributing sources in the PRDR as well as the long-range transport from other regions.

2. Methods

2.1. Sampling site and sample collection

The sampling campaign was conducted at Tianhu (TH), Guangzhou (23.65°N, 113.63°E), from March 2012 to February 2013. The sampling site is located in a recreational area that is at a distance of ~60 km north-east of the city center of Guangzhou (Fig. S1). There is no direct stationary source near the sampling site, and the major roads are more than 2 km away from the site.

During the campaign, a median volume sampler (300 L/min, Mingye, China) and a mini volume sampler (5 L/min, Airmetrics, US) were placed on the roof of a building to collect PM_{2.5} samples simultaneously at an interval of 6 days with a duration of 24 h. A quartz filter (25 × 20 cm, Pall, US) was used for the median volume sampler, and a PTFE filter (φ = 47 mm, Pall, US) was loaded into the mini volume sampler. A total of 51 quartz filters and 52 PTFE samples were collected during the campaign. Field blank samples were also collected to assure the consistence of the sampling procedure. The quartz filter was pre-baked in an oven at 550 °C to remove carbon content. After sampling, all filters were stored in the freezer at -20 °C to avoid contamination before analysis.

2.2. Chemical analysis

2.2.1. Mass

PTFE filters were analyzed gravimetrically for the mass concentration of PM_{2.5} using an electronic microbalance with a sensitivity of ±0.001 mg (Sartorius MC5, Germany). Before weighing, the sample filters were equilibrated at a temperature of 20–30 °C and at a relative humidity of 30–40% for 24 h. Each filter was weighed twice before and after sampling. The net mass was obtained by subtracting the average of the pre-sampling weights from the average of the post-sampling weights. The difference among the repeated weightings was less than 15 µg.

2.2.2. Water-soluble ions

A part of 2 × 2 cm² of each quartz filter sample was cut to analyze water-soluble ions. Six major ionic species (Na⁺, NH₄⁺, K⁺, Cl⁻, NO₃⁻, and SO₄²⁻) were measured by ion chromatography (DX90, Dionex, US). A CS12 column (4 × 250 mm) with 13.8 mM H₂SO₄ eluent and an AS14 column (4 × 250 mm) with 3.5 mM Na₂CO₃/1.0 mM NaHCO₃ eluent were used for the determination of cations and anions, respectively. The cut portion was extracted twice in 5 mL ultra-pure water (18 mΩ) and sonicated in an ice bath for 30 min. The total extract solution (10 mL) was filtered through a syringe filter (pore size: 0.45 µm) and stored in the refrigerator before analysis. After every 10 samples, a random replicate check was performed. The average value of the relative standard deviations (RSD) of replicate analyses were 1.2% for Cl⁻, 2.8% for NO₃⁻, 2.2% for SO₄²⁻, 5.0% for Na⁺, 2.0% for NH₄⁺, and 5.6% for K⁺.

2.2.3. Carbonaceous fractions

A 1.0 cm² punch of each quartz filter was taken to determine the content of organic carbon (OC) and elemental carbon (EC) using the thermo-optical transmittance (TOT) method (NIOSH protocol) by an aerosol OC/EC analyzer (Sunset Laboratory Inc., US) (Birch and Cary, 1996).

2.2.4. Elements

The PTFE filters were used to detect the elements by x-ray fluorescence spectrometry (XRF, Epsilon5, PANalytical, the Netherlands).

Thirteen elements, i.e., Al, Si, S, Cl, K, Ca, Ti, Mn, Fe, Cu, Zn, Br, and Pb, were measured. The limits of detection (LODs) are shown in Table S1.

2.3. Transport and dispersion simulation

Seven-day backward trajectories at an altitude of 100 m above ground level at 0:00 UTC (8:00 local time) on every sampling day were calculated by the Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPPLIT) model. Furthermore, backward Lagrangian particle dispersion modeling (LPDM) was also conducted using the HYSPPLIT model to investigate the potential source region. The model calculates the position of particles using mean wind and a turbulence transport component after they are released at a source point for forward simulations or a receptor for backward runs. The configuration and parameters of LPDM in this study are summarized in Table S2. More details of the LPDM can be found in a previous study by Ding et al. (2013a), in which the model was successfully used to simulate temporal variation of carbon monoxide measured at a station in the PRDR. Ding et al. (2013b) used the same method to identify potential source region of PM_{2.5} based on measurement at a site in East China.

3. Results and discussion

3.1. PM_{2.5} and the chemical composition

The concentrations of PM_{2.5} and the chemical composition at TH from March 2012 to February 2013 are summarized in Table 1. The annual average concentration of PM_{2.5} was 44.2 ± 25.8 µg/m³, which was higher than the annual limit of PM_{2.5} (35 µg/m³) in the recently released NAAQS-China. The annual average of PM_{2.5} at TH was lower than those reported in the urban area in Guangzhou but close to those reported in the rural area during 2002–2003 (Cao et al., 2003, 2004; Ding et al., 2011; Lai et al., 2007; Tao et al., 2014; Yang et al., 2011). The results from previous studies in Guangzhou are summarized in Table 2. Generally, the level of PM_{2.5} at TH was lower than those reported in the urban area. Out of the 52 collected PM_{2.5} samples, only 4 were observed to exceed the daily limit of 75 µg/m³. The variation and the enhancement of PM_{2.5} can be influenced by both local sources and long-range transport (Deng et al., 2008; Hagler et al., 2006; Li et al., 2013). A detailed discussion on the source region is presented in Section 3.5.

In this study, the chemical composition of PM_{2.5} includes organic matter (OM, defined as OC multiplied by a factor of 1.6 (Cao et al., 2003)), EC, water-soluble ions, crustal material (CM), non-crustal material (non-CM) (for definitions of crustal and non-crustal material, see Section 3.2), and an unidentified fraction (Fig. S2). Sulfate is the predominant species in PM_{2.5}, accounting for 28.6%. This is consistent with the results reported by previous studies (Lai et al., 2007; Tao

Table 1

Average concentrations of PM_{2.5} and the associated chemical components at Tianhu from March 2012 to February 2013 (µg/m³).

Component	Spring	Summer	Autumn	Winter	Average
PM _{2.5}	31.3 ± 12.4	36.8 ± 18.6	67.1 ± 33.2	40.8 ± 20.2	44.2 ± 25.8
OM ^a	7.2 ± 4.9	7.8 ± 5.8	12.2 ± 6.1	11.9 ± 7.6	9.8 ± 6.5
EC	0.7 ± 0.3	0.6 ± 0.3	0.9 ± 0.4	1.0 ± 0.4	0.8 ± 0.4
CM ^b	2.1 ± 1.9	1.3 ± 0.9	2.8 ± 1.5	2.0 ± 1.3	2.1 ± 1.5
Non-CM ^c	0.4 ± 0.3	0.3 ± 0.3	0.8 ± 0.3	0.5 ± 0.4	0.5 ± 0.4
Cl ⁻	0.2 ± 0.1	0.2 ± 0.1	0.2 ± 0.1	0.3 ± 0.2	0.2 ± 0.2
NO ₃ ⁻	1.8 ± 1.7	1.6 ± 2.0	1.4 ± 2.0	2.9 ± 3.4	2.0 ± 2.4
SO ₄ ²⁻	9.1 ± 2.8	9.5 ± 5.2	17.8 ± 11.5	12.6 ± 7.6	12.2 ± 8.0
Na ⁺	0.5 ± 0.3	0.5 ± 0.2	0.6 ± 0.2	0.5 ± 0.2	0.5 ± 0.2
NH ₄ ⁺	3.4 ± 0.9	3.7 ± 2.1	7.6 ± 3.4	5.8 ± 2.6	5.2 ± 2.9
K ⁺	0.6 ± 0.2	0.7 ± 0.3	0.8 ± 0.3	0.7 ± 0.3	0.7 ± 0.3

^a OM (organic matter) = OC × 1.6.

^b CM (crustal material) = 2.2 [Al] + 2.49 [Si] + 1.63 [Ca] + 2.42 [Fe] + 1.94 [Ti].

^c Non-CM (non-crustal material), the sum of the elements of S, Cl, K, Mn, Cu, Zn, Br, and Pb.

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