



Chemical composition and source apportionment of the ambient PM_{2.5} in Hangzhou, China



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ABSTRACT

To identify and apportion the sources of the ambient PM_{2.5} in the urban area of Hangzhou, China, PM_{2.5} samples were collected at three sites in the city from April 2004 to March 2005. Water-soluble ions, metal elements, and total carbon (TC) in PM_{2.5} samples were analyzed. The results indicated that the 24-h mean concentrations of PM_{2.5} ranged from 17.1 to 267.0 μg/m³, with an annual average value of 108.2 μg/m³. Moreover, the seasonal mean values for PM_{2.5} in spring, summer, autumn, and winter were 116, 73.1, 114.2, and 136.0 μg/m³, respectively. According to the Chinese ambient quality standard, at least 70% of the monitoring data exceeded the limit value. The total contribution of water-soluble ions, including F⁻, Cl⁻, NO₃⁻, SO₄²⁻, NH₄⁺, K⁺, and Na⁺, to PM_{2.5} mass varied from 32.3% to 36.7%. SO₄²⁻, NO₃⁻, and NH₄⁺ were the main constituents of the ions, with contributions to PM_{2.5} varying from 14.1% to 14.7%, 6.0% to 7.8%, and 6.4% to 7.7%, respectively. In addition, the annual mean mass fraction of TC in PM_{2.5} was 27.8%. The annual average total contribution of the group of elements of Zn, Pb, Cu, Mn, Cr, Ni, Se, Mo, Cd, Sb, and Ag to the aerosol was in the range of 1.7–2.0%. Furthermore, positive matrix factorization was applied to analyze the PM_{2.5} data collected from the central area, and five factors were identified. The factor contributions to PM_{2.5} mass were 12.8%, 31.9%, 10.1%, 17.2%, and 27.9%, respectively. Iron/steel manufacturing and secondary aerosol were the main sources for the fine particles. These findings may have significance for controlling the atmospheric contamination in the city.

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1. Introduction

Ambient PM_{2.5} pollution has various effects on human health (Jacobs et al., 2012; McDonnell, Nishino-Ishikawa, Petersen, Chen, & Abbey, 2000; Rohr & Wyzga, 2012). PM_{2.5} is composed of metal elements, water-soluble ions, elemental carbon (EC), and organic compounds. Metal elements in PM_{2.5} can be divided into crustal and anthropogenic elements. The former, including Fe, Al, Ca, and Mg, contribute 20% to the PM_{2.5} mass, while the latter, including Zn, Pb, Cu, Mn, Cr, Ni, Se, Mo, Cd, Sb, Ag, and Hg, contribute 1–2% to the aerosol content (Lonati, Giugliano, Butelli, Romele, & Tardivo, 2005; Vecchi, Marazzan, Valli, Ceriani, & Antoniazzi, 2004; Viana et al., 2005; Wang et al., 2004). F⁻, Cl⁻, NO₂⁻, NO₃⁻, SO₄²⁻, NH₄⁺, K⁺, Na⁺, Ca²⁺, and Mg²⁺ are common constituents of the water-soluble ions in PM_{2.5}, of which NO₃⁻, SO₄²⁻, Cl⁻, and NH₄⁺ are the major components. The contribution of these ions to the PM_{2.5} mass varies

from 30% to 50% (Giugliano et al., 2005; Kim et al., 2006; Lonati et al., 2005; Viana et al., 2005; Wang et al., 2004; Wang, Bi, Sheng, & Fu, 2006). The contents of the total carbon (TC) in PM_{2.5} in Beijing ranges from 20% to 40%, more than 70% of which is organic carbon (OC). In addition, the concentrations of both OC and EC are all greater in winter than in summer (Dan, Zhuang, Li, Tao, & Zhuang, 2004). In Milan (Italy), the average contents of EC in PM_{2.5} collected in summer and winter vary from 4.0% to 8.8% and 3.1% to 4.8%, respectively. In addition, the mean contents of OC collected in summer and winter vary from 19.3% to 37.3% and 26.6% to 46.8%, respectively (Giugliano et al., 2005; Lonati et al., 2005; Vecchi et al., 2004). Furthermore, the TC contents in PM_{2.5} in European urban backgrounds are in the range of 20–35% (Querol et al., 2004).

Chemical composition discrimination for PM_{2.5} with different dynamical diameters exists. In Meptitz (Germany), nitrate is distributed in both coarse and fine PM_{2.5}, whereas most of the sulfate is distributed in fine PM_{2.5} (Maenhaut, Cafmeyer, Dubtsov, & Chi, 2002). The content peaks for sulfates and nitrates are found in particles with a diameter of 1.1 μm in the rural and urban regions of Leeds (Clarke, Azsdi-Boogar, & Andrews, 1999). Moreover, more

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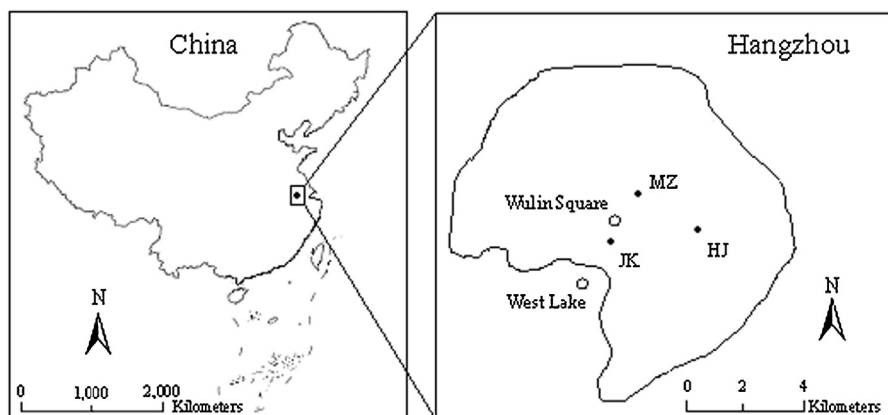


Fig. 1. Location of the sampling sites.

than fifty percent of chloride is in the coarse aerosol (Clarke et al., 1999). There are some differences in the temporal distribution for water-soluble salts in $PM_{2.5}$. For example, most of the sulfate exists in fine particles all of the time, whereas the distribution modes of chloride are distinct in $PM_{2.5}$ collected at day and night (Fang et al., 1999). In addition, crustal elements generally concentrate in coarse $PM_{2.5}$, whereas anthropogenic elements tend to be enriched in fine $PM_{2.5}$ (Balachandran, Meena, & Khillare, 2000).

Atmospheric $PM_{2.5}$ could be emitted from different sources. Iron and steel manufacturing, diesel-powered motor vehicles, coal combustion, road re-suspension dust, solid waste incineration, and open biomass burning are all emission sources of the aerosol. Furthermore, gaseous contaminants, such as SO_2 , NO_x , and NH_3 , are likely to transform into $PM_{2.5}$ through photochemical reactions. Chemical mass balance, positive matrix factorization (PMF), principal component analysis, and multivariable linear regression are often used to identify and apportion sources for $PM_{2.5}$ (Begum, Hopke, & Zhao, 2005; Hwang & Hopke, 2007; Marmur, Mulholland, & Russell, 2007; Pancras, Landis, Norris, Vedantham, & Dvonch, 2013; Viana et al., 2007; Zheng et al., 2005).

Since the policy of economic reform was put into practice in China since the late seventies of the last century, the Chinese economy has developed quickly. However, the air quality in the country has simultaneously declined, particularly in urban areas. Hangzhou is one of the most developed cities in China, but the ambient pollution level in the city is relatively high as well. PM_{10} is the primary pollutant in the city. However, less research work has been performed on $PM_{2.5}$ contamination. Zhu, Chen, Wang, and Shen (2004) studied the composition of PAHs in particulate matter. Okuda, Kumata, Naraoka, and Takada (2002) analyzed the stable carbon isotope ratio values of PAHs. Furthermore, the relationship between meteorological factors and the concentration of ambient $PM_{2.5}$ has also been studied (Hong, Jiao, & Ma, 2013). The aims of this paper are as follows: (1) to investigate the temporal and spatial distributions of ambient $PM_{2.5}$ in urban area of Hangzhou; (2) to determine the chemical compositions of water-soluble ions, metal elements and TC in $PM_{2.5}$; and (3) to identify and apportion the emission sources of ambient $PM_{2.5}$.

2. Sampling and analytical methods

2.1. Sample collection

Ambient $PM_{2.5}$ samples were collected at three sites in the urban area of Hangzhou from April 2004 to March 2005. One sampling site, HJ was located in the division campus of Zhejiang University,

Huajiachi (Fig. 1). Another, MZ was located in the second habitation area of Zhaohui. The third, JK lay south of Wulin Square, the urban center. Air samplers ($PM_{2.5}$ -2, Beijing Geological Instrument Factory, China) were placed on the roofs of buildings, which were ten to twenty meters above the ground and thirty meters away from nearest roads. Two samples were collected simultaneously at each site per week at the airflow rate of 77 L/min. The collection duration per sample was 24 h (from 9:00 a.m. to next 9:00 a.m.). Before sampling, glass fiber filters were baked for 2 h at 500 °C and then placed in a desiccator to reach humidity equilibrium for 24 h at room temperature. To assess sampling error, all air samplers were placed together at a site and used to collect experimental aerosol samples at one time. The relative errors for 24 h sampling between the three samplers varied from -2.6% to +2.3%.

2.2. Analytical methods

2.2.1. Mass and TC

The $PM_{2.5}$ mass was determined by gravimetry. Before and after sampling, the glass filters were weighed on an electronic balance with a reading precision of 0.01 mg. The samples were allowed to reach humidity equilibrium in the weighing room for 24 h before weighing. The aerosol samples were wrapped in aluminum foil and stored in a freezer under -18 °C until performing the chemical analysis after weighing.

The TC in $PM_{2.5}$ samples was analyzed using an elemental analyzer (Flash EA-1112, ThermoFinnigan Italia S.P.A.). A filter sample weighing 2 mg was burned under an oxygen environment in a quartz tube at 800 °C. The carbon dioxide produced was then carried by helium to a gas chromatograph and analyzed. Five blank filters were measured in the same process, and the average background was subtracted from all of the aerosol samples.

2.2.2. Water-soluble ions

To measure the water-soluble ions in $PM_{2.5}$, a quarter of a filter sample was cut into pieces and then placed into a polypropylene tube with screw cap. Afterwards, 13 mL of deionized water (electrical conductivity was equal to 18.2 MΩ cm) was added to the tube. The sample was extracted for 30 min under ultrasonic conditions. Finally, the liquor was centrifuged at 3000 rpm for 5 min and filtrated using a hydrophilic membrane with a pore diameter of 0.45 μm. Five blank filters were treated in the same way to subtract the background from the aerosol samples. An ion chromatograph (Metrohm-732, Metrohm, Switzerland) was used to analyze the water-soluble ions in the aerosol. Anions were eluted using an aqueous solution with concentration of (336 mg $NaHCO_3$ + 106 mg

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