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RESEARCH ARTICLE

Characteristics and source apportionment of $PM_{2.5}$ in Jiaxing, China

Zhipeng Zhao[1] · Sheng Lv[2] · Yihua Zhang[3] · Qianbiao Zhao[3] · Lin Shen[1] · Shi Xu[1] · Jianqiang Yu[1] · Jingwen Hou[1] · Chengyu Jin[1]

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Abstract

Herein we investigated the morphology, chemical characteristics, and source apportionment of fine particulate matter ($PM_{2.5}$) samples collected from five sites in Jiaxing. Scanning electron microscopy (SEM) and transmission electron microscopy (TEM) showed that soot aggregates and coal-fired fly ash were generally the most abundant components in the samples. All the samples were analyzed gravimetrically for mass concentrations and their various compositions were determined. Our results revealed that the $PM_{2.5}$ concentrations in the samples were in the following order: winter > spring > autumn > summer. The $PM_{2.5}$ concentrations in winter and spring were higher than those in autumn and summer, except for inorganic elements. Carbonaceous species and water-soluble inorganic ions were the most abundant components in the samples, accounting for 26.17–50.44% and 34.27–49.6%, respectively. The high secondary organic carbon/organic carbon ratio indicated that secondary organic pollution in Jiaxing was severe. The average ratios of NO_3^-/SO_4^{2-}, ranging from 1.01 to 1.25 at the five sites, indicated that mobile pollution sources contributed more to the formation of $PM_{2.5}$ than stationary sources. The BeP/(BeP + BaP) ratio (0.52–0.71) in samples reflected the influence of transportation from outside of Jiaxing. The positive matrix factorization (PMF) model identified eight main pollution sources: secondary nitrates (26.95%), secondary sulfates (15.49%), secondary organic aerosol (SOA) (19.64%), vehicle exhaust (15.67%), coal combustion (8.6%), fugitive dust (7.7%), ships and heavy oil (5.23%), biomass burning, and other sources (0.91%). Therefore, $PM_{2.5}$ pollution in Jiaxing during the winter and spring seasons was more severe than that in the summer and autumn. Secondary aerosols were the most important source of $PM_{2.5}$ pollution; therefore, focus should be placed on controlling gaseous precursors.

Keywords $PM_{2.5}$ · Morphology · Chemical characteristics · Source apportionment

Introduction

Atmospheric particulate matter is a general term for various solid and liquid particulate matter present in the atmosphere. Fine particulate matter ($PM_{2.5}$) (aerodynamic diameter ≤ 2.5 μm) has received widespread attention because of its harmful effects (Huang et al. 2015; Jung et al. 2009). $PM_{2.5}$ containing complex chemical composition (Hu et al. 2014) originates from both natural sources and anthropogenic emissions (Kelly and Fussell 2012). In urban areas, $PM_{2.5}$ is mainly produced from anthropogenic sources including both primary and secondary particles. The latter group is formed during the chemical conversion of gaseous precursor pollutants (Zhang et al. 2015a, b, c).

Since the 1980s, the USA and some European countries have conducted large-scale $PM_{2.5}$ research, with the source apportionment of $PM_{2.5}$ and its impact on human health emerging as important research focuses. Receptor models based on pollution sources and contaminated areas, such as

Highlights
- The concentrations of $PM_{2.5}$ in five sites were in accordance with the rule that winter > spring > autumn > summer.
- Secondary aerosols were the most important source of $PM_{2.5}$ pollution in Jiaxing.
- Mobile sources contributed more to the formation of $PM_{2.5}$ than stationary sources in Jiaxing.

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✉ Chengyu Jin
cyjin@sjtu.edu.cn

[1] Instrumental Analysis Center, Shanghai Jiao Tong University, Shanghai 200240, China

[2] Jiaxing Environmental Monitoring Station, Jiaxing 314000, China

[3] Shanghai Environmental Monitoring Center, Shanghai 200235, China

the chemical balance model (CMB) and positive matrix factorization model (PMF), have been the most widely used $PM_{2.5}$ source apportionment technique (Gibson et al. 2015). It is worth mentioning that, the PMF model is based on receptor chemical component data sets for source apportionment without source sample acquisition and is highly recommended when the sources were uncertain (Sharma et al. 2016). The source apportionment results in many parts of the world revealed that the main sources of $PM_{2.5}$ are secondary inorganic aerosols and organic aerosols, coal combustion, industrial emissions, traffic emissions, biomass combustion, and soil dust (Almeida et al. 2005; Amato and Hopke 2012; Heo et al. 2009).

In the past few decades, with the acceleration of urbanization, the pollution due to $PM_{2.5}$ in China has become severe, especially in the Pearl River Delta (PRD) (Ho et al. 2014), the Yangtze River Delta (YRD) (Li et al. 2011), and Beijing-Tianjin-Hebei regions (Zhao et al. 2013). For example, in the first quarter of 2013, China experienced extremely severe and persistent haze pollution, which affected a roughly 1.3-million-km^2 region and 800 million people (Huang et al. 2015). In recent years, the air quality in China has improved. $PM_{2.5}$ pollution has been reduced to a certain extent owing to the increased attention it has received. Chinese scholars have conducted a considerable amount of $PM_{2.5}$ research and obtained many results on $PM_{2.5}$ pollution characteristics (Sun et al. 2004), chemical composition (Cheng et al. 2015), and source apportionment (Wang et al. 2016; Jiang et al. 2018), the relationship between $PM_{2.5}$ and other particulate matter (Wang et al. 2008), and the health risks of $PM_{2.5}$ pollution (Hu et al. 2012). Studies in many regions of China showed that $PM_{2.5}$ concentrations had obvious seasonal characteristics, with the highest concentration occurring in winter and the lowest in summer (Zhang et al. 2014; Zheng et al. 2005; Louie et al. 2005; Xu et al. 2012; Geng et al. 2006). The source apportionment results showed that the types of pollution sources were similar to those in Europe and the USA, but their contribution rates were different (Wang et al. 2013; Liu et al. 2015; Tao et al. 2014; Zhang et al. 2015a, b, c). However, in the past, few sampling sites and short sampling times have been utilized in research on the $PM_{2.5}$ concentrations of various scale cities; furthermore, the analysis of samples has not been comprehensive enough.

Jiaxing, a medium-sized city with a population of over 3.5 million in the YRD, is located in the northeast of Zhejiang Province. It is less than 100 km away from cities such as Shanghai, Hangzhou, and Ningbo. The aerosol concentration in Jiaxing is affected by external sources of aerosols from cities around it. Meanwhile, the local pollution sources are complex. Therefore, it is necessary to determine the source apportionment of $PM_{2.5}$ pollution to develop effective control policies. At present, there are few studies on the source apportionment of $PM_{2.5}$ in Jiaxing. We, therefore, studied the differences between Jiaxing and surrounding cities, and focused on the differences between five sites in Jiaxing. In this work, we conducted multi-site, long-term sampling in Jiaxing. The morphology and comprehensive pollution characteristics of $PM_{2.5}$ in Jiaxing were investigated at five sites from October 2016 to November 2017. The PMF model was applied for the source apportionment using chemical composition (organic carbon, elemental carbon, major secondary ions, inorganic elements) of $PM_{2.5}$ mass.

Materials and method

Site description and $PM_{2.5}$ sampling

In this study, the $PM_{2.5}$ samples were collected at Jiaxing. We carried out sampling at five sites: Canlian (CL), Heyuan (HY), Qinghe (QH), Gangqu (GQ), and Tongxiang (TX). Among them, the CL, HY, and QH are located in the Jiaxing city area. CL is located in the new urban area of Jiaxing, and is mainly surrounded by residential areas. QH is located in the business district of the old city and has a dense population and well-developed catering industry. Although HY is located downtown, its surroundings are very special: it is less than 500 m away from the highway in the east, less than 50 m away from the main traffic artery in the west, 1 km away from a coach station, and it is also close to a logistics center and a heavily used military airport. GQ is located in the Gangqu District and is close to the Zhapu Port. There are five polluting enterprises within 1.5 km. TX is located in the outskirts of Jiaxing. The arable land area of Tongxiang accounts for 52.8% of the total land area and is far away from sources of pollution. The location of each representative site is shown in Fig. 1.

During the period from October 2016 to November 2017, we used pairs of Ø90-mm quartz fiber filters to collect $PM_{2.5}$ samples at five sites simultaneously with mid-volume samplers (Wuhan Tianhong TH Ш, China) at a rate of 100 L/min for a duration of 48 h. The heights of the sites for the CL, QH, HY, GQ, and TX were 20 m, 20 m, 5 m, 20 m, and 10 m, respectively. A total of 440 $PM_{2.5}$ valid samples were collected. The filters were placed in a humidity chamber (temperature 25 ± 1 °C and relative humidity 50 ± 3%) for 48 h before and after sampling, and weighed with an electronic balance (0.01 mg precision, Sartorius BT 25S, Germany). Then, all sample filters were stored in a − 20 °C freezer until further chemical physical and chemical characterization.

Morphological observation of $PM_{2.5}$

The morphology of $PM_{2.5}$ was studied by scanning electron microscopy (SEM) and transmission electron microscopy (TEM). For SEM observation, $PM_{2.5}$ filter of area 1 cm^2 was pasted onto the surface of a double conductive adhesive

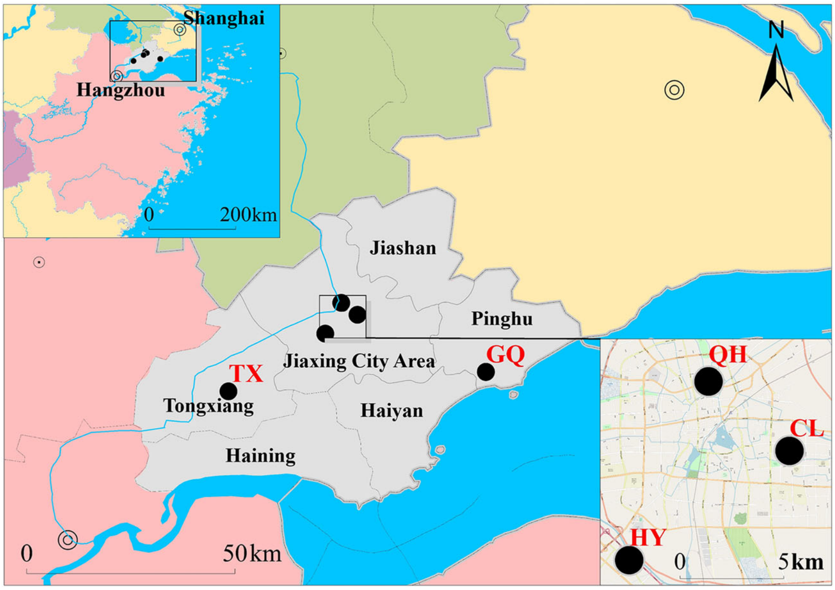

Fig. 1 Locations of the five sampling sites in Jiaxing, Zhejiang Province, China. Canlian site (CL), Heyuan site (HY), and Qinghe site (QH) are located in the Jiaxing city area. The Gangqu site (GQ) is located in Gangqu District. The Tongxiang site (TX) is located in Tongxiang District

fixed on an aluminum stub. Then, the sample was coated with gold and put into the sample compartment of the SEM (FEI, Nano 230, USA) for further analysis. For TEM analysis, a portion of the resuspended particles was placed onto carbon-coated copper grids and air-dried. The grids were analyzed with the TEM (FEI, Tecnai G2 spirit Biotwin, USA).

Inorganic elements and water soluble inorganic ions

To test for the presence of the inorganic elements and inorganic water-soluble ions in $PM_{2.5}$, one eight of the sample filters were cut and ultrasonically extracted for 45 min with a 25-mL nitric acid solution (5 mL HNO_3 + 20 mL Milli-Q purified water) and 25 mL Milli-Q purified water (specific resistivity = 18.2 MΩ cm^{-1}; Millipore, Massachusetts, USA), respectively. Then, the mixture was filtered with a 0.45-μm aqueous filter. The presence of inorganic elements (Al, As, Ca, Cr, Fe, K, Mg, Mn, Na, Ni, Si, V, Zn) was determined using an inductively coupled plasma (ICP, Agilent 7600, USA). The anions (NO_3^-, SO_4^{2-}) were detected by ion chromatography (ICS, ThermoFisher 5000+, USA), and the cations were detected by ion chromatography (ICS, ThermoFisher 900, USA). The analysis conditions of the instruments are listed in the Supplementary Materials (Table 1).

Carbon component

Carbonaceous aerosols in atmospheric particulate matter are mainly divided into two types: organic carbon (OC) and elemental carbon (EC). OC and EC concentrations on a 0.508 cm^2 quartz filter were measured by using a thermal/optical carbon analyzer (DRI, Model 2015, USA). The sample was gradually heated in a pure helium atmosphere at 140 °C (OC1), 280 °C (OC2), 480 °C (OC3), and 580 °C (OC4), and then in a 2% O_2/98% He atmosphere at 580 °C (EC1), 740 °C (EC2), and 840 °C (EC3) to release the carbon compounds. The carbon compounds were converted to CO_2 by passing the volatilized compounds through an oxidizer (heated manganese dioxide, MnO_2), and then the CO_2 was quantified by a nondispersive infrared (NDIR) CO_2 detector. The OC fraction of the sample might be underestimated, and the EC fraction might include some pyrolyzed OC (OCpyro) because of oxygen-free heating. According to the IMPROVE protocol (Chow et al. 2007), OC = OC1 + OC2 + OC3 + OC4 + OCpyro, EC = EC1 + EC2 + EC3 − OCpyro.

Polycyclic aromatic hydrocarbons

One-half of the sample filters were cut and smashed, and extracted with a mixture containing 20 mL of a dichloromethane and n-hexane mixture (CH_3Cl_2/C_6H_{14}, 1:1 *v*/*v*) for 60 min. The extract was then filtered and concentrated to 1 mL under a nitrogen atmosphere. Quantification of polycyclic aromatic hydrocarbons (PAHs) in the collected atmospheric aerosols was carried out by GC-MS (Agilent, 7890A-5975C) with a DB-5 ms capillary column (HP5-MS, 250 μm i.d., 30-m length, 0.25-μm film thickness) under the electron ionization (EI) mode (70 eV). One microliter of the sample was injected into a splitless injector (temperature 300 °C). Helium (purity = 99.999%) was used as the carrier gas at a constant flow rate of 1.0 mL min^{-1}. The temperatures of the transfer line, ion source, and quadrupole were set to 290, 230, and 150 °C, respectively. The temperature program was as follows: heat the sample at 80 °C for 2 min, increase the temperature to 140 °C at a rate of 20 °C/min, increase the temperature to 305 °C at a rate of 3 °C/min, and then hold the temperature for 5 min. The mass spectrometry data was acquired in the full-scan mode (*m*/*z* 33–500). Analyses of the average mass spectra calculated at the chromatographic peak middle height were carried out with NIST 2014. The integrations were performed with ChemStation software (Agilent MSD).

PMF

PMF is an effective data analysis method proposed by Paatero and Tapper in 1993 (Paatero and Tapper 1993). The procedure is as follows: First, use the weights to calculate the error of various chemical components in the samples, and then use the least square method to resolve the main sources of contribution and contribution of atmospheric particles. The PMF model is a novel and effective method for the analysis of atmospheric particulate matter sources. Compared with other methods, the PMF model has the following advantages: there is no need to input local source spectrum information, one can use the standard deviation of the data to optimize, the element sharing rate in the decomposition matrix is non-negative, and the model can handle inaccurate data and missing data.

Assuming that X is an $n \times m$ matrix, whereby n is the number of samples, and m is the number of chemical components, then matrix X can be decomposed into matrix G and matrix F, where G is the source contribution matrix of the particle emission source of $n \times p$, F is the spectrum matrix of the receptor points of $p \times m$, and p is the number of major pollution sources. The definition is as follows:

$$X = GF + E$$

where E is the residual matrix, which represents the difference between X and GF.

The purpose of PMF analysis is to minimize Q. Q is defined as

$$Q = \sum_{i=1}^{n} \sum_{j=1}^{m} \left(e_{ij}/S_{ij}\right)^2$$

$$e_{ij} = \chi_{ij} = \sum_{k=1}^{p} g_{ik} f_{kj}$$

$$i = 1, 2, \ldots, n; j = 1, 2, \ldots, m; k = 1, 2, \ldots, p_{\circ}$$

where s_{ij} is the uncertainty of the j_{th} species concentration in sample i; and x_{ij}, g_{ik}, f_{kj}, and e_{ij} are the elements of the X, G, F, and E matrices, respectively.

Under the constraints of $g_{ik} \geq 0$ and $f_{kj} \geq 0$, an iterative minimization algorithm is used to solve Q. Then, the pollution source contribution value G (relative value) and pollution source spectrum F (chemical composition relative concentration value) can be determined simultaneously.

Results and discussion

Morphology of $PM_{2.5}$

SEM and TEM results (Fig. 2) revealed that regular and irregular mineral particles, smooth and similar spherical particles, soot aggregates, etc., were common components of $PM_{2.5}$. Mineral particles were mainly from resuspended soil and road and construction dust. Smooth spherical particles, which might originate mainly from coal burning, are called “coal-fired fly ash” (Shi et al. 2003). The surface of similar spherical particles is rough and might be a mixture of salt and organic matter (Buseck and Posfai 1999). Coal, motor vehicle exhaust, and biomass combustion are the main sources of soot aggregation (Yue et al. 2006). In Fig. 2c, the chain and cluster-like soot aggregates were gathered by a large number of round particles with a diameter of 30–50 nm. Dye et al. (2000) found that newly generated particles tend to have a chain-like distribution. With the aging, the soot aggregate particles tend to aggregate and had a clustered distribution or a more tightly aggregated distribution. We observed particle types from a variety of sources, indicating that the sources of $PM_{2.5}$ in Jiaxing were diverse and their formation mechanism was complex.

Mass concentration of $PM_{2.5}$

According to the experimental results, the maximum daily average $PM_{2.5}$ concentration during the sampling period was 248.69 $\mu g/m^3$ (December 5, 2016, GQ), and the lowest value was 16.17 $\mu g/m^3$ (June 17, 2017, QH); the maximum monthly average value and lowest value of $PM_{2.5}$ concentration were 132.41 $\mu g/m^3$ (December 2016, GQ) and 26.74 $\mu g/m^3$ (July 2017, CL), respectively. Furthermore, the monthly average mass concentrations of $PM_{2.5}$ were all higher than the WHO average concentration standard (25 $\mu g/m^3$).

The variation trend of the average $PM_{2.5}$ concentration for each site was consistent with a “V” distribution (Fig. 3). The concentrations of $PM_{2.5}$ at the five sites were in the following order: winter > spring > autumn > summer. This seasonal feature was similar to that of the surrounding cities, such as Hangzhou (Bao et al. 2010) and Ningbo (Xiao et al. 2012). In the summer, Jiaxing is affected by the plum rain season (special type of rainfall that mainly occurs in southern China) and the typhoon period, both of which bring about high-frequency and high-volume precipitation. This increases the wet deposition of atmospheric particles during this season, providing effective air self-purification; good atmospheric diffusion conditions also reduce the concentration of pollutants (Wang et al. 2013). However, in winter, the weather conditions are relatively stable, with more inversion temperatures and poor atmospheric diffusion, resulting in relatively high concentrations of particulate matter (Tai et al. 2010). Among the five sites, the

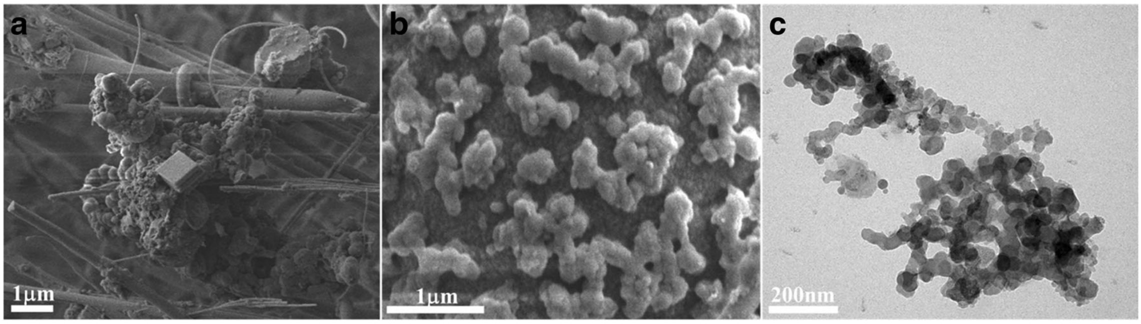

Fig. 2 Morphological characteristics of $PM_{2.5}$. **a** SEM images (scale bar 1 μm), all the species. **b** SEM images (scale bar 1 μm), typical soot aggregates; **c** TEM images (scale bar 200 nm), chain and agglomerated soot aggregates

$PM_{2.5}$ levels of GQ and HY were significantly higher than those of other sites. This might be because GQ is located in an industrial zone and is close to a port, while HY is greatly affected by traffic sources. As TX is located in the outer suburbs and away from industrial and traffic sources, its $PM_{2.5}$ concentration was low. The winter/autumn ratios for $PM_{2.5}$ concentrations at CL and QH were quite different from those at other sites. In fact, as CL and QH are located in the main urban area, they were not affected by special sources like HY. Therefore, the concentrations of $PM_{2.5}$ were lower than that at HY, especially in the summer and autumn, when the atmospheric diffusion conditions are relatively good.

Chemical characteristics of $PM_{2.5}$

General characteristics and chemical composition

This study carried out detailed chemical composition analyses of atmospheric particulates in Jiaxing, with an emphasis on the composition of 13 inorganic elements; four water-soluble ions; OC and EC; and 17 PAH compounds. Studies have shown that the amount of organic matter (OM) in the atmosphere is 1.4–1.6 times that of OC in urban areas (Pathak et al. 2011; Yang et al. 2005). The factor of 1.6 is adopted in this work. The mass ratios of the main chemical components of $PM_{2.5}$ and the composition of inorganic elements at the five sites were shown in Fig. 4. The total content of inorganic elements in $PM_{2.5}$ at each site was only 3.67%–8.54%. The content of inorganic elements in GQ was significantly higher than those at other sites because of the severe dust problem in GQ: The roads are badly damaged, and there are several irregular coal yards, docks, and unhardened parking lots (http://www.jepb.gov.cn/News/171/06229b8e959c71b4afa57eaf0241d1854d8b1c25.html). Furthermore, the crustal elements from natural sources at each site were abundant. Ca, Na, Fe, K, and Si accounted for 80% to 88% of the inorganic element concentration, while Mg and Al presented an average of 12%. The unidentified components accounted for 11 to 19% of the total $PM_{2.5}$ samples. In Fig. 5, except for inorganic elements, the concentrations of the main components at the five sites in winter and spring were all higher than those in autumn and summer. This feature was basically consistent with the seasonal distribution of $PM_{2.5}$ mass concentration.

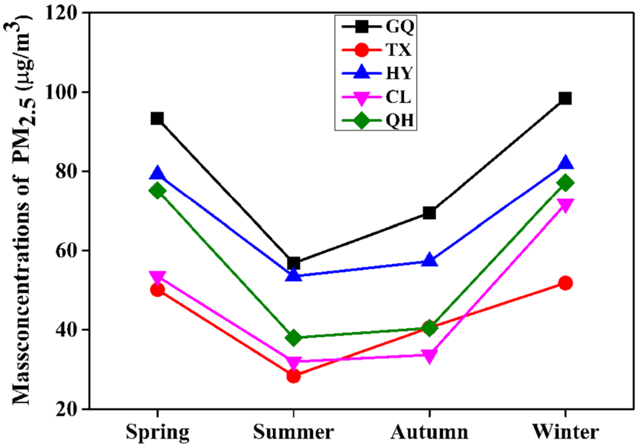

Fig. 3 Seasonal variation in $PM_{2.5}$ concentrations at the five sites: Gangqu site (GQ), Tongxiang site (TX), Heyuan site (HY), Canlian site (CL), Qinghe site (QH)

Water-soluble inorganic ions

It can be seen from Fig. 4 that the water-soluble ions at each site (except for HY) constituted the largest part of $PM_{2.5}$, accounting for 34.27–49.6%. SNA (sum of sulfate, nitrate and ammonium) was mainly the product of a photochemical reaction of NO_x, SO_2, NH_3, and other precursors (Squizzato et al. 2013). They were indicators of secondary inorganic aerosol (SIA). The high secondary ion content in $PM_{2.5}$ at each site indicated that secondary inorganic ions were an important cause of atmospheric heavy pollution. The value of $[NO_3^-]/[SO_4^{2-}]$ is often used to characterize the relative contributions of mobile sources and stationary sources. If the ratio of $[NO_3^-]/[SO_4^{2-}]$ is greater than 1, it indicates that mobile sources, such as vehicle exhaust, contribute more than stationary sources, such as coal combustion (Arimoto

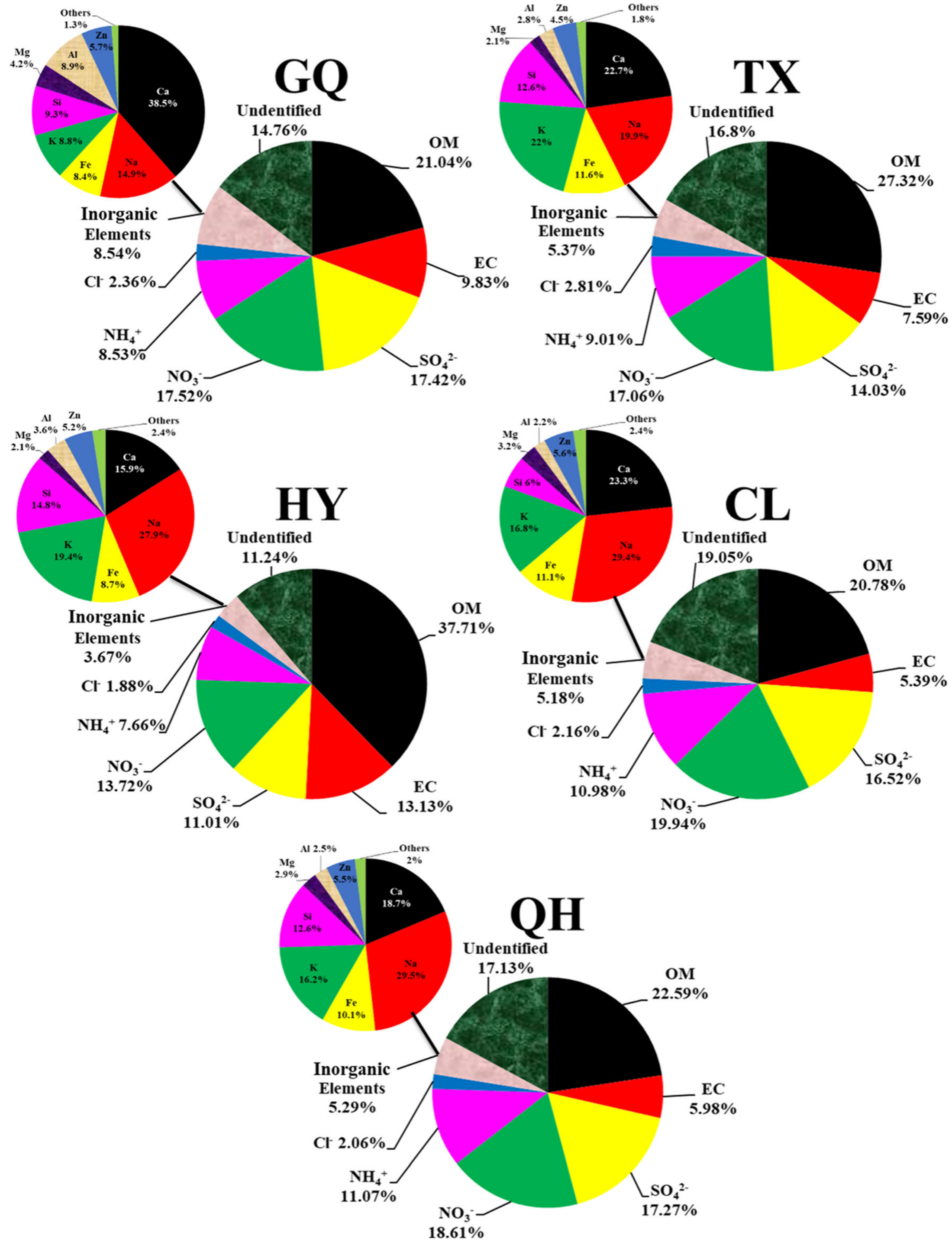

Fig. 4 Mass fraction (%) of major chemical components and some unidentified material in $PM_{2.5}$ and the composition of inorganic elements (others include Mn, Ni, V, As, Cr). Gangqu site (GQ), Tongxiang site (TX), Heyuan site (HY), Canlian site (CL), Qinghe site (QH)

et al. 1996). The average ratios of $[NO_3^-]/[SO_4^{2-}]$ at the five sites were greater than 1 (Fig. 6), indicating that the pollution from mobile sources in Jiaxing was severe. The average ratios of $[NO_3^-]/[SO_4^{2-}]$ at the five sites were also higher than that recorded for Shanghai (0.64) in 2005 (Wang et al. 2006a, b), indicating that mobile sources have become more and more dominant in the YRD.

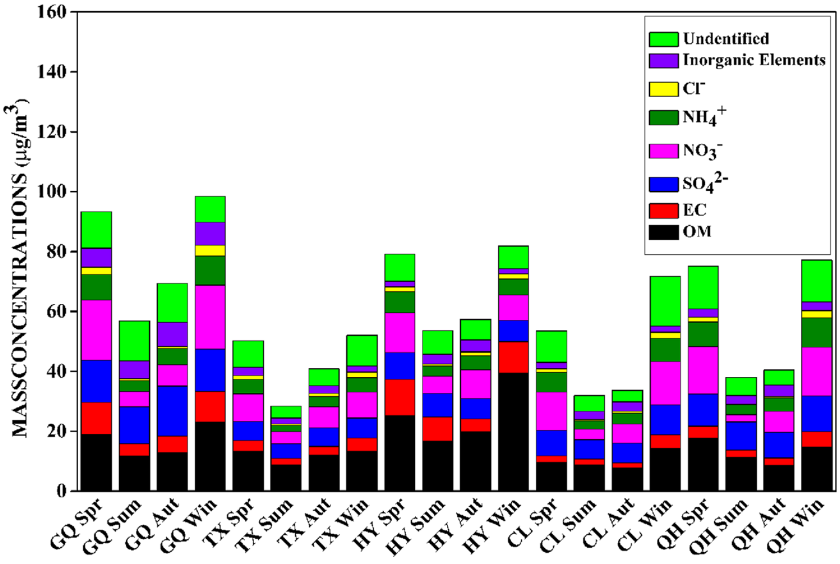

Fig. 5 Seasonal variations in components of $PM_{2.5}$ at the five sites: Gangqu site (GQ), Tongxiang site (TX), Heyuan site (HY), Canlian site (CL), Qinghe site (QH). Spring (Spri), Summer (Sum), Autumn (Aut), Winter (Win)

Carbon components

Carbonaceous aerosols, including OC and EC, mainly come from industrial boilers/kilns and residential stoves, steel production, and the incomplete combustion of automotive fuel (Bond et al. 2007). Total carbonaceous aerosols (TCA), i.e., the sum of EC and OM, contributed 31%, 50%, 26%, 29%, and 35% to the $PM_{2.5}$ mass concentrations in GQ, HY, CL, QH, and TX, respectively. In Nanjing (Li et al. 2015) and Shanghai (Cao et al. 2013), TCA accounts for 23% and 32% of the $PM_{2.5}$ mass concentration, respectively. It can be seen that the ratio of TCA/$PM_{2.5}$ in Jiaxing was significantly higher than that in Nanjing. Except in HY, the proportions of TCA in Jiaxing and Shanghai were similar. The ultra-high TCA/$PM_{2.5}$ ratio of HY might be related to the severe vehicle exhaust problem in this location: close to highway, coach station, logistics center, and military airfield.

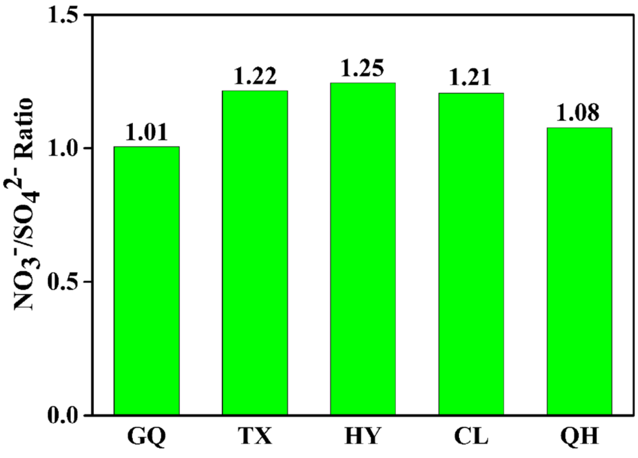

Fig. 6 NO_3^-/SO_4^{2-} ratios at the five sites: Gangqu site (GQ), Tongxiang site (TX), Heyuan site (HY), Canlian site (CL), Qinghe site (QH)

EC, which is a by-product of the incomplete combustion of carbonaceous materials or pollution migration in surrounding areas, has good chemical stability and is generally used as a tracer for human-made primary pollution sources (Turpin and Huntzicker 1995). The sources of OC are relatively extensive, ranging from primary organic carbon (POC) emitted directly from pollution sources to secondary organic carbon (SOC) generated by chemical reactions in the atmosphere. Therefore, the concentration ratio of OC to EC is often used to evaluate the contribution of secondary sources in atmospheric particles. Researchers compared OC/EC in cities around the world and found that OC/EC > 2 indicated the presence of secondary organic aerosol (SOA) (Chow et al. 1996; Turpin et al. 1990). The higher the ratio, the greater the SOA that have been generated. According to relevant research results (Song et al. 2007; Zhang et al. 2013), we could also adopt 1.6 for the conversion of SOA and SOC.

There is currently no direct way to separate POC from SOC. The EC tracer-based method is a widely applied approach, using EC as the tracer for primary sources and calculating POC in ambient samples with the OC/EC ratios obtained from primary emissions and measured EC levels (Turpin and Huntzicker 1995). However, under normal circumstances, the OC/EC ratio varies with different types of emission sources and can be influenced by many factors, such as the ambient temperature and the carbon element determination method. Therefore, the minimum ratio of test data ($(\mathrm{OC}/\mathrm{EC})_{\min}$) can be used instead of the OC/EC ratio (Castro et al. 1999). The estimation equation is as follows:

$$\mathrm{SOC} = \mathrm{OC} - \mathrm{EC} \times \left(\frac{\mathrm{OC}}{\mathrm{EC}}\right)_{\min}$$

Owing to the introduction of polar functional groups, such as oxygen and nitrogen, SOC has stronger polarity and hygroscopicity, and has an important influence on visibility reduction, ash formation, and climate change. Many studies have showed that SOC has become an important component of atmospheric particulates, especially fine particulates, and the proportion of SOC in organic carbon is 30%–52% (Cao et al. 2004; Lin and Tai 2001; Na et al. 2004). The SOC/OC ratio in each season at the five sites varied from 35% to 77% (Fig. 7), significantly higher than that in Shanghai (~30%) (Feng et al. 2009), indicating that the secondary organic pollution in Jiaxing was severe. Except in the autumn, the SOC/OC ratio at HY was significantly higher than those at other sites, which may be related to its location: close to highways, main roads, bus terminals, logistics centers, and military airports. The SOC/OC ratio at TX was significantly higher than those at other sites in the autumn. This may be due to the large area of cultivated land in Tongxiang District and the widespread burning of straw in the autumn. As shown in Fig. 7, the seasonal patterns of the SOC/OC ratios at each site were not consistent with other YRD cities where higher ratios were also found in the summer than in other seasons (Hou et al. 2011; Cao et al. 2007). The higher SOC/OC in winter and spring resulted partly from a lower $(OC/EC)_{min}$ than that in summer and autumn.

PAHs

PAHs are volatile fused ring compounds produced by incomplete combustion of organic matter such as coal, petroleum, tobacco, and wood. More than 200 species have been discovered so far. The US Environmental Protection Agency (EPA) has classified 16 compounds as superior-grade polycyclic aromatic hydrocarbons: naphthalene, acenaphthylene, acenaphthene, fluorene, phenanthrene, anthracene, fluoranthene, pyrene, benzo[a]anthracene, chrysene, benzo[b]fluoranthene, benzo[k]fluoranthene, benzo[a]pyrene, indeno[1,2,3-cd]pyrene, dibenzo[a,h]anthracene, and benzo[g,h,i]perylene.

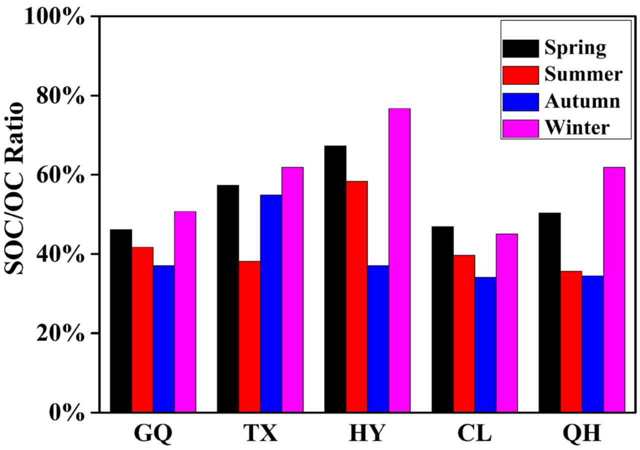

Fig. 7 Seasonal characteristics of SOC/OC ratios at the five sites: Gangqu site (GQ), Tongxiang site (TX), Heyuan site (HY), Canlian site (CL), Qinghe site (QH)

We analyzed 16 kinds of superior-grade PAHs and benzo[e]pyrene (BeP), of which there are one, five, four, five, and two kinds of 2–6 rings PAHs. PAHs in the atmosphere can be gaseous and particulate, with low molecular weight PAHs with 2–3 ring mainly existing in gaseous form. The 2-ring PAHs, such as naphthalene, are extremely volatile. The distribution of 4-ring PAHs in gaseous and granular states is basically the same. The PAHs with 5–7 rings, owing to their large molecular weight, exist mostly in particulate form. In Fig. 8a, we focused on the data of PAHs with 4–6 rings at each site for comparison. The total concentrations of PAHs with 4–6 rings at the five sites in winter and spring were significantly higher than those in summer and autumn. This can be partly attributed to the high temperature and strong sunlight, which causes the PAHs to undergo a photochemical reaction, generating regeneration contaminants such as nitro and oxi PAHs (Bidleman et al. 1986; Schauer et al. 2003).

Of the 16 superior-grade PAHs, benzo[a]pyrene (BaP) is highly carcinogenic, so it is often used as a representative of PAHs (Yassaa et al. 2001). The concentrations of BaP at the five sites ranged from 0.07 to 1.95 ng/m^3, with an average of 0.79 ng/m^3, which was significantly lower than that in Hangzhou (1.6 ng/m^3) (Feng et al. 2015). Only very few days exceeded the new air quality standard of China (1 ng/m^3). BaP is unstable and prone to photochemical reactions. Most of the fresh exhaust gases had similar BaP and BeP content, the ratio of BeP/(BeP + BaP) could be regarded as an index of the aging of particles (Muránszky et al. 2011). In addition, the sources of atmospheric particles can be divided into two types: local generation and external transportation. Owing to long-distance transportation, foreign particles inevitably age compared to local particles. Therefore, during the same season, under similar weather conditions, the ratio of BeP/(BeP + BaP) could also reflect the relative contribution of foreign particles. In this study, the ratios of BeP/(BeP + BaP) in the summer were the highest at the five sites (Fig. 8b) because of the preferential loss of BaP through photo-degradation. The ratios of BeP/(BeP + BaP) during all the seasons were greater than 0.5, indicating that $PM_{2.5}$ in Jiaxing had generally aged, and there was a certain degree of external transportation problems.

The source apportionment of $PM_{2.5}$

In this study, PMF 5.0 issued by the US EPA was used to calculate the total of 440 valid sample data from five sites. The input sample data included 19 chemical components, $PM_{2.5}$ mass concentration, and the uncertainty data for each component. Eight sources were obtained via minimized Q value with an optimal simulation result (Fig. 9).

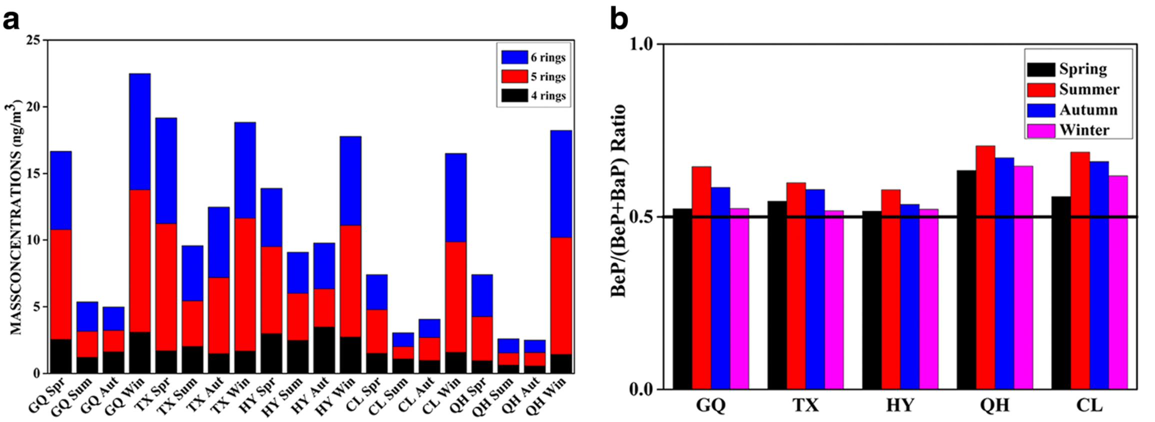

Fig. 8 **a** Concentrations of PAHs with 4–6 rings and their percentage contributions to the total concentration of PAHs. **b** Seasonal variations in the BeP/(BeP + BaP) ratios at the five sites: Gangqu site (GQ), Tongxiang site (TX), Heyuan site (HY), Canlian site (CL), and Qinghe site (QH)

Source 1 was designated as secondary nitrates because it was enriched with nitrate (NO_3^-) and ammonium (NH_4^+) ions, as well as some sulfate (SO_4^{2-}) ions. Source 2 was identified as secondary sulfates because of the high abundance of SO_4^{2-} and NH_4^+ in the source profile. These two sources could be collectively referred to as secondary inorganic aerosols (SIA). SNA is the main SIA component in $PM_{2.5}$ and is mainly present in the form of $(NH_4)_2SO_4$ and NH_4NO_3. Both are produced by neutralizing sulfuric acid (H_2SO_4) and nitric acid (HNO_3) with ammonia (NH_3), respectively (Squizzato et al. 2013; Stockwell et al. 2003).

OC had the highest contribution rate to source 3 and was much higher than EC, so source 3 was classified as SOA. Source 4 was vehicle exhaust, which was identified by the high contribution of EC. Many studies have shown that EC is a characteristic component of motor vehicle exhaust (Lin et al. 2015; Chan et al. 1999).

Source 5 represented coal combustion sources. The relative content of Cl^- in this source was high. Yao et al. (2003) found that, in many cities in China, owing to coal burning, high concentrations of Cl^- are often present in $PM_{2.5}$ in winter. In addition, some studies also showed that Cl^- was a tracer element of coal-fired sources (Xiu et al. 2004; Duan et al. 2006; Han et al. 2010; Yu et al. 2011). Coal still occupies a considerable proportion in the energy structure of cities in China. Therefore, coal-fired sources have always been an important source of urban particulate matter.

Source 6 was estimated to be fugitive dust because it contains high percentages of the elements Si, Ca, Mg, Al, Fe, Ca, and Mg, which are used to identify cement dust (Xue et al. 2010; Kim et al. 2004). Cement dust originates from cement, gypsum plants, and construction areas. Many studies have also confirmed that soil dust contains a large number of crustal elements, such as Si, Al, Fe, Ca, and Mg (Lee et al. 1999; Chueinta et al. 2000; Zhang et al. 2013; Kong et al. 2010). The average percentage of SiO_2, Al_2O_3, Fe_2O_3, MgO, and CaO in soil are 57.8%–64.8%, 14.8%–16.7%, 4.6%–9.1%, 2.7%–4.4%, and 3.4%–7.5%, respectively.

Source 7 represented ships and heavy oil. Heavy oil is the part of crude oil that remains after gasoline and diesel oil are extracted. A significant source of heavy oil combustion is ship emissions and might also include land transport and industrial sources. Ni and V are common tracers of heavy oil combustion (Vallius et al. 2005). Yuan et al. used the PMF model to analyze the sources of PM_{10} in Hong Kong and also found a heavy oil combustion source with V as the marker, and considered it to be mainly from ship emissions (Yuan et al. 2006). As Jiaxing has a port and the river network is densely populated, ship and heavy oil pollution were common.

Source 8 had the highest characteristic value of K, and the characteristic values of Na, Zn, Cr, and Fe were also high. Owing to the use of potash fertilizer, crops are rich in K, and the particles released from combustion are rich in K, so K is considered an important tracer for biomass combustion (Simoneit et al. 1999). Some studies showed that the concentrations of Na (Yli-Tuomi et al. 2003; Begum et al. 2004) were high in the by-products of biomass burning in China. Zn, Fe, and Cr might be derived from garbage incineration, which is often carried out with open-air biomass combustion. They might also originate from other sources of pollution. Therefore, we classified factor 8 as biomass burning and others.

The source apportionment results in five sites were shown in Fig. 10: secondary nitrates (26.95%), secondary sulfates (15.49%), SOA (19.64%), vehicle exhaust (15.67%), coal combustion (8.6%), fugitive dust (7.7%), ships and heavy oil (5.23%), and biomass burning and other sources (0.91%), including three secondary sources and five primary sources.

First, the contribution of secondary aerosols (including SIA and SOA) to $PM_{2.5}$ was generally large, from 43.33% to 69.76%. The contributions of SIA at the CL and QH sites were significantly higher than those at other sites, which was

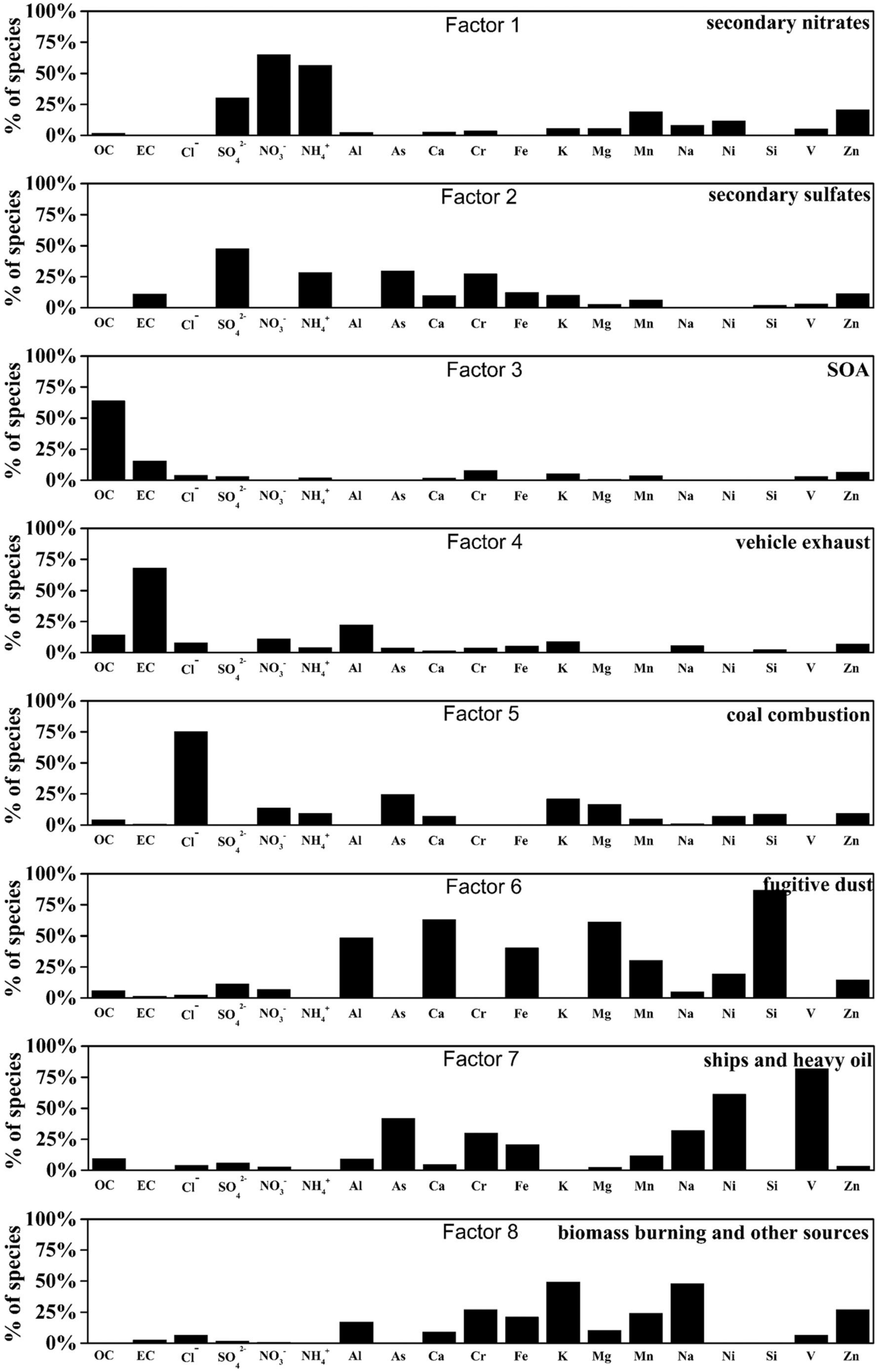
Factor 1
secondary nitrates
Factor 2
secondary sulfates
Factor 3
SOA
Factor 4
vehicle exhaust
Factor 5
coal combustion
Factor 6
fugitive dust
Factor 7
ships and heavy oil
Factor 8
biomass burning and other sources
% of species
100%
75%
50%
25%
0%
OC
EC
Cl^-
SO_4^{2-}
NO_3^-
NH_4^+
Al
As
Ca
Cr
Fe
K
Mg
Mn
Na
Ni
Si
V
Zn

Fig. 9 Source profiles (% of species) in Jiaxing obtained from the PMF analysis. Secondary nitrates, secondary sulfates, SOA, vehicle exhaust, coal combustion, fugitive dust, ships and heavy oil, biomass burning, and other sources were identified

consistent with the results of the component analysis. The contribution of vehicle exhaust at the HY site was much higher than those in the CL and QH sites because HY is close to highways, trunk roads, automobile passenger terminals, military airports, and logistics centers. The contribution rate of the SOA at the HY site was also the highest. In fact, motor vehicles produce OC and EC (Lin et al. 2015). Based on the findings in this study, the high SOC/OC ratio indicated that OC contained a large amount of SOC. According to relevant research results (Song et al. 2007; Zhang et al. 2013), SOA can be estimated by multiplying SOC by a factor of 1.6. Hence, there was a strong correlation between vehicle exhaust and SOA. The contribution rate of dust at GQ was the highest among the five sites, which was consistent with the severe dust pollution in GQ. The contribution rate of coal combustion at GQ was the highest compared to those at other sites and was almost twice that of TX. This is probably because of the industrial coal consumption in Gangqu District, which in 2015 was nearly seven times that of Tongxiang District (Report on

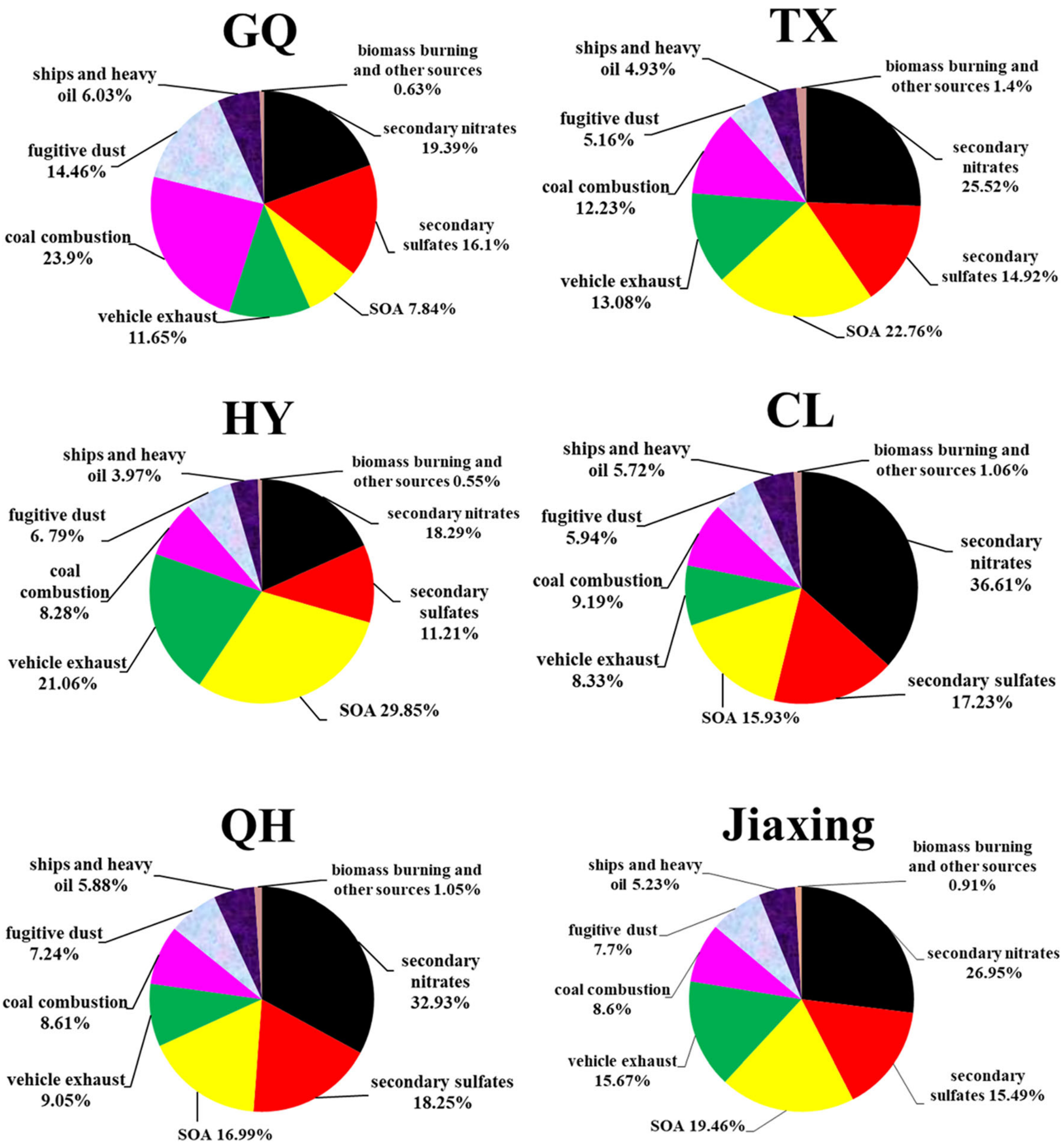

Fig. 10 Contribution rate of $PM_{2.5}$ pollution sources in Jiaxing. Secondary nitrates, secondary sulfates, SOA, vehicle exhaust, coal combustion, fugitive dust, ships and heavy oil, biomass burning, and other sources were identified. Gangqu site (GQ), Tongxiang site (TX), Heyuan site (HY), Canlian site (CL), Qinghe site (QH)

Source Release of Atmospheric Pollutants in Jiaxing City (2015)). Owing to the large number of rivers in Jiaxing, there are heavy oil pollution problems at each site, and there was little difference in contribution rates between the sites. The contributions due to ships and heavy oil on GQ was greater than those at other sites, as GQ is near a port. With respect to biomass burning and others, the contribution rates at all the sites were generally very low, and the contribution rate at TX was the highest, as TX is located in an ecological suburb and has a large area of cultivated land. A previous study found that, in 2010, biomass burning accounted for about 10% of the primary emission of $PM_{2.5}$ in the YRD region (Fu et al. 2013). Therefore, our findings seem to suggest that the problem of biomass burning in Jiaxing has greatly improved in recent years, even in Tongxiang District, where the cultivated land is large.

Conclusion

Jiaxing City is located in the Yangtze River Delta economic circle. The local and surrounding pollution sources are complex and diverse. Therefore, many kinds of particulate matter were observed in the SEM and TEM images; however, the artificial sources represented by vehicle exhaust and coal combustion might be the main sources.

The seasonal variation in $PM_{2.5}$ concentration in Jiaxing was as follows: winter > spring > autumn > summer, due to the climate characteristics of Jiaxing City. The $PM_{2.5}$ concentration levels in GQ (closing to the port) and HY (closing to many traffic sources) were relatively high, indicating that a mobile source was an important source of fine particles. Carbon components and water-soluble ions exceeded three quarters of the total mass of $PM_{2.5}$, and their seasonal variation was similar to that of $PM_{2.5}$ concentration.

The $[NO_3^-]/[SO_4^{2-}]$ ratio of Jiaxing City was higher than 1 and higher than that of Shanghai in 2005, indicating that the influence of mobile sources in YRD region had increased in recent years and was already dominant. The $TCA/PM_{2.5}$ ratio in Jiaxing was higher than that in Nanjing, which was equivalent to Shanghai, except for the HY. The SOC/OC ratio of Jiaxing was higher than that of Shanghai, and its seasonal characteristics were different from that obtained for the YRD region. The average annual concentration of BaP in Jiaxing (0.79 ng/m^3) was much lower than that in Hangzhou (1.6 ng/m^3), and lower than the Chinese national standard (1 ng/m^3). The BeP/(BeP + BaP) ratio showed that $PM_{2.5}$ in Jiaxing had generally aged and was affected by external sources.

The results of the source apportionment showed that secondary aerosols dominate; therefore, it is important to control the emissions of the gaseous precursors to reduce $PM_{2.5}$ concentrations. Compared with the pollution sources at other sites, vehicle exhaust in HY, coal combustion and fugitive dust in the GQ, and biomass burning in TX contributed prominently to the $PM_{2.5}$ concentrations.

In short, the pollution status of $PM_{2.5}$ in Jiaxing had both commonalities with and was different from those in the surrounding cities. There were also differences at each site. Therefore, control policies in different regions should focus on different sources, and a refined control strategy should be formulated.

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Compliance with ethical standards

Conflict of interest The authors declare no competing financial interest.

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