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Chemical compositions of $PM_{2.5}$ aerosol during haze periods in the mountainous city of Yong'an, China

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Abstract

Haze phenomena were found to have an increasing tendency in recent years in Yong'an, a mountainous industrial city located in the center part of Fujian Province, China. Atmospheric fine particles ($PM_{2.5}$) in the urban area during haze periods in three seasons (spring, autumn and winter) from 2007 to 2008 were collected, and the mass concentrations and chemical compositions (seventeen elements, water soluble inorganic ions (WSIIs) and carbonaceous species) of $PM_{2.5}$ were determined. $PM_{2.5}$ mass concentrations did not show a distinct difference among the three seasons. The carbonaceous species organic carbon (OC) and elemental carbon (EC) constituted up to 19.2%–30.4% of the $PM_{2.5}$ mass during sampling periods, while WSIIs made up 25.3%–52.5% of the $PM_{2.5}$ mass. The major ions in $PM_{2.5}$ were SO_4^{2-}, NO_3^- and NH_4^+, while the major elements were Si, K, Pb, Zn, Ca and Al. The experimental results (from data based on three haze periods with a 10-day sampling length for each period) showed that the crustal element species was the most abundant component of $PM_{2.5}$ in spring, and the secondary ions species (SO_4^{2-}, NO_3^-, NH_4^+, etc.) was the most abundant component in $PM_{2.5}$ in autumn and winter. This indicated that dust was the primary pollution source for $PM_{2.5}$ in spring and combustion and traffic emissions could be the main pollution sources for $PM_{2.5}$ in autumn and winter. Generally, coal combustion and traffic emissions were considered to be the most prominent pollution sources for this city on haze days.

Key words: $PM_{2.5}$; haze; metal elements; water soluble inorganic ions; carbonaceous species; Yong'an City
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Introduction

Haze is a phenomenon of heavy atmospheric pollution, which occurs when sunlight encounters tiny particles in the air. Haze is defined by the China Meteorological Agency as a phenomenon which leads to atmospheric visibility less than 10 km due to suspended particles, smoke, and vapor in the atmosphere, and it has adverse effects on air quality, human health, visibility and even the global climate (Chameides et al., 1999; Okada et al., 2001; Yadav et al., 2003).

Fine particles ($PM_{2.5}$) are considered to play an important role in the formation of haze (Watson, 2002). The chemical constituents of $PM_{2.5}$, such as water soluble inorganic ions (WSIIs) and carbonaceous species, mainly composed of organic carbon (OC) and elemental carbon (EC), have been found to be widely associated with health problems (Englert, 2004; Cormier et al., 2006; Ostro et al., 2007) and visibility degeneration (Christoforou et al., 2000; Kim et al., 2006). Therefore, it is crucial to investigate the chemical characteristics and source apportionment of particulate matter, especially for fine particles during haze.

The mountainous city Yong'an is one of three high-incidence haze areas in Fujian Province. Research has been focused on the increasing haze occurrence in Fuzhou and Xiamen, the other two cities in Fujian Province. However, haze pollution in Yong'an has received less attention.

According to the historical data of air quality observation during the last 40 years (1968–2007), supplied by the Fujian Provincial Environmental Monitoring Center, annual haze days in Yong'an increased rapidly from 1983 with more than 5 haze days in each month. Yong'an City has experienced especially rapid urbanization and industrialization in recent years, and atmospheric quality in the urban area was mainly between grades two and three of the GB3095–1996 criterion during 2006–2010, showing a deteriorating trend. However, there are relatively little data available on the chemical compositions of $PM_{2.5}$ in Yong'an City, particularly on haze days. Therefore, the purposes of this study were (1) to systematically investigate variations of chemical compositions of $PM_{2.5}$ on haze days during different seasons; (2) to preliminarily identify the sources of pollutants for haze days.

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1 Experiments

1.1 Sites and sampling

The industrial city Yong'an is located in the central part of Fujian Province. The city lies in the transition zone between the Wuyi and Daiyun Mountains, and the urban area is surrounded by rivers and mountains. The city has a typical subtropical monsoon climate, with sufficient annual rainfall and abundant sunshine. The annual average ambient temperature, relative humidity, and wind speed in Yong'an are 19.1°C, 78%, and 1.7 m/sec, respectively. The prevailing wind direction is southerly in summer but northeasterly in other seasons, and static wind days (< 0.5 m/sec) occur frequently.

$PM_{2.5}$ aerosol samples in this study were collected on the rooftop (18 m above the ground) of the Yong'an environmental monitoring station (117°21′32″N, 25°57′56″E), with no pollution source nearby. The station is situated in the mid-east of the urban area, downstream of the dominant wind direction, and might be influenced by traffic to a certain extent due to its short distance from the main street. The sampling site in the urban area is located in a basin surrounded by mountains, and the wind speed was low on most sampling days. As a consequence, atmospheric pollutants could not easily disperse out of the urban area quickly. Considering the terrain, the pollution condition of samples collected at the site should be representative of the urban area of Yong'an. A map of the sampling site and the urban area of Yong'an is depicted in Fig. 1.

A Minivol $PM_{2.5}$ sampler (AirMetrics Com., USA) was employed to collect particulates for WSIIs and elemental analysis with polypropylene filters (PPFs, Pall, USA), while a medium-volume sampler (TH-50C III Tianhong, China) was used to collect particulates for carbonaceous species analysis with quartz fiber filters (QFFs, Whatman, UK). The QFFs were pre-annealed for 5 hr at 450°C in a furnace, and PPFs were pre-heated for 2 hr at 60°C in an oven. All filters were kept in clean dishes within sealed polyethylene plastic bags before use and after sampling.

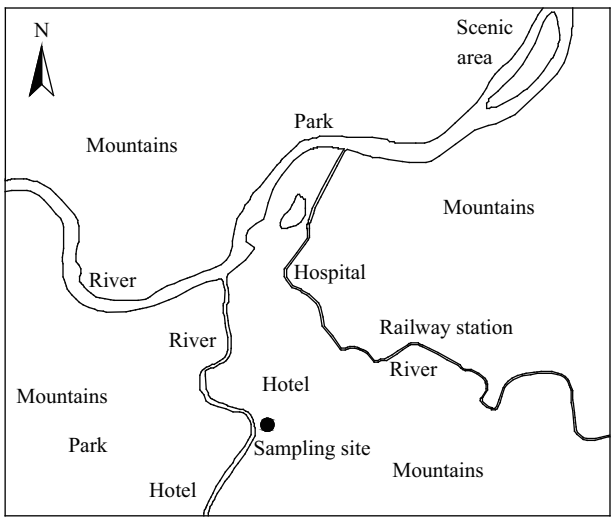

Fig. 1 Map of sampling site and the urban area of Yong'an, Fujian, China.

The loaded filters were stored at –20°C.

Aerosol sampling was conducted in three different seasons: spring (2007.4.11–20), autumn (2007.11.19–28) and winter (2008.1.10–19) when haze occurred (the visibility was less than 10 km while the relative humidity was lower than 85%). The duration of each sampling was about 24 hr, and a consecutive 10-day sampling period was carried out for each seasons. There were 10 samples collected for each season's analysis. The sampling process was paused when it rained.

1.2 $PM_{2.5}$ mass concentrations analysis

After the filters were stabilized under constant temperature (25 ± 1°C) and relative humidity (50% ± 1%) in a chamber (HWS-080, Jingyi, China) for 24 hr, the $PM_{2.5}$ particle mass loadings were obtained gravimetrically by an analytical balance (Sartorius 0.01 mg, Germany), and determined from the weights after sampling minus those before sampling. The typical uncertainty for the gravimetric measurements was ±20 μg, which represents less than ±5% of the total aerosol mass of the field samples.

1.3 Elemental analysis

Seventeen elements including Mg, Al, Si, K, Ca, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn, As, Se, Br and Pb were detected by the particle-induced X-ray emission (PIXE) method in the Institute of Low Energy Nuclear Physics of Beijing Normal University. The PIXE analysis was carried out using 2.5 MeV proton bombardments with a beam of 30–40 nA. The analysis instrument was a GIC4117 2 × 1.7 MV tandem accelerator (GIC Co., USA). All the elemental concentrations were corrected for the background from blank filters.

1.4 Water-soluble inorganic ions analysis

One-fourth of each blank and sample quartz fiber filter was cut into fine strips and dipped in 10 mL ultrapure water (18.2 MΩ·cm), and water-soluble inorganic ions (WSIIs) from the samples were extracted by ultrasonic bath for 40 min. The extract solutions were filtered with 0.45 μm PTFE syringe filters (Pall Co., Ltd., USA). An ion chromatography system (ICS-3000, Dionex, USA) was used to determine the concentrations of four anion (F^-, Cl^-, NO_3^- and SO_4^{2-}) and five cation (Na^+, K^+, NH_4^+, Ca^{2+} and Mg^{2+}) species. Anion analysis parameters: IonPac AS11-HC (250 mm × 4 mm) analytical column, ASRS-4 suppressor and conductivity detector, 10 mmol/L OH^- eluent generated automatically by the Dionex AutoRegen Accessory. Cation analysis parameters: IonPac CS12A (250 mm × 4 mm) analytical column, CSRS-4 suppressor and conductivity detector, 10 mmol/L H_2SO_4 used as eluent. Standard solution was purchased from the National Research Center of Certified Reference Materials, China.

1.5 Carbonaceous species (OC and EC) analysis

A Desert Research Institute (DRI) Model 2001 Thermal Optical Carbon Analyzer (Atmoslytic Inc., Calabasas, USA) was applied for OC and EC analysis. A 0.5-cm^2 punch from the quartz fiber filters was analyzed following

the IMPROVE thermal optical reflectance (TOR) protocol (Cao et al., 2003; Chow et al., 2004). This procedure produced stepwise: four OC fractions (OC1, OC2, OC3 and OC4 at 120, 250, 450 and 550°C, respectively, in a non-oxidizing atmosphere of helium) and three EC fractions (EC1, EC2 and EC3 at 550, 700 and 800°C, respectively, in an oxidizing atmosphere of 2% oxygen with 98% helium). The carbon compounds evolved at each temperature were oxidized to carbon dioxide, and then reduced to methane for quantification with a flame ionization detector (FID) system. The detection limit for EC and OC was 0.1 μg C/cm^2.

1.6 Haze observation and meteorological data

There is still no generally accepted definition of the relative humidity range when haze occurs. According to the climate traits in Fujian Province, the haze was judged to be occurring when the visibility was less than 10 km and the relative humidity was less than 85%. Meteorological data, including ambient temperature, relative humidity, wind speed, wind direction and concentrations of two kinds of gaseous pollutants (SO_2 and NO_2) were recorded during sampling periods.

2 Results and discussion

2.1 Haze pollution and relative meteorological factors

Meteorological conditions during the sampling periods are shown in Fig. 2. From the analysis of atmospheric gaseous and particulate pollution and the meteorological conditions in Yong'an during recent years (2005–2008), supplied by Fujian Provincial Environmental Monitoring Center and the Institute of Meteorological Science of Fujian Province, there were two factors that had a strong influence on atmospheric pollution: rainfall and temperature.

Rainfall in the spring was much more than in other seasons, and the concentrations of gaseous pollutants (SO_2 and NO_2) were the lowest in this season, while the concentrations of particulates ($PM_{2.5}$) did not present the same variations as the gaseous pollutants. It appeared that the rainfall has more scavenging ability for gaseous than particulate pollutants.

The daily data of temperature, relative humidity and wind speed, and the daily variation of these meteorological conditions is presented in Fig. 2. Relative humidity stayed relatively high during the 30 sampling days, while the temperature and wind speed for most days changed slightly during each of the sampling periods of the three seasons. The average values of relative humidity in the three sampling periods were similar to each other, which was also the case for wind speed. It was found that high relative humidity and low wind speed were favorable for haze formation in Yong'an.

2.2 Mass concentrations of $PM_{2.5}$

The mass concentrations of $PM_{2.5}$ are presented in Table 1 and Fig. 2. Daily variation of $PM_{2.5}$ concentrations was apparent during each 10-day period, however, the average $PM_{2.5}$ concentrations of the three sampling periods were close to each other. Most of the $PM_{2.5}$ concentrations exceeded the National Ambient Air Quality Standard (NAAQS) daily average value of 65 μg/m^3 for $PM_{2.5}$ (US EPA, 1997), and the three average concentrations for each season were much higher than the NAAQS annual average value of 15 μg/m^3. The average concentration

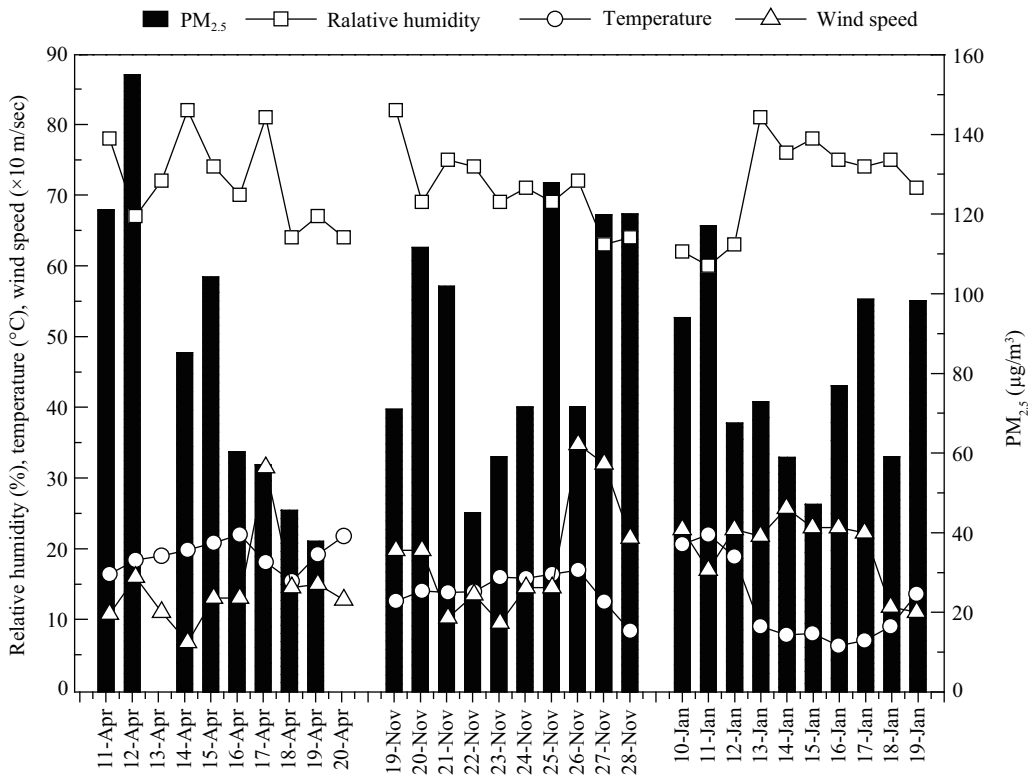

Fig. 2 Daily variation of meteorological parameters in haze sampling periods ($PM_{2.5}$ data of 13-Apr and 20-Apr are not available).

Table 1 Meteorological conditions and concentrations of $PM_{2.5}$, carbonaceous species, SO_2 and NO_2 in haze sampling periods

	Spring	Autumn	Winter
Temperature (°C)	19.09 ± 2.14	14.03 ± 2.52	12.21 ± 6.09
RH (%)	71.83 ± 6.64	70.73 ± 5.41	71.30 ± 7.26
WS (m/sec)	1.44 ± 0.65	1.90 ± 0.86	2.01 ± 0.51
$PM_{2.5}$ (μg/m^3)	83.26 ± 40.90	89.93 ± 29.54	79.01 ± 22.15
SO_2 (μg/m^3)	56.6 ± 32.4	46.6 ± 13.3	66.0 ± 45.5
NO_2 (μg/m^3)	46.5 ± 24.2	32.1 ± 0.17	30.8 ± 11.4
TC (μg/m^3)	15.98 ± 5.18	17.28 ± 6.23	24.05 ± 6.45
OC (μg/m^3)	12.90 ± 4.31	13.25 ± 5.17	16.81 ± 4.79
EC (μg/m^3)	3.08 ± 0.92	4.03 ± 1.14	7.24 ± 2.05
OC/EC	4.18 ± 0.57	3.24 ± 0.53	2.38 ± 0.51

Data are presented as mean ± standard deviation.

of $PM_{2.5}$ during haze days was 84.02 μg/m^3, which was correspondingly lower than those in Shanghai (90.3–95.5 μg/m^3) (Feng et al., 2009), Beijing (222.81 μg/m^3) (Sun et al., 2006) and Guangzhou (235.50 μg/m^3) (Tan et al., 2009a) in China.

The concentrations of SO_2, NO_2 might not have a direct relationship with the mass of $PM_{2.5}$, except in the spring period, when SO_2 and the mass of $PM_{2.5}$ had a certain degree of correlation with $R^2 = 0.61$. This implied that the ambient SO_2 and its derivatives can be important contributors to $PM_{2.5}$ in spring.

2.3 Carbonaceous species in $PM_{2.5}$

Total carbon (TC), OC and EC are summarized in Table 2. OC had complicated sources, including direct emissions and secondary formation of OC via gas-to-particle conversion, whereas EC was mostly emitted from primary combustion (Lonati et al., 2007). The average concentration of OC in $PM_{2.5}$ in Yong'an (14.32 ± 4.94 μg/m^3) was higher than those in Hong Kong (9.6 μg/m^3) (Cao et al., 2003), Kaohsiung (10.4 μg/m^3) (Lin and Tai, 2001) and some European cities such as Amsterdam (5.3 ± 2.0 μg/m^3) (Viana et al., 2007), and Ghent (4.1±1.9 μg/m^3) (Viana et al., 2007), but lower than those in Guangzhou (18.4 ± 11.1 μg/m^3) (Feng et al., 2006), Beijing (21.4 ± 5.4 μg/m^3) (Feng et al., 2006), Taiyuan (28.9 ± 14.8 μg/m^3) (Meng et al., 2007) and Xi'an (61.9) (Cao et al., 2005), and it was close to that in Shanghai (14.7 ± 17.4 μg/m^3) (Feng et al., 2009). The average concentration of EC in $PM_{2.5}$ in Yong'an (4.78 ± 2.29 μg/m^3) was higher than those in Kaohsiung (4.0 μg/m^3) (Lin and Tai, 2001), Amsterdam (1.8 ± 0.1 μg/m^3) (Viana et al., 2007), and Ghent (1.0 ± 0.3 μg/m^3) (Viana et al., 2007), but lower than those in Beijing (5.7 ± 0.0 μg/m^3) (Feng et al., 2006), and Guangzhou (6.4 ± 0.4 μg/m^3) (Feng et al., 2006), Xi'an (12.3 μg/m^3) (Cao et al., 2005), and it was close to those in Taiyuan (4.8 ± 2.2 μg/m^3) (Meng et al., 2007) and Hong Kong (4.7 μg/m^3) (Cao et al., 2003). Generally, it seemed that the concentrations of OC and EC in $PM_{2.5}$ in Yong'an were higher than those in the European cities, and lower or close to those in domestic cities.

It was obvious that concentrations of TC, OC and EC peaked in winter. TC constituted up to 19.2%, 19.2% and 30.4% of $PM_{2.5}$ mass in the three seasons, respectively, which indicated that carbonaceous aerosols were one of the key components of fine particles in Yong'an. At the same time, TC had an apparent correlation with $PM_{2.5}$, especially in spring (R^2: 0.82, 0.43, and 0.73 in the three seasons, respectively). Therefore, TC had a comparatively important influence on the $PM_{2.5}$ mass concentration.

Ratios of OC to EC concentrations (OC/EC) can be used to study the emission source and transformation characteristics of carbonaceous species. Typical emission sources include diesel- and gasoline-powered vehicular exhaust (OC/EC: 1.0–4.2) (Schauer et al., 1999), wood combustion (16.8–40.0) (Schauer et al., 2001), residential coal smoke (2.5–10.5) (Chen et al., 2006), kitchen emissions (32.9–81.6) (He et al., 2004) and biomass burning (7.7) (Zhang et al., 2007), etc. The maximum OC/EC ratio in Yong'an could be found in spring (4.18 ± 0.57), and the minimum OC/EC ratio appeared in winter (2.38 ± 0.51). Therefore, vehicle emission, coal combustion and biomass burning could be the major contributors to the carbonaceous species in $PM_{2.5}$. The OC/EC ratio of Yong'an was close to the results of Guangzhou (3.2–4.7 for haze days and 2.0–2.8 for normal days) (Tan et al., 2009b). Based on the correlation ($R^2 = 0.56$) between OC and K (an indicator of biomass burning as discussed below) in spring, a biomass burning origin with higher OC/EC ratios (Fine et al., 2004) could be deduced in this season. The relationships between OC and EC could be used to infer the origins of carbonaceous particles (Turpin et al., 1991; Turpin and Huntzicker, 1995). OC concentrations were well correlated ($R^2 = 0.88$, 0.85 and 0.55) with EC in the three seasons, especially in spring, showing that most OC and EC was derived from common dominant sources in this period.

2.4 Elemental compositions

Different elements' concentrations varied over a large range as shown in Table 2. Si, K, Pb, Zn, Ca, and Al dominated the detected elements in $PM_{2.5}$ in spring, accounting for 93.6% of total elemental mass, while K, Si, Pb, Ca accounted for 75.0% of total elemental mass in autumn $PM_{2.5}$, and K, Ca, Si accounted for 81.2% of total elemental mass in winter. Si is known as the second most abundant element in the Earth's crust and existed in oxide form in granules. Ca, Fe, K, and Al are also indicator elements for crustal dust (Yang et al., 2004). The concentration of the typical crustal element Si was much higher in spring than the other two seasons, indicating an important contribution of dust emitted from the desert or semiarid areas in North or Northwestern China (Wai et al., 2005; Fang et al., 2002) to Yong'an during this season. Biomass was a major additional source for K (Yang et al., 2009). The high K mass concentrations during spring, autumn, and winter were likely due to biomass burning in the surrounding rural area of Yong'an, e.g., rice straw was often burned in farm fields in autumn and winter.

Al is considered as a typical marker of soil and Ca can be derived from both soil dust and construction materials in urban aerosols. The Ca/Al ratio also could be calculated to assess the contribution of soil dust and suspended construction materials to urban aerosols (Wang

et al., 2005). The average Ca/Al ratios were compared in spring (1.07), autumn (4.40), and winter (12.82). The higher Ca/Al ratio in winter was due to the lower Al concentrations rather than higher Ca concentrations (Ca concentrations showed only slight difference during the three seasons) as shown in Table 2. The lower Ca/Al ratio could be accounted for the increased windblown dust during spring, which was coincident with the fact that another typical crustal element Si was much higher in spring. The lack of apparent variation in the Ca concentration might reflect the contribution of construction activity to atmospheric particulates, which remained stable during the three seasons. The mass concentrations of the coal-related elements As and Se (Tian et al., 2010) were lowest in winter, indicating that coal combustion for civil heating in this season might not have obvious significant influence on ambient particles compared with the other two seasons. This temporal characteristic was quite different from most other areas in China, especially the northern Chinese cities, which suffer severe air pollution resulting from coal combustion for heating during cold seasons. Pb and Br were the fingerprints for traffic emissions (Cao et al., 2009), and Ni and V could originate from oil combustion (Hueglin et al., 2005). Concentrations of Br, Ni and V varied slightly during the three seasons; however, concentrations of Pb fluctuated sharply, which indicated that Pb had a different source besides traffic emissions.

In order to identify the source and evaluate the extent of anthropogenic influences, enrichment factors (EF) were calculated for the measured elements in each season (Hsu et al., 2010):

$$\mathrm{EF}_X = \frac{(C_X/C_\mathrm{R})_\mathrm{aerosol}}{(C_X/C_\mathrm{R})_\mathrm{crust}}$$

where, C_X is the mass concentration of element X and C_R is concentration of reference element. The subscripts of aerosol and crust refer to particles in the aerosol samples and crustal materials, respectively. Ti was selected as the reference element in this study and crustal compositions were given by Chen et al. (1991). Values of EF below 1 indicated that local crust was the major origin of elements; EF between 1 and 5 implied that these elements were emitted from other pollution sources beside crustal sources, while EF over 5 suggested that anthropogenic emission was predominant (Hsu et al., 2010). Figure 3 shows the values of EF for the studied elements in $PM_{2.5}$ in Yong'an.

Table 2 Elemental concentrations in haze sampling periods (unit: ng/m^3)

	Spring	Autumn	Winter
Mg	511.7 ± 643.7	403.9 ± 290.7	506.2 ± 929.2
Al	1335.7 ± 1501.4	277.4 ± 255.3	104.3 ± 103.3
Si	24586.9 ± 14830.6	2690.5 ± 2144.6	1335.4 ± 894.9
K	4588.9 ± 2808.3	5395.8 ± 3773.4	6072.2 ± 5344.0
Ca	1432.3 ± 1203.0	1219.3 ± 726.2	1337.1 ± 1084.7
Ti	23.1 ± 22.2	17.7 ± 15.8	11.3 ± 7.8
V	10.7 ± 9.6	12.5 ± 4.4	10.5 ± 3.5
Cr	15.2 ± 11.8	30.4 ± 29.7	23.4 ± 12.9
Mn	841.7 ± 494.8	685.5 ± 536.1	96.1 ± 56.3
Fe	736.1 ± 510.3	930.2 ± 444.7	582.1 ± 323.1
Ni	16.8 ± 8.4	17.3 ± 7.2	18.2 ± 8.6
Cu	16.5 ± 9.8	23.4 ± 10.4	19.6 ± 10.6
Zn	1547.4 ± 866.7	878.3 ± 483.7	260.2 ± 141.7
As	152.9 ± 79.8	126.7 ± 104.9	68.0 ± 45.0
Se	44.3 ± 19.8	49.5 ± 46.6	32.2 ± 25.3
Br	47.2 ± 28.6	57.5 ± 38.3	55.1 ± 11.6
Pb	1793.4 ± 1258.2	1231.5 ± 877.8	239.0 ± 175.5

Data are presented as mean ± standard deviation.

From Fig. 3, it could be found that all enrichment factors peaked in autumn, which meant strong enrichment occurred in this season for all the studied elements. For most elements, the enrichment factors between spring and winter were similar. As illustrated in Fig. 3, the EF of Al was below 5 during the three seasons, while EFs of other elements were greater than 5. Taking K as an example, its EF was in the range of 74.5–436.8; biomass burning was considered to be a contributor in addition to crustal sources (Cao et al., 2009). According to the EF values of Zn, As, Cu, and Se, non-crustal sources were the major origins of these elements. Pb, Zn, Br, and Cu have often been regarded as markers associated with traffic sources (Querol et al., 2001; Manoli et al., 2002; Fang et al., 2003; Cao et al., 2009). Therefore, the traffic source probably contributed a great amount of the contents of these elements in $PM_{2.5}$. Furthermore, As and Se are elements known to be derived from coal combustion (Tian et al., 2010). Cu and Zn shared similar sources of fuel burning, industrial metallurgical processes and waste incineration, while Zn was also emitted from traffic sources (Nriagu and Pacyna, 1988; Chueinta et al., 2000). The experimental result of the elements in $PM_{2.5}$ was coincident with the fact that Yong'an is an industrial city, and stationary sources, including power plants and a cement plant, were the major contributors to the city's atmospheric pollution.

2.5 WSIIs compositions

Water soluble inorganic ions (WSIIs) are associated with particle formation, growth, migration and transformation. Previous studies showed that WSIIs accounted for one-third or more of the fine particles in Chinese urban aerosols (He et al., 2001; Hu et al., 2002; Wang et al., 2002). As given in Table 3, WSIIs accounted for (25.3 ± 0.8)%, (52.5 ± 10.8)% and (46.4 ± 9.3)% of $PM_{2.5}$ mass in the three seasons, respectively, suggesting the WSIIs play an

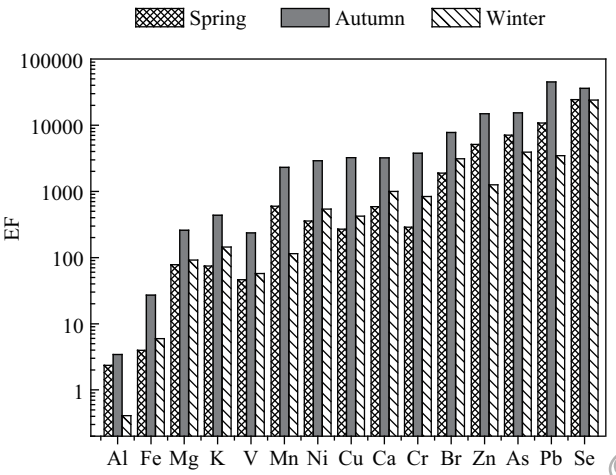

Fig. 3 Enrichment factors (EF) for elements in different seasons.

important role in the compositions of fine particles in Yong'an.

The major WSIIs species were a slightly different in different seasons. SO_4^{2-}, NO_3^- and Cl^- were the three major species in spring, accounting for (73.9 ± 14.4)%, (23.9 ± 4.5)%, and (8.8 ± 5.5)% of the total WSIIs, while SO_4^{2-}, NO_3^- and NH_4^+ were the three major species accounting for (79.0 ± 6.8)%, (6.9 ± 2.9)% and (4.8 ± 4.2)% of the total WSIIs in autumn and (57.6 ± 10.0)%, (14.1 ± 12.7)%, and (11.6 ± 9.2)% of the total WSIIs in winter, respectively. The top three major WSIIs species varied among the three seasons in Yong'an, while the top three major WSIIs species stayed relatively constant in other cities (SO_4^{2-}, NO_3- and NH_4^+ were the three most abundant ions in fine particulates in most areas) (Tan et al., 2009; Chen et al., 2003).

The ratios of the mass concentrations of NO_3^-/SO_4^{2-}, and values of the nitrogen oxidation ratio (NOR) and sulfur oxidation ratio (SOR) are summarized in Table 4. Previous studies demonstrated that the ratio of NO_3^-/SO_4^{2-} is useful to evaluate the contribution of mobile and stationary sources to nitrogen and sulfur in the atmosphere of China (Wang et al., 2005; Cao et al., 2009).

Ratios of NO_3^-/SO_4^{2-} during sampling periods in Yong'an were in the range of (0.09 ± 0.04)–(0.42 ± 0.08), with an average value and SD of 0.15±0.12. Compared with other cities, it was clearly found that the NO_3^-/SO_4^{2-} ratio in Yong'an was evidently lower than that in the downtown of Log Angeles and Rubidoux (2–5) (Kim et al., 2000) in Southern California (where no coal was used) and evidently lower than those in Guangzhou (0.79) (Tan et al., 2009b), Beijing (0.71) (Wang et al., 2005) and Shanghai (0.64) (Wang et al., 2006), all of which had heavy traffic densities. The ratio of NO_3^-/SO_4^{2-} in Yong'an was comparable to those in Hangzhou (0.36), (Cao et al., 2009), Qingdao (0.35) (Hu et al., 2002), Hong Kong (0.26) (Pathak et al., 2003), and Taiwan (0.20) (Fang et al., 2002). The ratio in Yong'an was close to that in the typical coal combustion pollution city, Guiyang (0.14) (Xiao and Liu, 2003), suggesting that coal burning made a major contribution to particulate pollutants in Yong'an. At the same time, the effect of vehicle exhaust should not be neglected against the background of the rapid increase of motor vehicles in the urban area.

Table 3 WSIIs concentrations and NO_3^-/SO_4^{2-}, NOR and SOR in haze periods (unit: μg/m^3)

	Spring	Autumn	Winter
WSIIs			
F^-	ND	1.23 ± 0.22	0.22 ± 0.15
Cl^-	1.55 ± 0.25	1.77 ± 2.55	0.74 ± 0.55
NO_3^-	6.99 ± 0.89	4.09 ± 2.30	2.93 ± 1.79
SO_4^{2-}	12.19 ± 4.55	37.22 ± 15.88	20.38 ± 5.85
NH_4^+	1.37 ± 1.48	3.71 ± 2.48	4.22 ± 4.25
K^+	0.52 ± 0.62	1.44 ± 0.79	2.85 ± 3.06
Na^+	0.96 ± 1.60	0.39 ± 0.23	0.39 ± 0.25
Ca^{2+}	0.20 ± 0.06	0.27 ± 0.22	–[a]
Mg^{2+}	0.31 ± 0.28	0.38 ± 0.17	0.27 ± 0.18
NO_3^-/SO_4^{2-}	0.42 ± 0.08	0.09 ± 0.04	0.12 ± 0.08
SOR	0.15 ± 0.06	0.35 ± 0.11	0.24 ± 0.16
NOR	0.09 ± 0.03	0.08 ± 0.05	0.07 ± 0.08

Data are presented as mean ± standard deviation.
[a] exceptional data which is canceled.

To determine the degrees of atmospheric conversion of SO_2 to SO_4^{2-} and of NO_2 to NO_3^-, the sulfur and nitrogen oxidation ratios (SOR and NOR, respectively) were employed and defined as follows (Ohta and Okita, 1990): $SOR = nSO_4^{2-}/(nSO_4^{2-} + nSO_2)$, $NOR = nNO_3^-/(nNO_3^- + nNO_2)$. SOR and NOR in the atmosphere of urban Yong'an during the sampling periods are shown in Table 4. Higher values of SOR and NOR suggested that photochemical oxidation may occur and more secondary aerosols can exist in the atmosphere (Ohta and Okita, 1990; Kaneyasu et al., 1995). Gas-to-particle conversion could be accomplished by vapor condensation (heterogeneous nucleation), adding mass onto primary aerosol particles, or by direct nucleation from gas phase reaction (homogeneous nucleation) (Foltescu et al., 1996).

In this study, SO_2 had a lower concentration in autumn than the other two seasons, while the SO_4^{2-} concentration and SOR had their highest value in autumn. SOR had a certain degree of correlation with SO_2 as the R^2 were 0.46, 0.38 and 0.75 in the three seasons, respectively. SOR only had a relatively good correlation with $PM_{2.5}$ mass (R^2 = 0.74) in autumn, which indicated that gas phase reaction and heterogeneous nucleation both took place in this season. SOR values could be affected by other factors besides SO_2 concentrations; therefore the meteorological and other conditions in this autumn sampling period might be favorable for the formation of sulfate from gaseous SO_2. For sulfate primary pollutant the value of SOR is lower than 0.10 (Pierson et al., 1979); when the SOR value is higher than 0.10, photochemical oxidation of SO_2 occurs in the atmosphere (Ohta and Okita, 1990). The SOR of $PM_{2.5}$ aerosols in Yong'an were higher than 0.10 in the sampling periods (from 0.15 ± 0.06 to 0.35 ± 0.11), and the result showed that the formation of secondary pollutants from the transformation of SO_2 to SO_4^{2-} occurred throughout the year, especially during haze days. This SOR value was comparable to that in Kaohsiung City (0.30 on haze days and 0.12 for clear days) (Lin, 2002), which was a major SO_2 emission area in Taiwan.

Sulfate material (SO_4^{2-}) accounted for a considerable proportion of $PM_{2.5}$ mass concentrations in high coal-fired electric utility regions, and the visibility reduction was an outcome of rapid SO_4^{2-} accumulation in humid atmosphere (Chen et al., 2003). A similar result was also found in this study, which signified the role of SO_4^{2-} in haze formation.

NOR stayed at a comparatively stable value during the three seasons, in the range of (0.07 ± 0.08) to (0.09 ± 0.03). Values of the NOR were generally lower than those for SOR (Colbeck and Harrison, 1984). The NOR values obtained in this study were also similar to the resulted measured in Kaohsiung, Taiwan, where there was a NOR value of 0.09 on haze days (Lin, 2002). The high NOR values obtained in this study suggested that secondary formation of NO_3^- from NO_2 occurred frequently in the atmosphere during the sampling periods in Yong'an.

Table 4 Concentration and percentage of different compositions in $PM_{2.5}$

		Secondary ions	Total carbon	Crustal elements	Sea salt	Coal/oil burning elements
Spring	Concentration (μg/m^3)	20.55	15.98	33.52	2.82	12.38
	Percentage (%)	24.7	19.2	40.3	3.4	14.9
Autumn	Concentration (μg/m^3)	45.02	17.28	11.38	2.95	11.67
	Percentage (%)	50.1	19.2	12.7	3.3	13.0
Winter	Concentration (μg/m^3)	27.53	24.05	9.53	1.40	7.03
	Percentage (%)	34.8	30.5	12.1	1.8	8.9

2.6 Composition characteristics of $PM_{2.5}$

Compositions of $PM_{2.5}$ are displayed in Table 4. The determinant elements and compounds were classified as six types. The crustal element (Si, K, Ca, Al, Fe, Mn, etc) species was the most abundant component in spring $PM_{2.5}$, accounting for 40.3% of the total $PM_{2.5}$ mass, followed by secondary ions (SO_4^{2-}, NH_4^+, NO_3^-, etc.) and TC, accounting for 24.7% and 19.2%, respectively. $PM_{2.5}$ had different correlation with OC, EC, SO_4^{2-}, Si, K, Ca and Fe, indicating that civil construction activity, dust, traffic emissions and coal combustion can have respective influence on atmospheric particle formation. $PM_{2.5}$ also had a strong correlation with the coal burning indicator element Ti ($R^2 = 0.89$), which showed that coal combustion was another major source of the atmospheric pollution in spring.

Different from spring, secondary ions became the most abundant components in $PM_{2.5}$ in autumn, accounting for 50.1% of the total $PM_{2.5}$ mass, followed by TC and coal/oil burning elements, accounting for 19.2% and 13.0%. $PM_{2.5}$ had strong correlation with SO_4^{2-} and NO_3^- (R^2: 0.62, 0.63, respectively), while the correlations with crustal elements, OC and EC were relatively weak, indicating that coal burning was still a major source of the atmospheric pollution in autumn.

The most abundant component in winter $PM_{2.5}$ was secondary ions, accounting for 34.8% of the total mass of $PM_{2.5}$, followed by TC and crustal elements, accounting for 30.5% and 12.1%. The mass of $PM_{2.5}$ had relatively good correlations with NO_3^-, OC and EC ($R^2 = 0.50$, 0.70 and 0.53, respectively), and weak correlations with crustal elements, indicating that $PM_{2.5}$ in this season was affected mainly by traffic emission and coal combustion.

Elements and chemical species relating to coal combustion had a slightly different variation in different seasons. Accordingly, the effect of coal combustion on $PM_{2.5}$ in the three seasons should be discussed in more detail. Elemental analysis of the typical coal-related elements As and Se showed a lower concentration in winter, while the lowest and highest concentrations of the secondary ion SO_4^{2-} were present in spring and autumn respectively, and the gaseous pollutant SO_2 presented the lowest and highest concentrations in autumn and winter respectively. The inconsistency of the SO_4^{2-} and SO_2 concentration distribution was due to different SOR in different seasons. The inconsistency of coal-related elements and sulfuric pollutants was due to their differences in formation mechanisms, particle size distribution, transportation and retention in the ambient atmosphere. In addition, exogenous SO_2 from other areas to this city should be taken into consideration.

Generally, higher concentrations of crustal constituents was the most prominent characteristic of $PM_{2.5}$ in spring; while the secondary ions and carbonaceous species became the two major contributors in autumn and winter, indicating the significance of combustion and traffic emission contributions.

3 Conclusions

In this study, the chemical compositions in fine particles $PM_{2.5}$ sampled in haze periods during spring, autumn and winter in Yong'an were analyzed, and various possible pollution contributors and meteorological conditions related to haze were discussed.

Although the concentrations of $PM_{2.5}$ sometimes varied during the same sampling period, the average $PM_{2.5}$ concentrations of the three sampling periods were close to each other. Similar variations were also presented in relative humidity and wind speed, such that average values of these meteorological parameters did not show a distinct difference among the three seasons. Low wind speed and high relative humidity in the 30 sampling days were both favorable for haze formation.

OC and EC constituted up to 19.2%–30.4% of $PM_{2.5}$ mass, and TC showed a good correlation with $PM_{2.5}$. Si, K, Pb, Zn, Ca and Al were the major elements in $PM_{2.5}$, and different elements showed different distribution characteristics in the three seasons. WSIIs constituted up to (25.3±0.8)%–(52.5±10.8)% of $PM_{2.5}$ mass. The major ions in $PM_{2.5}$ were SO_4^{2-}, NH_4^+, NO_3^-, etc. The ratios of NO_3^-/SO_4^{2-} were evidently lower than those of cities mentioned in the literature, and close to that in the typical coal combustion pollution city, Guiyang.

$PM_{2.5}$ composition analysis indicated that the crustal element species were the most abundant component in spring, while the secondary ion species were the most abundant component in autumn and winter. Considering the city's development conditions, road construction, soil dust, biomass burning, coal combustion and traffic emissions could all have different significant influences on $PM_{2.5}$ components. It could be deduced that dust was the primary pollution source for $PM_{2.5}$ in spring and combustion and traffic emissions could be the main pollution sources for $PM_{2.5}$ in autumn and winter. As a mountainous industrial city, Yong'an's landform is disadvantageous for the dispersion of atmospheric pollutants, which mainly accounted for the deterioration of air quality and increase of haze days. Although the atmospheric pollution of Yong'an

discussed in this research was based on the analysis results of $PM_{2.5}$ samples collected in three haze periods (with a 10-day sampling length each period), and could not reflect the atmospheric pollution in the entire city completely, this research could enable a better understanding of the main pollution contributors to haze formation in this city.

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