



## Characteristics of indoor/outdoor PM<sub>2.5</sub> and elemental components in generic urban, roadside and industrial plant areas of Guangzhou City, China

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### Abstract

Quantitative information on mass concentrations and other characteristics, such as spatial distribution, seasonal variation, indoor/outdoor (I/O) ratio, correlations and sources, of indoor and outdoor PM<sub>2.5</sub> and elemental components in Guangzhou City were provided. Mass concentration of PM<sub>2.5</sub> and elemental components were determined by standard weight method and proton-induced X-ray emission (PIXE) method. 18 elements were detected, the results showed positive results. Average indoor and outdoor PM<sub>2.5</sub> concentrations in nine sites were in the range of 67.7–74.5 µg/m<sup>3</sup> for summer period, and 109.9–123.7 µg/m<sup>3</sup> for winter period, respectively. The sum of 18 elements average concentrations were 5362.6–5533.4 ng/m<sup>3</sup> for summer period, and 8416.8–8900.6 ng/m<sup>3</sup> for winter period, respectively. Average concentrations of PM<sub>2.5</sub> and element components showed obvious spatial characteristic, that the concentrations in roadside area and in industrial plant area were higher than those in generic urban area. An obvious seasonal variation characteristic was found for PM<sub>2.5</sub> and elemental components, that the concentrations in winter were higher than that in summer. The I/O ratio of PM<sub>2.5</sub> and some elemental components presented larger than 1 sometimes. According to indoor/outdoor correlation of PM<sub>2.5</sub> and element concentrations, it was found that there were often good relationships between indoor and outdoor concentrations. Enrichment factors were calculated to evaluate anthropogenic versus natural elements sources.

**Key words:** PM<sub>2.5</sub>; elemental components; indoor air

### Introduction

Particle matter (PM), especially PM<sub>2.5</sub> (particle with aerodynamic diameter less than 2.5 µm), is the most ubiquitous and most complicated air pollutant in urban area (Pope, 1995; Reichhardt, 1995). There is now a great deal of epidemiological evidence associating exposure to ambient particles with short- and long-term effects on health (Anderson, 2000; Kim and Jaques, 2000). With improvement of measurement techniques, the effects became clearer when smaller size particles were considered (Wichmann and Peters, 2000; Lippmann and Ito, 2000). Besides health effects, atmospheric fine particles play an important role in controlling a number of atmospheric processes such as the deposition of different compounds, the optical properties, etc. (Molnár *et al.*, 1999). From these points of view, it is crucial to know the characteristics of the fine particles to develop effective strategy for the control of fine aerosol pollution.

Elemental components constitute a small portion by mass of the particles, however, the existence of some heavy metals such as lead, arsenic, selenium, and iron, may exhibit detrimental health impact to human body even in trace amount (Patterson and Eatough, 2000; Chao and Wong, 2002). Therefore, in order to understand the properties of fine particle pollution, there is a need for the studies on concerning components of fine particle (Pakkanen *et al.*, 2001).

PM is usually generated from many different outdoor sources, such as automobile exhaust, industrial production processes, secondary conversion from gaseous pollutants (Gartrell and Friedlander, 1975) and also from indoor sources, such as smoking (Øie and Magnus, 1997), cooking (Naeher *et al.*, 2000) and other activities (Chan, 2002). People spend most of their time indoors. Yet the majority data of particle concentration are based on measurements conducted outdoors. Actually, outdoor particle concentrations may not be reliable indicators of indoor and personal particle exposures (Geller *et al.*, 2002). Studies on indoor particles characteristics are as important as the outdoors'.

Guangzhou is a sub-tropical city and has warm weather and high relative humidity (RH). It is the largest city of

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southern China, with population approximately 10 millions and some industrial factories are located in urban and suburban areas of the city (Cao *et al.*, 2003a). The city is seriously crowded with population and traffic vehicles, and the quality of indoor and outdoor air is causing increasing concern. Studies on fine particles in Guangzhou are scarce, and almost no specific research on simultaneous indoor and outdoor PM<sub>2.5</sub> characteristics has been carried out.

The main objectives of this study were to provide quantitative information on mass concentrations and other characteristics, such as spatial distribution, seasonal variation, indoor/outdoor (I/O) ratio correlations and sources, of indoor and outdoor PM<sub>2.5</sub> and elemental components in Guangzhou City.

## 1 Experiment

### 1.1 Sampling sites

Guangzhou City (22°36′–24°18′N, 112°33′–114°35′E) is one of the most developed and industrialized city in South China, with high population density, where the amount of population and motorcar is increasing year by year, excessive air pollutants are released, and atmospheric haze happens more often. Nine pairs sampling sites (indoor and outdoor) classified in 3 kinds of areas (general urban area, roadside area, the area near industrial plant) of Guangzhou City were selected for this study. All indoor environments were located in domestic home. Three pairs sampling sites (GS1, GS2, GS3) were selected in general urban area without impacts of heavy vehicle traffic and industrial production, three pairs sampling sites (RS1, RS2, RS3) were selected by roadside with heavy motor vehicle traffic, and three pairs sampling sites (IS1, IS2, IS3) were selected near industrial plant. Detailed characteristics of nine pairs sampling sites are given in Table 1.

### 1.2 Sample collection

Both indoor and outdoor PM<sub>2.5</sub> samplings were collected simultaneously from 2 July to 13 August 2004 (Summer period) and from 29 November 2004 to 6 January 2005 (Winter period) by using two mini-volume portable samplers (Airmetrics, USA) with a PM<sub>2.5</sub> cyclone

operating with the flow rates of 5 L/min for continuous 24 h. All samples were collected on 47 mm Whatman quartz microfibre filters. The filters were pre-heated before sampling at 800°C for 3 h. After collection, loaded filters were stored in a refrigerator at about 4°C before weighing and chemical analysis. Twenty-four hour sampling lasted 4 d in every site, so 144 pieces filters were collected in 9 sites for indoor and outdoor environment during two sampling season. In addition, 4 pieces blank experience filters were also collected. Blank experience means sampling with electricity power off and other condition same as routine sampling. The samplers were simultaneously put in the living room and in the balcony, or platform, or flat roof, which represent outdoor environment. The indoor sampling height was in the range of 1–1.5 m above ground to simulate the breathing zone and avoid potential interferences from excessive re-suspension of particles.

### 1.3 Measurement and quality control of PM<sub>2.5</sub>

PM<sub>2.5</sub> mass was determined gravimetrically using an electronic microbalance with a 1 µg sensitivity (Mettler M3, Switzerland). Each filters were weighted after being equilibrated for 24 h in a silica gel desiccators and at a constant (within ± 2°C) temperature between 20°C and 23°C. Each filter was weighed at least three times before and after sampling. The average values were used. Mass concentrations of PM<sub>2.5</sub> were obtained by subtracting the initial mass of the blank filter by the final mass of the sampled filter and dividing the difference by the total volume of air passing through the filter. The difference among the three repeated weightings was less than 10 µg for a blank filter and less than 20 µg for a sampled filter. Reweighing of the filter is necessary if the difference between the former and the latter weighting is out of the range.

### 1.4 Analysis and quality control of elemental components

After weighing the sampled filter for PM<sub>2.5</sub> quantification, elemental components in PM<sub>2.5</sub> were analyzed by proton-induced X-ray emission (PIXE) method without prior extraction. PIXE technique is a method for elemental analysis, which relies on the analysis of the energy spectra

**Table 1** Characteristic of indoor/outdoor sampling locations in Guangzhou City

Site no.	Site location	Types	No. of occupants	Cooking	Floor	Size (m <sup>2</sup> )	Smoking	Build year	Decoration year
GS1	SunYatSen University	The site in generic urban area	4	Yes	4	100	No	2000	2000
GS2	Jinan University	The site in generic urban area with light traffic effect	4	Yes	6	100	No	2000	2001
GS3	Nanyayuan Community	The site in generic urban area	2	Yes	5	86	No	1989	1990
RS1	Mingyue er Road	The site by the side of third-class road	3	Yes	6	106	No	1992	1998
RS2	Xinggangxi Road	The site by the side of main road	5	Yes	7	95	Yes	1986	1997
RS3	Tianshou Road	The site by the side of sub-main road	5	Yes	6	70	No	2000	2000
IS1	Huangpu xingwei xingchun	The site near a ethylene plant	2	Yes	1	220	No	2000	2000
IS2	Huangpu shachong	The site near two power plant	3	Yes	1	150	No	1997	1998
IS3	Huangpu shihua yard	The site near several chemical plants	2	Yes	6	60	No	1990	1993

of characteristic X-rays emitted from a sample, bombarded with a flux of high energy protons (about 1–3 MeV). Due to its low background noise, a high sensitivity for trace element analysis can be achieved. In this study, element PIXE analysis was performed by the Institute of Low Energy Nuclear Physics, Beijing Normal University, using 2.5 MeV protons with a 50 nA beam current from a 1.7×2 MV accelerator. The PIXE system was calibrated using standards produced by MicroMatter Co. Blank filter background spectra were subtracted prior to peak integration. Analysis of eight samples of standard reference material from the National Bureau of Chemical Exploration Analysis, China, showed that satisfactory precision (<10%) and accuracy (<15%) were achieved. Eighteen elements detected and showing positive results in our study were sulfur, chlorine, potassium, calcium, titanium, vanadium, chromium, manganese, iron, nickel, copper, zinc, arsenic, bromine, strontium, zirconium, and lead.

### 1.5 Meteorological factor

During summer sampling period, the mode wind direction was pre-dominantly south. The wind speed 10 m above the ground level usually fell into the range 0.8–3.5 m/s. The temperature during diurnal period ranged from 25 to 37°C and ambient pressure varied insignificantly between 998.3 and 1007.2 hPa. The relative humidity (RH) was between 50%–90%. During winter sampling period, the mode wind direction was dominantly northeaster and north. The wind speed 10 m above the ground level was in the range of 0.0–5.3 m/s. Daily average temperature ranged from 6.5 to 20.6°C and ambient pressure varied insignificantly between 1010.0 and 1025.6 hPa. Average relative humidity was between 30%–78%.

### 1.6 Data processing and statistical analysis

One hundred and forty-three pieces loaded filters and 4 pieces blank experience filters were weighted except one loaded filter invalid. The mass concentration of PM<sub>2.5</sub> in sampling state was transformed to corresponding value in standard atmospheric state. At every site, arithmetic daily average value of PM<sub>2.5</sub> mass concentration was obtained based on four 24 h average concentration values. Statistical analysis was performed using SPSS 11.5 (statistical program for social science) statistical software packages and Microsoft Excel 2000 program.

## 2 Results and discussion

### 2.1 Mass concentration of PM<sub>2.5</sub>

Average mass concentrations of indoor and outdoor PM<sub>2.5</sub> of total samples in this study were given in Table 2. As shown in Table 2, average indoor and outdoor PM<sub>2.5</sub> concentrations in the nine sites were 67.7 and 74.5 μg/m<sup>3</sup> for summer period and 109.9 and 123.7 μg/m<sup>3</sup> for winter period, respectively.

USA National Ambient Air Quality Standard (NAAQS, 1997) requires PM<sub>2.5</sub> 24-h average concentration in populated area to be less than 65 μg/m<sup>3</sup>. Compared with

**Table 2 Average concentrations of PM<sub>2.5</sub> in Guangzhou (μg/m<sup>3</sup>)**

Sites	Summer		Winter	
	Indoor	Outdoor	Indoor	Outdoor
GS1	58.0±7.1	52.4±15.8	89.1±25.2	120.6±46.1
GS2	67.1±21.3	56.5±14.1	95.8±29.8	175.0±39.4
GS3	43.6±17.2	44.7±16.7	64.0±28.9	64.0±30.6
Average	56.2±17.1	51.2±15.0	83.0±29.1	119.9±59.1
RS1	62.2±22.5	69.5±11.6	92.9±19.4	134.8±23.2
RS2	71.1±5.2	88.6±8.5	212.4±48.3	216.0±48.1
RS3	87.1±32.2	80.1±31.4	101.6±21.4	72.2±16.7
Average	73.5±23.3	79.4±19.8	135.6±64.0	141.0±68.1
IS1	65.7±15.4	66.0±19.0	85.1±4.2	83.5±8.5
IS2	54.6±25.0	111.2±18.9	121.0±31.7	117.0±37.6
IS3	99.8±13.8	101.5±22.0	126.8±34.1	130.1±33.2
Average	73.4±26.3	92.9±27.2	111.0±31.1	110.2±33.6
Average in all	67.7±23.6	74.5±27.1	109.9±48.3	123.7±55.5

the limit value, out-standard state of each daily PM<sub>2.5</sub> concentration in this study of Guangzhou City was very severe, that out-standard percents of indoor and outdoor PM<sub>2.5</sub> were 57.1% and 55.6% in summer, and 88.9% and 86.1% in winter, respectively. Therefore, more efforts should be taken for the control of fine particle pollution in Guangzhou. Currently there is no PM<sub>2.5</sub> standard in China; so more efforts also should be taken towards the establishment of a PM<sub>2.5</sub> concentration standard of China.

Average indoor and outdoor PM<sub>2.5</sub> mass concentrations in Guangzhou were compared with the results of other studies on PM<sub>2.5</sub> in other cities (Table 3). As shown in Table 3, a significant difference was observed between the value in Guangzhou and that in some US and Europe cities. The concentrations of PM<sub>2.5</sub> in Guangzhou were 2.2–6.1 and 6.3–7.2 times more than that of US cities, and 2.1–8.6 and 3.1–5.4 times more than that of Europe cities for indoor and outdoor environment, respectively. It implied that both indoor and outdoor PM<sub>2.5</sub> qualities in Guangzhou were worse. Compared with research results in several Asia cities, the concentrations of PM<sub>2.5</sub> in Guangzhou were 4.5 times and 3.8 times higher than that of Osaka, Japan, and were 1.4 and 1.3–1.6 times higher than that of Hong Kong for indoor and outdoor environment, respectively. In addition, outdoor concentrations of PM<sub>2.5</sub> were 0.3–1.0 times higher than that of Shanghai, and 1.0 times higher than that of Shenzhen and Zhuhai. It indicated that indoor and outdoor PM<sub>2.5</sub> concentrations in Guangzhou were much severe by comparing to some Asia even China cities. It was doubted that the high PM<sub>2.5</sub> concentrations in Guangzhou were due to the huge population and motor vehicles and industrial production.

### 2.2 Mass concentrations of elemental components in PM<sub>2.5</sub>

#### 2.2.1 General characteristics

Average concentrations of indoor and outdoor elemental components in PM<sub>2.5</sub> by overall samples in Guangzhou City are shown in Table 4. During summer, the sum of average concentrations of 18 elements were 5362.6 and 5533.4 ng/m<sup>3</sup> for indoor and outdoor environment, respectively. During winter, the sum of average concentrations of

**Table 3** PM<sub>2.5</sub> concentrations in Guangzhou and comparison with other cities (μg/m<sup>3</sup>)

Sites	Summer			Winter		
	Indoor	Outdoor	Sampling period	Indoor	Outdoor	Sampling period
Guangzhou, China (this study)	67.7	74.5	2 July to 13 August 2004	109.9	123.7	29 November 2004 to 6 January 2005
Shanghai, China	-	36.3	Summer of 1999	-	91.2	Winter of 1999
Guangzhou, China	-	-		-	105.9	January–February 2002
Shenzhen, China	-	-		-	60.8	January–February 2002
Zhuhai, China	-	-		-	59.3	January–February 2002
Hong Kong, China	-	-		-	54.5	January–February 2002
Hong Kong, China	-	-		45.0	47.0	October 1999 to March 2000
Hong Kong, China	-	-		-	50.2	November 2000 to February 2001
Osaka, Japan	-	-		20.0	26.0	November 1994, October 1995, December 1997
Suffolk, New York, USA	-	-		33.8	16.9	January–April 1986
Minneapolis-St. Paul metropolitan area, USA	13.9	10.3	Spring, summer and fall of 1999	-	-	
Coachella, California, USA	-	-		15.5	15.0	Winter and Summer of 2000
Baltimore, USA	-	-		15.7	17.0	January–February 1997
London, UK	14.0	18.0	April–October 1998	-	-	
Oslo, Norway	22.0	13.0	1992–1993	-	-	
Amsterdam, Netherlands	-	-		19.5	23.0	January–April 1995
Athens, Greece	-	-		11.4	-	January–June 2000
Brisbane, Australia	9.9	11.6	March–August 1999	-	-	

The values in the table were average of each study.

indoor and outdoor 18 elements were 8416.8 and 8900.6 ng/m<sup>3</sup>, respectively. The average percentages of indoor and outdoor 18 elements in PM<sub>2.5</sub> were 8.03% and 7.42% by mass for summer period, and 7.66% and 7.20% for winter period, respectively.

In comparison with the findings from literature (Table 4), average concentrations of single element except S in PM<sub>2.5</sub> in Guangzhou city were higher than the corresponding average element concentration in Birmingham (Hidy *et al.*, 2000), Atlanta (Cao *et al.*, 2003a), Helsinki (Pakkanen *et al.*, 2001), Brisbane (Chan *et al.*, 1999), Sihwa (Park *et al.*, 2001) and Hong Kong (Chao and Wong, 2002). It indicated that both indoor and outdoor fine particle pollution in Guangzhou were much severer than these cities. The high loadings of elemental component in Guangzhou might be due to: firstly, there were near 10 million populations and more than 1 million motor vehicles in the city; secondly, some industrial emission sources were located in the city; thirdly, coal-combustion was used occasionally by residents, and fourthly weather condition during summer, such as high temperature, high humidity and low wind speed were disadvantaged to the pollutants diffusion, but were advantaged to the formation of secondary aerosol. It implied that fine particle pollution prevention and control in Guangzhou is imperative.

### 2.2.2 Characteristic of sulfur

Among 18 involved elements, S element was found to be the dominating elemental component. It implied that the sulfate emissions, including secondary aerosol production from the oxidation of SO<sub>2</sub> and industrial production involved in coal combustion or oil combustion, contributed a significant part to the fine particle, in that traditionally sulfur has been a tracer for secondary sulfate particles (Artaxo, 1999; Park *et al.*, 2001), coal combustion (Ramadan *et al.*, 2000; Song *et al.*, 2001), and oil combustion (Fung and Wong, 1995).

### 2.2.3 Characteristic of lead and zinc

Indoor/Outdoor Pb and Zn concentrations in Guangzhou were higher than that in Birmingham, Atlanta, Helsinki, Brisbane, Sihwa and Hong Kong. It suggested the contribution from motor vehicle exhaust and incineration sources accounted for a larger proportion than these 5 cities did, since Pb and Zn are commonly considered as the marker of motor vehicle emission source and incineration (Baek *et al.*, 1997; Artaxo *et al.*, 1999; Song *et al.*, 2001).

### 2.2.4 Characteristic of iron

In our study, average indoor/outdoor Fe element concentrations were 0.382 and 0.396 μg/m<sup>3</sup>. Fe is a representative trace element of fugitive soil dust (Fung and Wong, 1995; Biegalski and Landsberger, 1998; Lee *et al.*, 1999; Chueinta *et al.*, 2000). The contribution of fugitive soil dust to the PM<sub>2.5</sub> can be evaluated by the ratio of Fe concentration (μg/m<sup>3</sup>) to 3.5%. Based on the above experiential theory, the contribution of fugitive soil dust to the PM<sub>2.5</sub> in Guangzhou were 10.9% and 11.3% for indoor and outdoor environment during summer, and were 13.1% and 15.0% during winter, respectively. The contribution is comparable or lower than those measured at Helsinki, Finland (12%, 1996–1997) (Vallius *et al.*, 2003), Chianjen site, Kaohsiung, Taiwan (17%) (Chen and Lin, 2001), Hong Kong (17.7%) (Fung and Wong, 1995), Taegu City, Korea (29.2%) (Baek *et al.*, 1997), Pohang City, Korea (25.0%) (Baek *et al.*, 1997). It indicated that in Guangzhou city, the total contribution from traffic related source, fuel oil combustion, coal related source, secondary aerosols, and so on, accounted for larger proportion than those places did.

### 2.3 Spatial distribution of PM<sub>2.5</sub> and elemental components

In Table 2, it is found that during summer period, indoor average PM<sub>2.5</sub> concentrations in generic urban area,

**Table 4 Element concentrations in PM<sub>2.5</sub> of Guangzhou and the comparison with other cities (ng/m<sup>3</sup>)**

	Site	S	Cl	K	Ca	Ti	V	Cr	Mn	Fe
Indoor	Guangzhou (summer)	2445.3	302.8	670.3	771.1	41.3	38.3	10.9	24.6	371.4
	Guangzhou (winter)	3266.7	782.6	1640.4	1146.8	49.2	28.2	4.5	41.7	457.9
	Guangzhou (Ave.)	2856.0	542.7	1155.4	959.0	45.3	33.3	7.7	33.2	414.7
	Birmingham	-	-	-	19	10	15	0.1	1	54
Outdoor	Hong Kong	3879.2	301.9	1026	138.2	7.7	4.6	-	10.7	140.1
	Guangzhou (summer)	2475.7	243.4	726.6	861	45.8	39.2	14.1	24.8	396.4
	Guangzhou (winter)	3316.2	940.1	1685	1267.8	55.7	28.5	4.7	42.4	523.8
	Guangzhou (Ave.)	2896.0	591.8	1205.8	1064.4	50.8	33.9	9.4	33.6	460.1
	Birmingham	-	-	-	32	6	3	0.1	2	62
	Atlanta	-	-	143.3	101.3	8.9	0.6	2.7	6.9	37.7
	Hong Kong	4830.3	235.8	1258.8	172.3	12.5	4.9	-	16.7	211.6
	Hong Kong	-	-	967	620	5.9	4.2	3.7	13.0	307
	Helsinki	-	-	85	71	0.8	5	-	3.3	96
	Brisbane	3100	2000	56	30	6	-	-	4	50
	Sihwa	2068	-	369	119	3	2	3	71	192
	Site	Ni	Cu	Zn	As	Se	Br	Sr	Zr	Pb
Indoor	Guangzhou (summer)	22.7	15.5	357.2	26.4	10	17.1	11.5	117.6	108.6
	Guangzhou (winter)	16.6	57.3	458.5	17.2	7.1	67.1	18.4	95.1	261.5
	Guangzhou (Ave.)	19.7	36.4	407.9	21.8	8.6	42.1	15.0	106.4	185.1
	Birmingham	0.8	3	8	5	2	2	1	1.6	70
Outdoor	Hong Kong	1.8	10.7	130.6	-	-	7.8	-	-	88.1
	Guangzhou (summer)	22.8	13.4	367.3	26.6	9.9	16.8	17.3	114.9	117.4
	Guangzhou (winter)	17.4	58.4	473.5	22.2	8.1	70.8	22.4	97.2	266.4
	Guangzhou (Ave.)	20.1	35.9	420.4	24.4	9.0	43.8	19.9	106.1	191.9
	Birmingham	0.8	9	12	5	1	3	1	1.6	55
	Atlanta	3.6	-	14.3	-	11.5	-	-	-	67.3
	Hong Kong	2.1	13	144.9	-	-	8.5	-	-	103.6
	Hong Kong	5.9	21.3	263	3.9	-	-	-	-	76.2
	Helsinki	2	3.1	14	0.8	-	-	0.5	-	5.8
	Brisbane	-	-	27	-	-	18	-	-	51
	Sihwa	7	54	239	7	1	-	-	-	131

roadside area and industrial plant area were 56.2, 73.5 and 73.4  $\mu\text{g}/\text{m}^3$ , respectively, and the corresponding outdoor average PM<sub>2.5</sub> concentrations were 51.2, 79.4 and 92.9  $\mu\text{g}/\text{m}^3$ , respectively. During winter period, indoor average PM<sub>2.5</sub> concentrations were 83.0, 135.6 and 111.0  $\mu\text{g}/\text{m}^3$ , respectively, and the corresponding outdoor average PM<sub>2.5</sub> concentrations were 119.9, 141.0 and 110.2  $\mu\text{g}/\text{m}^3$ , respectively. Obviously, both indoor and outdoor average PM<sub>2.5</sub> concentrations in roadside area and in industrial plant area were higher than those in generic urban area except the situation of winter outdoor. It indicated that traffic exhaust and industrial exhaust were the major outdoor source for concentrations of PM<sub>2.5</sub>, which lead to the high loadings of PM<sub>2.5</sub> concentrations in the residences by roadside and near industrial plant.

By the comparison of element concentrations in generic urban area, roadside area and the area near industrial plant, spatial distribution characteristics of most of the single element concentrations for 3 different environments were also found, that the element concentrations in generic urban area were the lowest, the concentrations by roadside were in the middle, and the concentration near industrial plant were the highest, with some elements concentrations higher by roadside. It indicated that the residential near industrial plant and by roadside need concern more about how to avoid element pollution in PM<sub>2.5</sub>.

Concerning the effects of traffic conditions on PM<sub>2.5</sub>, two categories were classified to represent the different degrees of traffic effects on PM<sub>2.5</sub> and elemental compo-

nents. One category was the generic urban area sites, far away from main road; the other was the sites by roadside. As shown in Table 2, average indoor and outdoor PM<sub>2.5</sub> concentrations of the roadside sites were higher than that of the sites far away from the traffic by 30%–60% and 18%–60%. Further, there was a significant difference between two category sites for Zn, Br and Pb concentrations. The 3 elements are traditional marker of traffic source. During summer, average Zn, Br and Pb concentrations of the roadside sites were higher than that of the sites far away from the traffic by 114.9%, 96.1% and 49.4% for indoor environment, and by 86.2%, 65.7% and 84.8% for outdoor environment, respectively. During winter, average Zn, Br and Pb concentrations of the roadside sites were higher than that of the sites far away from the traffic by 8.0%, 3.7% and 10.9% for indoor environment, and by 35.7%, 19.1% and 30.8% for outdoor environment, respectively. Obviously, high PM<sub>2.5</sub> and Zn, Br and Pb concentrations were observed to be in the sites close to heavy traffic. It is consistent with our assumption that high traffic sources, vehicle exhaust and related wind blowing dust impacted a great deal effect on PM<sub>2.5</sub> and associated elements concentrations.

Comparing and analyzing the two data sets at generic urban sites and the sites near industrial plant, the impact of industrial production on PM<sub>2.5</sub> and elemental component can be found. Similarly, it was observed there were significant differences of PM<sub>2.5</sub> and elemental component concentrations between the sites near industrial plant and

the sites far away from industry. The ratios of average concentration at the sites near industrial plant to that correspondingly at generic sites (I/G) for PM<sub>2.5</sub> and elemental component were commonly larger than 1. Indoor and outdoor I/G ratios of S, As, Se and Pb element were 1.79 and 1.76, 1.34 and 1.26, 1.20 and 1.05, and 1.74 and 2.14 for summer period, and 1.13 and 1.18, 1.08 and 1.24, 1.17 and 1.28, and 1.11 and 1.31 for winter period, respectively. According to the reports previously, high S loading is always taken as tracer of oil combustion (Fung and Wong, 1995; Baek *et al.*, 1997; Chueinta *et al.*, 2000; Kavours *et al.*, 2001; Park *et al.*, 2001; Song *et al.*, 2001; Vallius *et al.*, 2003), high As and Se loadings are markers of coal combustion (Fung and Wong, 1995; Baek *et al.*, 1997), and high Pb loading also suggests industrial sources (Vallius *et al.*, 2003; Ramadan *et al.*, 2000). Since the sites were near industrial plant, it was implied that the oil and coal combustion were from local industrial production.

## 2.4 Seasonal variation of PM<sub>2.5</sub> and elemental components

Comparing the concentration of PM<sub>2.5</sub> (Table 2) and element components (Table 4) in summer with that of the same site in winter, an obvious seasonal variation

characteristic was found that the concentration in winter was higher than that in summer. Element concentrations showed similar seasonal variation with that of PM<sub>2.5</sub>, which is accorded with that the percentage of element to PM<sub>2.5</sub> is generally relatively stable. It was believed that main reasons for seasonal variation might ascribe to that: (1) under the effect of Asian monsoon, there were stronger air convection activities in summer than in winter in Guangzhou, therefore precipitation was more frequent, which is helpful for diffusion and dilution of PM<sub>2.5</sub>; (2) the prevailing wind direction in summer was from south, which bring more clearer air from equator Pacific Ocean and South China Ocean to Guangzhou, while the prevailing wind direction in winter was from north, which bring more polluted air from inland of China; (3) during winter, atmosphere condition in Guangzhou was more stable than that in summer, so it is more difficult for pollutants to be diffused.

But there was an exception for V, Cr, Ni, As, Se, Zr, that the concentrations of them were higher in summer than that in winter. Traditionally, these elements are associated with combustion sources, such as industrial emission and oil and coal combustion, especially As and Se, which are direct index of coal combustion. It was suspected the

Table 5 Pearson correlation coefficient (*r*), significant value (*P*) and I/O ratio between the indoor and the outdoor

		Generic area			Roadside area			Industrial area		
		<i>r</i>	<i>P</i>	I/O ratio	<i>r</i>	<i>P</i>	I/O ratio	<i>r</i>	<i>P</i>	I/O ratio
Summer	PM <sub>2.5</sub>	0.8383	0.001	1.098	0.7342	0.007	0.926	0.4182	0.088	0.790
	S	0.960	0.000	0.9699	0.938	0.000	1.0031	0.919	0.000	0.9865
	Cl	0.462	0.153	1.0891	0.874	0.000	1.2543	0.961	0.000	1.2902
	K	0.934	0.000	0.8754	0.918	0.000	0.9600	0.953	0.000	0.9178
	Ca	-0.065	0.850	0.9878	0.239	0.454	0.8325	0.291	0.358	0.8834
	Ti	-0.597	0.052	0.8398	0.518	0.084	0.9706	0.445	0.147	0.8930
	V	0.902	0.000	0.9628	0.929	0.000	0.9800	0.618	0.032	0.9876
	Cr	0.345	0.299	0.5018	0.340	0.279	1.5680	-0.491	0.105	0.7664
	Mn	0.405	0.216	0.9701	0.881	0.000	0.9830	0.833	0.001	1.0194
	Fe	0.533	0.091	0.7710	0.697	0.012	1.0824	0.897	0.000	0.9437
	Ni	0.623	0.040	1.0481	0.295	0.353	1.0394	0.601	0.039	0.8919
	Cu	-0.629	0.038	0.4974	0.440	0.153	1.1095	0.528	0.078	2.0497
	Zn	0.981	0.000	0.8935	0.958	0.000	1.0195	0.893	0.000	0.9672
	As	0.540	0.087	0.7754	0.304	0.336	1.3326	0.498	0.099	0.8308
	Se	0.551	0.079	0.8548	0.014	0.966	1.3008	-0.147	0.649	0.9823
	Br	0.341	0.304	1.0292	0.454	0.138	1.2144	0.502	0.097	0.8752
	Sr	0.024	0.944	0.7233	-0.194	0.546	0.6892	-0.476	0.117	0.6024
	Zr	0.340	0.306	0.8958	0.444	0.148	1.1294	0.189	0.556	1.0682
	Pb	0.656	0.028	1.0900	0.794	0.002	0.8806	0.655	0.021	0.8850
Winter	PM <sub>2.5</sub>	0.8571	0.000	0.692	0.8729	0.000	0.962	0.9784	0.000	1.007
	S	0.946	0.000	1.0279	0.964	0.000	0.9546	0.987	0.000	0.9786
	Cl	0.907	0.000	0.8791	0.872	0.000	0.8562	0.995	0.000	0.7854
	K	0.928	0.000	1.0443	0.863	0.000	0.9088	0.998	0.000	0.9785
	Ca	0.203	0.526	0.8533	0.749	0.005	0.9327	0.813	0.001	0.9375
	Ti	-0.107	0.741	0.8237	0.854	0.000	0.9400	0.852	0.000	0.9000
	V	0.832	0.001	0.9820	0.975	0.000	0.9331	0.933	0.000	1.0726
	Cr	0.047	0.886	0.8627	-0.015	0.964	1.0000	0.476	0.118	1.0698
	Mn	0.854	0.000	1.0424	0.892	0.000	0.9061	0.915	0.000	1.0085
	Fe	0.590	0.044	0.9183	0.789	0.002	0.8112	0.978	0.000	0.8858
	Ni	0.587	0.045	0.8624	0.829	0.001	0.9205	0.584	0.046	1.0949
	Cu	0.691	0.013	1.1080	0.863	0.000	0.7269	0.998	0.000	1.0554
	Zn	0.918	0.000	1.0879	0.865	0.000	0.8658	0.993	0.000	0.9845
	As	-0.135	0.675	0.8659	0.868	0.000	0.7450	0.211	0.510	0.7500
	Se	0.385	0.216	0.9028	0.007	0.982	0.9103	-0.049	0.879	0.8261
	Br	0.875	0.000	1.0264	0.666	0.018	0.8945	0.866	0.000	0.9325
	Sr	0.140	0.665	0.7600	0.153	0.636	0.8245	0.195	0.544	0.8809
	Zr	0.246	0.440	0.9683	0.169	0.600	0.9506	-0.151	0.639	1.0086
	Pb	0.922	0.000	1.0392	0.708	0.010	0.8813	0.917	0.000	1.0319

reason for the exception was the emergency of electric providing during summer of 2004 in Guangzhou, when almost all of the electric power plants produced by using coal or oil produced in their maximum.

### 2.5 I/O ratio, indoor/outdoor correlation and source implication

Ratio of indoor concentration to outdoor concentration (I/O ratio) is an indicator for evaluating the difference between indoor concentrations and the corresponding outdoor levels (Li and Lin, 2003). The I/O ratios of PM<sub>2.5</sub> and elemental components are shown in Table 5. It was shown that average I/O ratios of PM<sub>2.5</sub> in generic urban area, in roadside area and in industrial plant area were 1.098, 0.926 and 0.790 for summer period, and 0.692, 0.962, 1.007 for winter period, respectively. Similarly, the I/O ratios of some elemental components presented larger than 1. It indicated that the indoor PM<sub>2.5</sub> quality are not always better than outdoors' as people thought, and implied that there might be major sources of PM<sub>2.5</sub> indoors, such as smoking, cooking and cleaning solvent in indoors. Therefore indoor air quality should be concerned urgently and thoroughly.

The correlation between indoor and outdoor paired samples for PM<sub>2.5</sub> and elemental concentrations were tested using Pearson paired *t*-test and the results were also shown in Table 5. The indoor/outdoor correlation was indicated by the Pearson correlation coefficients. The significant-value for the hypothesis tests of the correlation was used to judge whether the confidence level was strong or weak.

In Table 5, it is found that there were good relationships between indoor and outdoor PM<sub>2.5</sub> concentrations except the situation in industrial area during summer, indicating that indoor PM<sub>2.5</sub> concentrations were impacted mainly by the penetration of outdoor PM<sub>2.5</sub> concentrations. It was suspected that one important reason for the exception was very weak ventilation between outdoor and indoor.

During summer, in 3 kinds of area environments, S, K, V, Zn and Pb element had a good correlation between indoor and outdoor concentrations. Nevertheless for Ca, Ti, Cr, Cu, As, Se, Br, Sr and Zr, no matter in which areas, there was no correlation between indoor and outdoor concentrations. During winter, S, Cl, K, V, Mn, Fe, Ni, Cu, Zn, Br and Pb element had a good correlation between indoor and outdoor concentrations, while the things of Cr, Se, Br, Sr and Zr were reverse. Theoretically, good correlation indicates that the elements have their indoor concentrations impacted mainly by the infiltration of corresponding elements of outdoor air, while weak correlation implies much higher removal rate of corresponding elements during the penetration of outdoor air, or closing of windows/doors to prevent the outdoor pollution, or existence of indoor sources of these elements, the 3 factors often accompanied by I/O<1, I/O=1, and I/O>1, respectively. Combining the correlation extent with the corresponding I/O ratio (Table 5), it appeared that when I/O>1 or much lower than 1, the correlation between indoor and outdoor concentrations were more likely not so good.

There was a very strong correlation between sulfur's

indoor and outdoor concentrations. S mostly exists as sulfate fine particles from the oxidation of sulfur dioxide or fuel combustion. The I/O ratios of sulfur were 0.9699, 1.0031 and 0.9865 in generic area, roadside area and the area near industrial plant for summer period, and 1.0279, 0.9546, 0.9786 for winter period, respectively. Somewhat lower indoor levels than outdoors were due to much particle removal during penetration to indoors, a little higher indoor level than outdoors in industrial environment were due to indoor activities, such as cooking, smoking and incense.

Generally, Zn and Pb are taken as the marker of motor vehicle emission sources, and are classic marker of atmospheric outdoor aerosols with no known indoor sources, which explain the very high correlation between their indoor and outdoor concentrations.

Similarly, the results shown in Table 5 indicated that the indoor and outdoor concentrations of K concentrations were highly correlated. A possible explanation for this might be K is a main soil constituent and a marker for burning organic matter (Geller *et al.*, 2002).

Good correlations between indoor and outdoor concentrations obtained for V and Ni indicated that the two metals also more likely originated from outdoor sources, with approximately equal I/O ratio of 0.9628, 0.9800, 0.9876 for V in generic urban area, in roadside area and in the area near industrial plant, and 1.0481, 1.0394, 0.8919 for Ni in 3 environments, respectively.

Weak indoor/outdoor correlation were found for As and Se in the 3 kinds of areas, which suggested that besides outdoor coal combustion source, it was possible that indoor sources might be responsible for a fraction of the observed indoor As and Se levels.

### 2.6 Enrichment factor analysis

Enrichment factor analysis provides a preliminary picture of whether the indoor particle matters come from the crust or from industrial processes or some indoor activities. This method was widely used in particle source apportionment studies (Winchester, 1981; Davidson *et al.*, 1986; Chao and Wong, 2002; Cao, 2003b). Enrichment factor (EF) of average element concentrations of the indoor and outdoor particle relative to the elements of the Earth's crust were used for identifying the major particle contributors.

In our study the EF was calculated by using the equation as follows:

$$EF_{(\text{Fe, reference})} = \frac{(\text{Element/Fe})_{\text{PM}_{2.5}}}{(\text{Element/Fe})_{\text{Earth crust}}} \quad (1)$$

Where the  $(\text{Element/Fe})_{\text{Earth crust}}$  was cited from Winchester (1981).

EF of various elements is shown in Fig.1. Measured EF varied widely in a range of 1.3–26991.6 and the EF of most of the indoor PM<sub>2.5</sub> for each element was higher than those of the outdoor. Element EF includes some degree of uncertainty related to the natural variations of the earth crust component. For this reason it is usually assumed that the EF should be more than an order of magnitude higher than unity to suggest an anthropogenic

origin (Bilos *et al.*, 2001). Elements were categorized as “non-enriched”, “moderately enriched”, or “enriched” if their average enrichment values were less than 10, between 10 and 200, or above 200, respectively. The classification was chosen by the authors after observing breaks in the enrichment factor values.

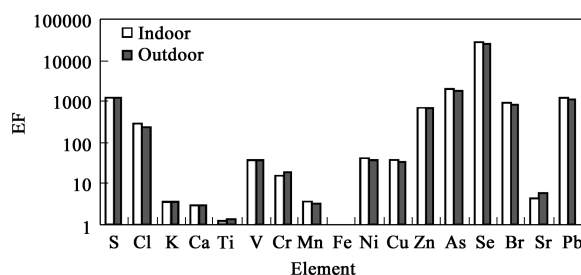


Fig. 1 Enrichment factors (EFs) of elements.

EFs of Se, As, S, Pb, Br, Zn and Cl, were larger than 200, that of Sr, K, Mn, Ca and Ti were smaller than 10. Elements with moderate EF ranging from 15.2 to 41.2 were Ni, V, Cu and Cr. It indicated that Se, As, S, Pb, Br, Zn and Cl were highly enriched and Ni, V, Cu and Cr were secondly enriched, these elements were mainly influenced by anthropogenic sources. Pb, Br and Zn are traditional tracers of vehicle emissions (Fung and Wong, 1995; Chao and Wong, 2002) and their high EF values reflected a large contribution of vehicle exhaust emission. The high EF values of Se, As, Cl, Ni, and Cu indicated that industrial operation (Chao and Wong, 2002), especially the coal-fired plants for Se and As (Fung and Wong, 1995) were a major contributor. High EF values of S and V could be explained by oil combustion sources (Fung and Wong, 1995). With low EF value, Sr, K, Mn, Ca and Ti mainly originated from natural source, soil dust.

### 3 Conclusions

This study fills research blank associated with simultaneous indoor and outdoor PM<sub>2.5</sub> and elemental components in China. Average indoor and outdoor PM<sub>2.5</sub> concentrations in Guangzhou were 67.7 and 74.5  $\mu\text{g}/\text{m}^3$  for summer period, and 109.9 and 123.7  $\mu\text{g}/\text{m}^3$  for winter period, respectively, out the standard of NAAQS. The average concentrations of almost all of single element components in PM<sub>2.5</sub> in Guangzhou were higher than that in Birmingham, Atlanta, Helsinki, Brisbane, Sihwa and Hong Kong. High loadings of PM<sub>2.5</sub> and elements mainly ascribe to huge population, motor vehicles and industrial production. Average concentrations of PM<sub>2.5</sub> and elemental components showed spatial distribution characteristic, the concentrations in roadside area and in industrial plant area were higher than that in generic urban area, indicating traffic exhaust and industrial exhaust were the major outdoor sources. Obvious seasonal variation was found for PM<sub>2.5</sub> and elemental components, the concentrations in winter were higher than that in summer. I/O ratio presented larger than 1 sometimes, indicating indoor PM<sub>2.5</sub> quality are not always better than the outdoors’.

The results of indoor/outdoor concentration correlation indicated indoor concentrations of PM<sub>2.5</sub> and elements were impacted strongly by the penetration of outdoor concentrations. Through Enrichment factors (EF) analysis, it was found that Se, As, S, Pb, Br, Zn, Cl, Ni, V, Cu and Cr were enriched, indicating anthropogenic sources. High EF of Pb, Br and Zn reflected a large contribution of vehicle emission. High EF of Se, As, Cl, Ni, and Cu indicated high contribution of industrial production. High EF of S and V could be explained by oil combustion.

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