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Urban roadside aromatic hydrocarbons in three cities of the Pearl River Delta, People's Republic of China

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Abstract

Urban roadside levels of benzene, toluene, ethylbenzene and xylenes (BTEX) were investigated in three typical cities (Guangzhou, Macau and Nanhai) in the Pearl River Delta Region of south China. Air samples were collected at typical ground level microenvironments by multi-bed adsorbent tubes. The BTEX concentrations were determined by thermal desorption–gas chromatography–mass selective detector (TD–GC–MSD) technique. The mean concentrations of benzene, toluene, ethylbenzene and xylenes were, respectively, 51.5, 77.3, 17.8 and 81.6 $\mu g/m^3$ in Guangzhou, 34.9, 85.9, 24.1, 95.6 $\mu g/m^3$ in Macau, and 20.0, 39.1, 3.0 and 14.2 $\mu g/m^3$ in Nanhai. The relative concentration distribution pattern and mutual correlation analysis indicated that in Macau BTEX were predominantly traffic-related while in Guangzhou benzene had sources other than vehicle emission. In Nanhai, both benzene and toluene had different sources other than vehicle emission. The samples collected from Guangzhou showed that BTEX had significant higher concentrations in November than those in July.

Keywords: Aromatic hydrocarbons; Urban air pollution; Benzene; Vehicle emission

1. Introduction

The Pearl River Delta in Guangdong province has become China's most populated and developed region since the early 1980s. From 1978 to 1997, the oil consumption and gross industrial output in Guandong province had increased about 140 and 60 times, respectively (YCCG, 1998). With the rapid urbanization and industrialization in the region, more and more chemicals are introduced into the environment, and environmental pollution, including air pollution, has attracted increasing public and regulatory concerns.

The majority of previous studies on air pollution in the Pearl River Delta region focused on non-VOC pollutants like carbon monoxide (CO), sulfur dioxide (SO_2), nitric oxides (NO_x), total suspended particulates (TSP) and semi-volatile compounds in aerosols. Qin and Chan (1993) investigated traffic emission and street levels of CO and NO_x in the urban areas of Guangzhou, and found that 87% of CO and 67% of NO_x were sourced from vehicle exhaust. The dispersion of vehicle emission in the street canyons of Guanzhou was also discussed (Qin, 1993; Chan et al., 1995). Chan et al. (1994) studied the exposure of bicycle commuters to vehicle emission in the urban area of Guangzhou, and found CO and NO_x levels at some sampling sites exceeded those set in the China National Air Quality Standards. Simoneit et al. (1991), Fu et al. (1997) and

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Cheng et al. (1998a) studied organics in aerosols from Guangzhou and other cities in the Pearl River Delta. They used molecular markers and their assemblages to trace the origins of organics in airborne particulates and concluded that vehicle exhaust was the dominant source.

In some developed countries, especially the United States and Europe, there have been various studies on the nature and ambient concentrations of the aromatic hydrocarbons (e.g. NRC, 1980; Singh et al., 1985; Grosjean, 1992; Bowman et al., 1995; Brocco et al., 1997; Fraser et al., 1998; Grosjean et al., 1999; Skov et al., 2001), for their roles as toxic air contaminants and as important precursors to the formation of ozone (e.g. Carter, 1990; Bowman et al., 1995) and of secondary organic aerosols (e.g. Grosjean, 1992; Odum et al., 1997). However, information on ambient levels of aromatic hydrocarbons is not available for the Pearl River Delta. In our present study, we report urban roadside benzene, toluene, ethylbenzene and xylenes (BTEX) concentrations in the three cities, namely Guangzhou, Macau and Nanhai (Fig. 1) in the Pearl River Delta region. Cocheo et al. (2000) pointed out that people's actual outdoor benzene exposure is higher than the value calculated from the daily urban average benzene concentration and the time spent outdoors, since people are not exposed to urban background values but they are exposed in streets where the highest concentrations are found. Moreover, they are in the street canyons during rush hours when the concentration reaches its maximum. Since people usually start their work at about 9:00 a.m. in the morning and about 2:00 p.m. in the afternoon in the Pearl River Delta region, we collected our samples at 9:00–9:30 in the morning and at 2:00–2:30 in the afternoon, and measured BTEX at ground level to represent human exposure levels when people commute to their workplaces.

2. Sampling and analysis

2.1. Sampling locations

Table 1 summarizes the basic information of Guangzhou, Macau and Nanhai in terms of the population, urban areas, numbers of vehicles and others. These cities are very similar in climate in terms of their annual average temperature and are obviously different in their city layouts as well as their social and economic status. Compared to Nanhai, Guangzhou and Macau are

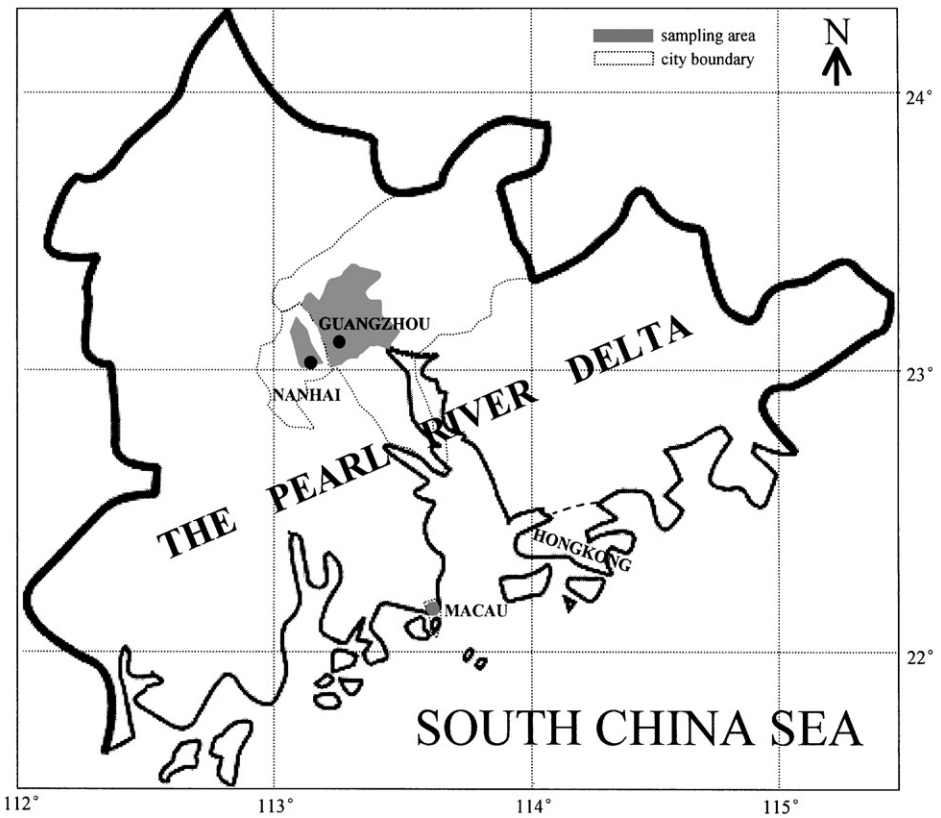

Fig. 1. Locations of the three cities and sampling areas in the Pearl River Delta, south China.

Table 1
Summaries of basic information about the three cities

	Guangzhou	Macau	Nanhai
Permanent residents (1996)	3,901,840 (urban) 2,658,668 (non-urban)	410,500 (urban) 7168 (Taipa) 3146 (Coloane)	1,043,186
Area (km^2)	1443.6 (urban) 5990.8 (non-urban)	7.0 (urban) 4.1 (Taipa) 7.2 (Coloane)	1152.7
Population density (person/km^2)	2703 (urban) 444 (non-urban)	58,643 (peninsular) 1748 (Taipa) 437 (Coloane)	905
Motor vehicles (1996)	583,778 (urban)	62,871 (1992)	~250,000
Annual average air temperature (°C)	21.6 (urban) (1996)	23.1 (1994)	21.8 (1996)

densely populated cities with much heavier traffic and more high-rise buildings and large mansions. Guangzhou lies in the north of the Pearl River Delta, it is an old city and has long been the political, economical and cultural center of south China. Commerce, service and high-technology industries are becoming dominant in its economy and manufacturing industries are becoming a smaller part. Macau is a seaside city with tourism and the gambling industry dominating its economy; its other industrial outputs are relatively trivial. Nanhai was historically an agricultural county before 1980. At present, however, agriculture contributes less than 10% of its GDP. In recent years there has been rapid development of village-, town-, and private-owned factories/mills, most of which are small- to medium-sized enterprises involved in producing ceramic, metal and wood products.

Samples were all taken at the curbsides at least 20 m away from bus stops, junctions and traffic lights. In each city, sampling locations were chosen based on typical urban microenvironment considering the traffic, population density and human activity pattern in the neighborhood. We also took advantage of the local air monitoring networks representative of different environments in the cities and collected part of the samples in the close vicinity of these monitoring stations.

2.2. Air sampling

Sorbent tubes and passivated canisters were typically used for sampling VOCs in air (Rudolph et al., 1990; Seeley and Broadway, 1994). In this study, multi-bed adsorbent tube based procedures were adopted for sampling. The commercial $7'' \times 1/4''$ multi-bed stainless steel sampling tubes (Tekmar Company, USA) were packed with Tenax and Carbosieve S-III. Before using the sampling tubes were conditioned by heating at 180°C for 24 h while purging with helium. After each use or storage they were reconditioned at 250°C for 2 h. Samples were all collected 1.2 m above the ground level. Volatile organic compounds were sampled by drawing the air through the sampling tubes with air sampling pumps (MP-5G, Amtek, USA), the flow rate and sampling times were set to 100 ml/min and 30 min, respectively. Both bubble flow meters and digital flow meters (DC-Lite, BIOS, USA) were used to check the flow rate. A digital flow meter can read the real time flow rate every 15 s, and store this data in its memory for retrieving in a computer to check the variation of flow rate in the whole sampling process. In all of our practice, after we tuned the flow rate to 100 ml/min, the variations of flow rate were less than 2%.

In Guangzhou two batches of samples, one on 10 July 1996 and one on 22 November 1996, were collected at 16 urban roadside locations. Each batch of samples was collected at 9:00–9:30 a.m. and 2:00–2:30 p.m. on the same day. In Guangzhou air samples were also collected in the Zhujiang Tunnel (~1240 m in length) near its open maintenance door in the middle of the tunnel (two samples at 9:00–9:30 and two at 14:00–14:30). Samples in Macau were collected on 20 November 1995. The Macau peninsular is the urban area containing more than 95% of Macau's population. Six roadside sampling sites were chosen in the Macau peninsular and samples were collected at 9:00–9:30 a.m. and 2:00–2:30 p.m. on the same day. Apart from air samples, gasoline from gasoline stations was also obtained in Guangzhou and Macau for BTEX analysis. In Guangzhou most gasoline sold was from CNPC with octane numbers of 90 (90#) and 97 (97#), respectively. We collected the two types of gasoline at five different gasoline stations scattered in urban Guangzhou. The three types of gasoline mostly used in Macau were also obtained from gas stations of three different oil companies. In Nanhai, four sites were selected for a three consecutive day sampling on 10–12 July 1996. Samples were also collected at 9:00–9:30 a.m. and 2:00–2:30 p.m. Each batch of air sampling had one sorbent tube as a field blank.

2.3. Laboratory analysis

By coupling a thermal desorption system (Tekmar 6000 AeroTrap) to a Hewlett Packard Gas Chromatograph/Mass Selective Detector (HP 5972 GC/MSD), the analysis of VOCs was performed with thermal desorption–gas chromatography–mass selective detector (TD–GC–MSD). A HP-VOC (30 m × 0.20 mm i.d. × 0.51 μm) capillary column was used with helium as carrier gas and a temperature program of initially 35°C for 2 min, at a rate of 5°C/min to 250°C for 10 min.

Compounds were identified by their retention times and their mass spectra. Standard gas mixtures (1.0 ppm) (Supelco TO-14 Calibration Mix) were first dynamically diluted with zero air, then sampled and analyzed using identical conditions to those used for the field samples, and multipoint calibration was performed for quantifying the BTEX in the air samples. *P*-xylene and *m*-xylene co-eluted in the GC columns in this study, and their concentrations were reported as their total instead of their individuals as most of previous researches have done. The method detection limits for benzene, toluene, ethylbenzene, (*m*+*p*)-xylene and *o*-xylene were 0.2, 0.1, 0.1, 0.2 and 0.1 $\mu g/m^3$, respectively. All gasoline samples have also been analyzed for their contents of BTEX by a HP 5890 gas chromatography equipped with a FID detector.

3. Results and discussion

3.1. BTEX concentration levels

Table 2 shows detected mean concentrations and ranges of BTEX in the roadside samples collected. Mean concentrations of B, T, E and X in the three cities were 51.5, 77.3, 17.8 and 81.6 $\mu g/m^3$ (in Guangzhou); 34.9, 85.9, 24.1 and 95.6 $\mu g/m^3$ (in Macau); 20.0, 39.1, 3.0 and 14.2 $\mu g/m^3$ (in Nanhai), respectively. BTEX levels in Guangzhou and Macau were much higher when compared with those reported in other North American and European cities, and BTEX levels in Nanhai lay within levels in these cities (Singh et al., 1985; Finlayson-Pitts and Pitts, 1986; Harkov et al., 1987; Bruckmann et al., 1988; Edgerton et al., 1989; Scheff and Wadden, 1993; Jose et al., 1998). The highest benzene concentration (185 $\mu g/m^3$) was found in a mixed area in northwest Guangzhou. The highest toluene concentration (314 $\mu g/m^3$) occurred in urban Macau. Benzene is a carcinogenic compound causing leukaemia. The World Health Organization (WHO, 1999) has estimated that a life time exposure of 1 $\mu g/m^3$ of benzene leads to about 6 cases of leukaemia per 1,000,000 inhabitants. From our measurements, benzene exposure in the urban street canyons may pose health risks especially to those who spend a longer time in the roadside microenvironment, such as policemen patrolling in the street and people working in roadside stands and shops. In July total BTEX levels in Guangzhou were more than twice those observed in Nanhai. This may be explained by their source strength and dilution conditions. Photochemistry is in this connection of minor importance. The lifetimes of benzene, toluene, ethylbenzene, *m*-xylene (the fastest reacting BTEX) are, respectively, 22, 4.7, 3.9 and 1.2 h at 10^7 molecules/cm^3 OH radical, a relative high concentration. The residence time of a compound in a street canyon is a few seconds and at the wind speeds of 2–3 m/s in the sampling periods in Nanhai and Guangzhou an air mass has travelled nearly 9 km or more within 1.2 h. In fact there exist street canyons in urban Guangzhou, which hinder the dispersion of these pollutants. Nanhai, however, is a newly developed and decentralized city, where buildings are usually buffered from the main traffic. Also there is less on-road traffic in Nanhai. So the less on-road source strength and better dilution conditions led to lower BTEX levels measured in Nanhai when compared to those in Guangzhou in July.

3.2. BTEX correlations

Fig. 2 shows the scatter plots of ethylbenzene against benzene, toluene and xylenes in the three cities. Table 3

Table 2
Detected means and ranges of benzene, toluene, ethylbenzene and xylenes in the three cities (in $\mu g/m^3$)

Sampling locations and periods		Benzene		Toluene		Ethylbenzene		Xylene	
		Mean	Range	Mean	Range	Mean	Range	Mean	Range
Guangzhou (07/10/96)	09:00–09:30	47.4	8.3 ~ 115	69.3	7.9–162	17.7	2.1–35.0	79.2	7.1–190
	14:00–14:30	38.1	10.1 ~ 102	65.7	16.0–131	13.8	1.7–37.6	50.9	12.2–161
Guangzhou (22/11/96)	09:00–09:30	67.3	14.9–185	92.1	20.7–202	22.5	5.9–56.7	102.9	14.6–243
	14:00–14:30	53.2	12.3 ~ 130	82.2	14.5–178	17.2	4.5–47.2	95.5	18.3–203
Macau (20/11/95)	09:00–09:30	40.7	12.0–109	106.9	18.6–314	31.2	4.9–59.2	111.5	33.7–281
	14:00–14:30	29.1	8.4–89.2	64.9	35.0–118	17.0	10.4–31.1	79.6	15.6–167
Nanhai (10 ~ 12/07/96)	09:00–09:30	24.6	2.9–70.4	49.6	11.3–164	3.6	1.1–7.2	17.0	5.0–47.1
	14:00–14:30	15.4	5.9–53.9	28.6	8.4–131	2.4	0.5–6.2	11.4	2.9–26.4

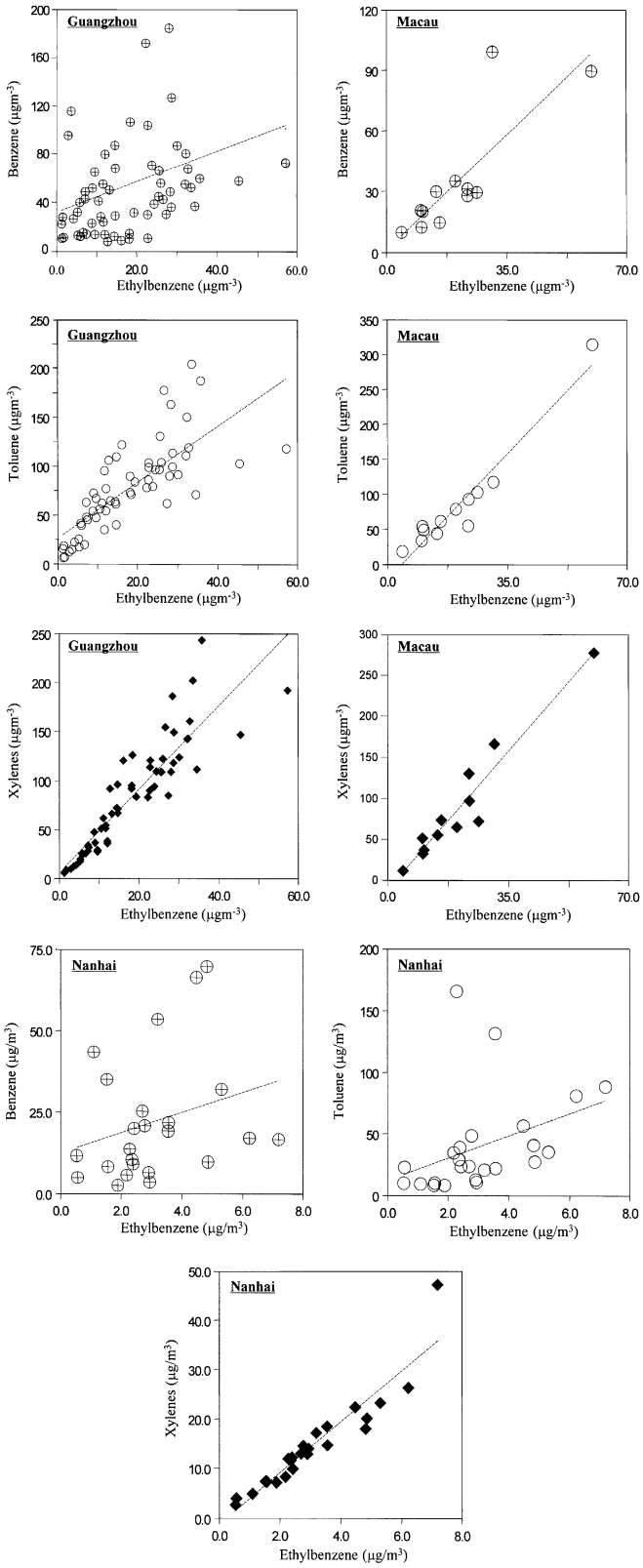

Fig. 2. Scatter plots of ethylbenzene levels against those of benzene, toluene and xylenes in the three cities.

Table 3
Summary of BTEX linear correlation coefficients (R) in the three cities

	Guangzhou ($n=64$)				Macau ($n=24$)				Nanhai ($n=12$)			
Compound	B	T	E	X	B	T	E	X	B	T	E	X
B	1	0.15	0.29	0.21	1	0.77	0.82	0.84	1	0.06	0.27	0.22
T		1	0.76	0.94		1	0.97	0.92		1	0.39	0.45
E			1	0.91			1	0.96			1	0.94
X				1				1				1

summarizes the BTEX mutual linear correlation coefficients in the three cities. Noticeably BTEX were significantly correlated ($P<0.01$) in Macau, and ethylbenzene and xylenes showed good linear correlations ($P<0.01$) in the three cities. In urban air, BTEX, especially ethylbenzene and xylenes, are typically related to emissions from gasoline vehicles and gasoline stations (McClenny et al., 1989; Cohen et al., 1991; Jose et al., 1998). The good mutual correlations among the aromatic hydrocarbons in Macau might indicate that they came predominantly from a single source, most possibly traffic-related emission. In Guangzhou, poor correlations between benzene and other BTEX species might imply that benzene had sources other than vehicle emission. Careful examination revealed that there were site-specific BTEX patterns, implying possibly different sources in different sites, especially for benzene. In Nanhai, even toluene had poor correlations with other species. The poor mutual correlations of BTEX in Nanhai except between ethylbenzene and xylenes might suggest that there were various sources of BTEX in the city. Moreover, as shown in Fig. 2, toluene was found in relatively high levels in two samples collected the same day at one site in Nanhai, indicating an irregular emission source of toluene other than traffic-related emissions.

3.3. B:T:E:X ratios

Table 4 shows the average B:T:E:X ratios in the three cities. X/E ratios in the three cities were relatively close when compared with B/E and T/E ratios. B/E and T/E ratios were much higher in Nanhai than those in Macau and Guangzhou. Previous studies in tunnels indicating motor vehicle emissions found B/E ratios typically <5 and T/E ratios <6 (Lonneman et al., 1986; Doskey et al., 1992; Haszpra and Szilagyi, 1994; Chang et al., 1996). Compared to samples from a tunnel in Guangzhou, the X/E ratio was almost the same as urban roadside samples from both Guangzhou and Nanhai, but B/E and T/E ratios were significantly higher in Nanhai (Table 4). In Nanhai and Guangzhou, the types of gasoline used by vehicles, as well as vehicle types and their maintenance conditions, were very similar. The unusual B/E and T/E ratios (6.7 and 14.4, respectively) in Nanhai might also confirm the significant contribution of sources other than traffic-related emission to BTEX in the city. Compared to Guangzhou and Nanhai, the gasoline used in Macau was different in its composition of BTEX, the maintenance of vehicles in Macau were also much better. These might also be reasons for the difference of their BTEX ratios. Compared the B/E ratios in tunnel and roadside samples to those in gasoline samples (Table 4), an enhancement of benzene were observed relative to ethylbenzene, possibly due to the generation of benzene during combustion (Christensen et al., 1999; Palmgren et al., 2001).

Table 4
Average BTEX ratios relative to ethylbenzene (in weight) in the air samples, gasoline and tunnel air samples

	B	T	E	X
Guangzhou	2.9	4.3	1	4.6
Macau	1.4	3.5	1	4.0
Nanhai	6.7	14.4	1	4.7
Gasoline (90#) (Guangzhou)	0.8	3.1	1	3.7
Gasoline (97#) (Guangzhou)	1.3	4.2	1	3.8
Tunnel air samples (Guangzhou)	3.3	5.0	1	4.9
Macau Gasoline type 1	0.5	43	1	4.4
Macau Gasoline type 2	0.12	2.9	1	4.0
Macau Gasoline type 3	0.14	3.0	1	3.9

Macau is a city dominated by tourism and other industries play a less important role than those in Guangzhou and Nanhai. So vehicle emission becomes the single prominent source. That is why BTEX showed good linear correlation in Macau. Nanhai had less traffic and far more industrial activities, so contribution of traffic-related emission shared a lesser portion and other sources (like industry activities) of benzene and toluene became more important. These resulted in higher B/E and T/E values in Nanhai.

3.4. Comparison of summer and autumn BTEX in Guangzhou

In Guangzhou, autumn (November) BTEX concentration levels were found higher than their summer

(July) concentration levels when the observed wind speeds were similar (SE/2–3 m/s on 10 July, 1996 and NNE/2–3 m/s on 22 November, 1996). Average BTEX concentrations in the autumn time samples were 26% (ethylbenzene) to 51% (xylenes) higher than those in the summer time samples. Cheng et al. (1998b) also found much higher concentrations of polycyclic aromatic hydrocarbons in aerosols collected in autumn than in summer. As a subtropical city, Guangzhou has smaller annual air temperature variation (in 1996, monthly average air temperature in July is 28.8°C, and 21.3°C in November). The pattern of anthropogenic activities that leads to BTEX emission, to a reasonable extent, also had little changed between July and November. As mentioned above, photochemistry might be of minor importance to the gap of the urban roadside BTEX levels. Therefore, the summer and autumn concentration differences might not be the consequence of variation of source input or photochemistry, but the result of variation in other factors such as meteorological conditions. The seasonal change of monsoon winds in the Pearl River Delta is very important in seasonal variation of pollutants in the region (Chan et al., 1998). In summer time air masses from the South China Sea have a dilution effect for the BTEX levels in the Pearl River Delta cities. In November, however, continental outflows from continent China are much richer in air pollutants than marine air masses. Also in November the air is much more stable than in July (Chan et al., 1998), thus the dispersion of gas-phase pollutants are hindered to a larger extent in urban roadside microenvironments.

4. Conclusion

The urban roadside BTEX levels in Guangzhou and Macau were much higher than those in Nanhai and reported levels in other North American and European cities. Mean concentrations of benzene, toluene, ethylbenzene and xylenes in the three cities were 51.5, 77.3, 17.8 and 81.6 $\mu g/m^3$ (Guangzhou); 34.9, 85.9, 24.1, 95.6 $\mu g/m^3$ (Macau); 20.0, 39.1, 3.0 and 14.2 $\mu g/m^3$ (Nanhai), respectively. The difference of these BTEX levels in urban roadside microenvironments might be explained by their source strengths and dilution conditions. Of particular concern is the high urban roadside benzene concentrations ($> 20\ \mu g/m^3$) in the three cities, especially in Guangzhou and Macau. In Guangzhou and Macau, the high levels of BTEX, particularly benzene, which were probably related to the street canyon effect and traffic density, might cause health risks to people working or residing in the roadside microenvironments. The BTEX pattern and correlation analysis indicated that in Macau emissions were predominantly traffic-related, and in Guangzhou and Nanhai there were sources other than vehicle emission. In Guangzhou BTEX concentrations in autumn samples are found significantly higher than those in summer samples. The present study is only a preliminary one indicating a likely problem of high BTEX concentrations in urban cities like Guangzhou and Macau in the Pearl River Delta. Further study is necessary for a comprehensive estimation of emissions as well as people's exposure and risks of these aromatic hydrocarbons in the region.

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