A source study of atmospheric polycyclic aromatic hydrocarbons in Shenzhen, South China

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Abstract Air pollution has become a serious problem in the Pearl River Delta, South China, particularly in winter due to the local micrometeorology. In this study, atmospheric polycyclic aromatic hydrocarbons (PAHs) were monitored weekly in Shenzhen during the winter of 2006. Results indicated that the detected PAHs were mainly of vapor phase compounds with phenanthrene dominant. The average vapor phase and particle phase PAHs concentration in Shenzhen was 101.3 and 26.7 ng m^{-3}, respectively. Meteorological conditions showed great effect on PAH concentrations. The higher PAHs concentrations observed during haze episode might result from the accumulation of pollutants under decreased boundary layer, slower wind speed, and long-term dryness conditions. The sources of PAHs in the air were estimated by principal component analysis in combination with diagnostic ratios. Vehicle exhaust was the major PAHs source in Shenzhen, accounting for 50.0% of the total PAHs emissions, whereas coal combustion and solid waste incineration contributed to 29.4% and 20.6% of the total PAHs concentration, respectively. The results clearly indicated that the increasing solid waste incinerators have become a new important PAHs source in this region.

Keywords Shenzhen · Air · Polycyclic aromatic hydrocarbons

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Introduction

In the Pearl River Delta (PRD), South China, air pollution has become a serious problem with significant negative impact on the public health and social economy. Similar to many other metropolitan cities, the air pollution problem in this region is caused by the ever-increasing demand of energy by a growing population and economic bloom. Heavy smog caused by coal burning, industrial pollution, and vehicle exhausts have ruined the air quality and reduced visibility over the PRD region. The serious air pollution has taken its toll on public health and daily life; the Municipal Environmental Bureau sometimes has to warn local residents to stay home and away from the smog-filled air.

Polycyclic aromatic hydrocarbons (PAHs) are ubiquitous pollutants in the environment, arising mainly from the incomplete combustion of fossil fuels, organic materials, and wood (Benner et al.

1989; Baek et al. 1991). Anthropogenic activities such as vehicles exhaust, residential heating, and power generation are considered to be the principal PAHs sources to the environment (Mastral and Callen 2000; Mcdonald et al. 2000; Wornat et al. 2001). Nowadays, PAHs have been identified to be one of the major toxic air pollutants in Chinese urban air, and numerous studies have been carried out to understand the level, sources, and the health effect (Guo et al. 2003; Li et al. 2006; Chang et al. 2006; Lu et al. 2007).

Located at the eastern side of the PRD region, Shenzhen is a Special Economic Zone (SEZ) and one of the most important industrial centers in China, with an area of 2,021 km^2 and a population approaching ten million. Since the establishment of the SEZ in 1980, Shenzhen has made rapid economic progress; its gross domestic product surpassed US $60 billion in 2006, which has grown 1,800 times over the past 26 years. However, the rapid urbanization and industrialization may have posed negative effect on the environment. As reported in the Shenzhen Environmental Bulletin, hazy days have been increasing year on year since 2003 as the air quality deteriorates; the major culprit is believed to be vehicle and industrial emissions. With the rapid economic development and city expansion, municipal solid waste (MSW) is becoming a severe problem in China. MSW incineration has been recently emerged as a primary treatment method for volume reduction, harmlessness, and energy recycle from MSW. Until 2006, there were eight MSW incineration power plants in operation in Shenzhen, treating 6,000 tons of MSW daily. The thermal process in MSW incineration has the potential of emitting diverse types of organic pollutants to the environment, such as PAHs, polychlorinated biphenyls, and polychlorinated dibenzo-*p*-dioxins/dibenzofurans, which may endanger the ambient environment and people's health.

To our knowledge, few studies are available on PAHs pollution in Shenzhen. The present work aimed to investigate the PAHs level in the urban air of Shenzhen and to understand the sources and contributions. The results will be of particular value for a better understanding of the air pollution in megacities and for developing effective control strategies.

Materials and methods

Air sampling

The sampling site was selected on the roof top of the library at Shenzhen University (Fig. 1). Sampling was conducted for a consecutive 24-h period during the day and night on a weekly base from December 2006 to February 2007. Samples were collected with a high-volume air sampler, at a flow rate of 335 m^3 day^{-1}. Suspended particulate was retained on a Whatman glass microfiber filter (GFF; grade GF/A, 20.3 × 25.4 cm), and vapor phase absorbed on a polyurethane foam plug (PUF; length 8.0 cm, diameter 6.25 cm, density 0.035 g cm^{-3}). Prior to sampling, the filter papers were baked at 450°C for 4 h; the PUFs were pre-extracted in dichloromethane (DCM) for 48 h, dried under vacuum, and sealed in precleaned glass jars prior to use. After sampling, the filters were folded and sealed in aluminum foil envelopes until weighing for determination of total suspended particulate mass. The PUFs were sealed in the jar and stored in a freezer at −20°C until further analysis.

Analytical procedure

Filter and PUF samples were spiked with surrogates (naphthalene-d8, acenaphthene-d10, phenanthrene-d10, pyrene-d10, chrysene-d12, and perylene-d12) and Soxhlet-extracted for 48 h with DCM. Activated copper granules were added to the collection flask to remove elemental sulfur. Concentrated extracts were loaded on a 10-mm i.d. silica/alumina column, packed from the bottom to top with neutral alumina (6 cm, 3% deactivated), neutral silica gel (2 cm, 3% deactivated), and anhydrous sodium sulfate (1 cm, 450°C baked), eluted with 60 mL of hexane/dichloromethane (6:4 *v/v*), and the final volume was concentrated to 200 μL under a gentle N$_2$ stream. A known amount of internal standard (hexamethybenzene) was added to all the samples prior to PAH analysis.

PAHs were quantified on a HP-5971 GC-MSD system operating at 70 eV under scan mode. Analytics were separated with a DB5-MS column (30 m × 0.25 mm diameter, 0.25 μm film

Fig. 1 Map of the Pearl River Delta with sampling locations

thickness). The GC temperature program was as follows: initially at 80°C for 5 min, increased to 290°C at 3°C/min, and held for 30 min. A 1-μL sample was injected into the splitless injector with a 5-min solvent delay. Peak confirmation and quantification was performed on a DOS-based HP ChemStation system. The following 16 PAHs were selected as target compounds in the analysis: naphthalene (Nap), acenaphthylene (Acy), ace-naphthene (Ace), fluorene (Flo), phenanthrene (Phe), anthracene (Ant), fluoranthene (Flu), py-rene (Pyr), benzo[*a*]anthracene (BaA), chrysene (Chry), benzo[*b*]-fluoranthene (BbF), benzo[*k*] fluoranthene (BkF), benzo-[*a*]pyrene (BaP), dibenz[*a*, *h*]-anthracene (DiA), indeno[1,2,3-*c*, *d*] pyrene (InP), and benzo[*g*, *h*, *i*]perylene (BghiP).

Quality assurance

Field blanks, which accompanied samples to the sampling sites, were used to determine background contamination. Lab blanks and field blanks showed no detectable PAHs. In addition, surrogate standards were added to all the samples (including quality assurance samples) to monitor the procedural performance and matrix effects. The mean recoveries in field samples were naphthalene-D_8 30%, acenaphthene-D_{10} 79%, phenanthrene-D_{10} 101%, chrysene-D_{12} 95%, and perylene-D_{12} 88%. PAH concentrations were corrected for recovery efficiency. A backup PUF was attached to the sampler to check the possible breakthrough of gaseous PAHs. Typically, the backup PUF contained <5% of the total vapor phase component, with greater penetration to the second plug observed for the lighter PAH compounds (phenanthrene 3.3%, fluorene 1.6%, anthracene 0.9%, and fluoranthene 0.5%).

Results and discussion

General remarks on gas/particle phase distribution and compounds detected

In this study, the gas (PUF) and particle (GFF) phase samples were analyzed separately. In general, the lower molecular weight PAHs (two to three rings) appeared to be associated with the vapor phase, and high molecular weight PAHs (five to six rings) were primary attached with the particulate phase. As shown in Table 1, the total PAHs occurred primarily in the gas phase; the

Table 1 PAHs concentrations in gas and particle phase/ng m^{-3}

Compounds	PUF			GFF		
	Min	Max	Mean	Min	Max	Mean
Nap	0.9	9.3	5.0	0.0	0.2	0.1
Ace	1.8	17.6	7.8	0.0	0.1	0.1
Acy	0.7	2.3	1.4	0.0	0.0	0.0
Flo	13.2	30.5	20.7	0.0	0.2	0.1
Phe	37.0	69.3	49.2	0.2	3.6	1.6
Ant	2.5	8.6	5.1	0.0	0.2	0.1
Flu	4.2	11.9	9.1	0.1	5.7	2.8
Pyr	3.2	9.0	6.7	0.1	4.8	2.6
BaA	0.0	0.3	0.2	0.2	2.0	1.3
Chry	0.2	0.9	0.6	0.3	5.2	3.3
BbF	0.0	0.2	0.1	0.6	4.5	3.0
BkF	0.0	0.2	0.1	0.3	3.1	2.2
BaP	0.0	0.2	0.2	0.3	2.6	1.8
InP	0.0	0.4	0.1	0.8	4.3	3.3
DiA	0.0	0.4	0.3	0.6	1.1	0.9
BghiP	0.0	0.4	0.2	0.7	3.3	2.4
ΣPAHs	73.2	160.1	101.3	15.2	30.7	26.7

most abundant PAHs in the air are, on average, Phe (21.8% of the total PAHs), Flu (11.6%), Pyr (6.7%), and Chry (7.3%). The profiles of PAHs in ambient air were dependent on their source and environmental variables. For remote areas like ocean and high mountain regions, PAHs are often low molecular compounds as they are more volatile and can be transported over longer distances (Halsall et al. 1997; Liu et al. 2005a, b; Ding et al. 2007). In contrast, local sources like industrial and vehicular emission frequently result in higher percentages of particle phase PAHs (Tasdemir and Esen 2007). By comparison, the PAH distribution pattern in Shenzhen was similar to other studies for urban air (Ohura et al. 2004; Li et al. 2006).

Levels and trends of PAHs

The concentration of total PAH ranged from 109.1 to 189.6 ng m^{-3}, with a mean value of 128.0 ng m^{-3}. It was several times higher than the level reported 10 years ago (averaged ∼17 ng m^{-3}; Chen et al. 1998). Atmospheric PAHs have been extensively studied in urban areas. The total concentration of PAHs in urban sites of Beijing (Zhou et al. 2005) and Tianjin (Wu et al. 2006a, b) was in the range of 1.4–152 ng m^{-3} and 239–537 ng m^{-3}, respectively. In Seoul, South Korea, the average total PAHs concentration was 89.3 ± 74.3 ng m^{-3} (Park et al. 2002), and the mean concentration of total PAH was 456 ± 524 ng m^{-3} in the city of Bursa, Turkey (Tasdemir and Esen 2007). In general, atmospheric PAH concentrations in Shenzhen are within the range of PAH levels reported in urban air elsewhere; it was slightly lower than those found in Guangzhou, where the average total gaseous and particulate PAHs concentrations were 313 and 24 ng m^{-3}, respectively (Li et al. 2006), but were much higher than those found in the background sites of the Pearl River Delta (Liu et al. 2006).

The meteorological conditions have a large effect on PAH concentrations. A positive relationship between air temperature and concentration was found for the lighter molecular weight PAHs, whereas the heavier molecular weight PAHs exhibited a negative relationship with temperature (Lee and Jones 1999). In addition to temperature, wind speed, air humidity, and the concentration of atmospheric particulates will also influence PAH concentrations in the air (Chang et al. 2006). Figure 2 shows the temporal variation of total PAH concentrations during the sampling period from December 2006 to February 2007. The fluctuation of PAH levels was small except in heavy hazy days (January 11–12, 2007). Shenzhen features a typical monsoon-controlled climate, wet and hot in summer and dry and cool in winter. Due to the meteorology of this region which was affected by mountainous landscape, the river delta, and surrounding ocean, the temperature inversion frequency is high in winter, and particulate pollutants are often trapped in the air result in so called haze episode (with visibility less than 10 km and under relative humidity <80%). It has been suggested that the proportion of fine particles (0.1–2 μm) in total suspended particles will increase during haze days (Wu et al. 2006a, b). As most of the particle PAHs are bounded to fine particles especially below 2.0 μm (Duan et al. 2007), the higher PAHs concentration observed in haze episode may result from the accumulation of pollutants under decreased boundary layer, slower wind speed, and long-term dryness conditions.

Fig. 2 Time variation of atmospheric PAHs concentration in Shenzhen
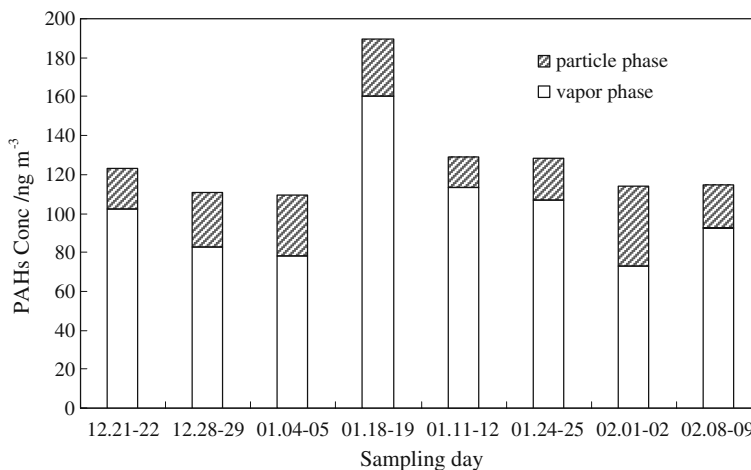
Principal component analysis and sources

PAHs are ubiquitous pollutants in urban atmosphere; understanding the contribution of the various sources is critical to appropriately control PAH levels in the environment. It is suggested that the concentration of some marker compounds and their ratios could be used to distinguish different PAH sources (Simoneit et al. 1991; Li and Kamens 1993; Gogou et al. 1996). Chemical mass balance (CMB) model and factor analysis have also been used to identify the sources of organic compounds (Duval and Friedlander 1981; Pistikopoulos et al. 1990; Li and Kamens 1993; Harrison et al. 1996; Simcik et al. 1999; Larsen and Baker 2003). However, there are certain limitations as CMB models require the input of source emission profiles to calculate source contributions, and molecular markers often lack suitable "source signatures" (Sporstol et al. 1983). Principal component analysis (PCA) modeling, providing information on source contributions based on the time variation of the organic composition, has been widely utilized for source characterization of air pollution (Harrison et al. 1996; Simcik et al. 1999; Larsen and Baker 2003). In this study, PCAs, in combination with multiple linear regression (MLR), were performed to identify and apportion the air PAHs in Shenzhen.

There are two significant concerns regarding the use of PAHs in source apportionment studies (Larsen and Baker 2003). PAHs partitioning between gas and particulate phases complicates both the sampling methodology and characteristic source signatures. In this study, the measured gas- and particulate-phase PAH concentrations were combined to avoid problems associated with partitioning. Another concern is the loss of the source signature by the destruction of PAHs by photochemical processes. Assumed that the atmospheric conditions were relatively constant during the sampling days, the photochemical loss should have only a minor effect on the source apportionment of PAHs.

PCA followed by multiple linear regression of the data was performed using SPSS software. Factor analysis was conducted for 15 PAHs, except for Nap due to its high volatility and low recovery. Three principal factors were identified by PCA, explaining 97.6% of the total variance in the data (Table 2). Factor 1, accounting for the majority of the total variance (49.1%), is highly loaded with BaA, Chry, BbF, BkF, InP, and BghiP. Among these compounds, BkF, Ind, and BghiP were considered as the tracers of vehicle emission (Miguel and Pereira 1989; Li and Kamens 1993; Harrison et al. 1996). Previous studies have also suggested that vehicle exhausts were the dominant PAH source in the air of PRD region (Bi et al. 2003; Liu et al. 2005a, b; Li et al. 2006). Therefore, we suggested that factor 1 was related to vehicle emissions. The second factor, which accounted for 28.5% of total variance, was highly loaded on light weight PAHs (Acy, Ace, Flo, Phe, and Ant). Flo was reported as the dominant PAH in coke oven emission (Khalili et al. 1995) and coal

Table 2 Factor pattern of air PAHs in Shenzhen

Compound	Factors		
	Factor1	Factor 2	Factor 3
Acy	0.261	0.955	0.113
Ace	−0.107	0.781	−0.537
Flo	−0.078	0.981	−0.128
Phe	−0.150	0.711	0.679
Anth	−0.154	0.946	0.186
Fluo	0.179	−0.129	0.965
Pyr	0.142	0.118	0.972
BaA	0.991	−0.002	0.057
Chry	0.959	−0.099	0.247
BbF	0.962	−0.140	0.165
BkF	0.981	−0.102	0.089
BaP	0.991	−0.112	0.037
InP	0.988	0.096	−0.017
BghiP	0.974	0.140	−0.028
Variance (%)	49.1	28.5	19.9

combustion (Masclet et al. 1987) and was often used as the tracer of coal combustion factor in PCA (Simcik et al. 1999), whereas Phe, Ant, Flu, Pyr, BaA, Chr, BaP, and BghiP are identified to be the source fingerprints of coal combustion and coke production (Duval and Friedlander 1981). Therefore, factor 2 was selected to represent coal combustion. In Shenzhen, the fire power generation accounts for more than 70% of the total electricity generated, and most are coal-fire power plants (Bureau of Statistics of Shenzhen 2006). Although the major thermal process that generate PAHs have changed to vehicle exhaust, coal combustion (especially for power generation) is still an important PAH source in this region.

Factor 3, accounting for 19.9% of the total variance, is highly loaded with Flu and Pyr. Flu and Pyr are originated from many types of combustion processes typically found in urban areas (Rogge et al. 1993a, b; Müller et al. 1998; Schauer et al. 1996). However, previous studies indicated that Pyr was also emitted in large amounts by biomass burning such as grass and savannah fires (Masclet et al. 1995) or wood fires (Freeman and Cattell 1990; Jenkins et al. 1996). Pyrene and fluoranthene have been used as an indicator of incineration (Yan et al. 2004; Wang et al. 2007). Besombes et al. (2001) found that the PAH profile in the surrounding municipal incinerator was characterized by a high relative concentration of Flu and Pyr.

In Shenzhen, there were eight municipal waste incineration plants in operation till 2006, with an expected capacity rating of 6,000 ton/day each and an electricity capacity of 100,000 Kw h^{-1} each. These plants were used to incinerate more than 50% of all the treated municipal solid waste, and Nanshan MSW incineration power plant (capacity of 800 ton/day each) is only about 8.0 km from our sampling site. Based on the above information, we suggested that factor 3 might relate to MSW incineration.

The ultimate goal of performing PCA/MLR is to determine the percent contributions of different PAH sources for a given sample of ambient air. Following the MLR procedure described by Larsen and Baker (2003), the mean percent contributions of different PAH sources were quantified, and the result was 50.0% for the vehicular source, 29.4% for the coal source, and 20.6% for the municipal solid waste incinerator source.

Conclusion

Atmospheric PAHs were weekly monitored in Shenzhen during the winter of 2006. The detected PAHs were mainly of vapor phase compounds and dominated by Phe, Flu, Pyr, and Chry. The total PAHs concentration ranged from 110 to 190 ng m^{-3}, with a mean value of 128 ng m^{-3}, which was several times higher than the level reported 10 years ago. Higher PAH concentration observed during haze weather may result from the enhanced accumulation of pollutants under decreased boundary layer, lower wind speed, and long-term dryness conditions. Vehicle emission was found to be the major PAH source in Shenzhen, accounting for 50.0% of the total PAHs concentration, whereas coal combustion and solid waste incineration contributed to 29.4% and 20.6% of the total PAH concentration, respectively. The increasing number of solid waste incinerators have become a new important PAH source in this region.

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