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A comparison study of atmospheric polycyclic aromatic hydrocarbons in three Indian cities using PUF disk passive air samplers

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HIGHLIGHTS

- Total PAH concentrations exhibited a trend of Kolkata > Mumbai > Chennai.
- There was no significant urban–rural pattern in India.
- Industrial emission could be important source in Indian cities.

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ABSTRACT

A passive air sampling campaign was conducted to measure polycyclic aromatic hydrocarbons (PAHs) in Kolkata, Mumbai and Chennai, the three major cities of India. The measured total PAH concentrations ranged from 6480 to 54,800 ng sample^{-1}, comparable to the highest levels across the globe. Three- to four-ring PAHs were the dominant components in the atmosphere. According to the spatial distribution, the PAH concentrations were the highest in Kolkata and the lowest in Chennai. Kolkata and Mumbai were characterized by a relatively high proportion of HMW (high molecular weight) PAHs, which can be ascribed to the difference in the economic and energy structures in the urban areas. Surprisingly, there was not significant decrease in PAH concentrations from urban to rural sites. Rural sources, generally associated with traditional biomass combustion, could be as important as urban sources in India. In this study, the total BaP_{eq} (BaP toxic equivalent) concentrations generally exceeded the human exposure limit, posing potential risk to the health of the local residents.

1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are a group of carcinogenic, genotoxic and ubiquitous semivolatile organic compounds (SVOCs) in the atmosphere. Although natural processes are potential sources of PAHs, these compounds are mostly associated with anthropogenic activities such as incomplete combustion and the pyrolysis of fossil fuels or wood. High levels are generally observed in densely populated areas, posing adverse effects on human health. Due to health concerns, great efforts have been devoted to the study of PAHs in cities in recent years.

India is a developing country challenged by a dense population and low energy efficiency. In India approximately 85%–90% of cooking energy is from the burning of biofuels such as wood, agricultural waste and dunk cake, which account for 47% of the energy consumption (Tata Energy Research Institute). Zhang and Tao (2009) estimated that the annual PAH emissions from India was 90 Gg y^{-1}, constituting 23.6% of the total global emissions. Traditional biomass resources are still used to meet the overwhelming proportion of India's energy demand (Masih et al., 2012), which could lead to a significant amount of PAH release. Some reports have indicated that atmospheric PAH levels in Indian cities are higher than in other urban sites around the world (Sharma et al., 2007; Singh et al., 2011). Hence, a large portion of the world's population is exposed to high levels of PAHs (Simth et al., 2004). However, previous studies have mainly focused on particle-associated PAHs (Karar and Gupta, 2006; Mohanraj et al., 2010, 2011), and limited work has been carried out on both gaseous and particulate phase PAHs in India (Sahu et al., 2004; Pandit et al., 2006). In addition, previously measured local PAH

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concentrations ranged from several ng m^{-3} (Pandit et al., 2006) to several ug m^{-3} (Singh et al., 2011), making assessment on a regional scale difficult.

The passive air sampling (PAS) method (Shoeib and Harner, 2002) is an economical alternative to traditional active air sampling (AAS) when monitoring SVOCs on a large scale and for surveying atmospheric PAHs (Jaward et al., 2004). PAS has been applied to assess the seasonal and spatial trends of other SVOCs in India (Zhang et al., 2008; Pozo et al., 2011), and it has been proved to be an efficient tool to study the regional variation of organic compounds compared with high volume air sampling in India (Chakraborty et al., 2010). The present study uses the same method in three of India's major coastal cities during the wintertime: Kolkata in the east, Mumbai in the west and Chennai in the south. According to India's 2011 census, Mumbai, Chennai and Kolkata had 12.3, 4.68, 4.49 million residents, respectively. Similarly, the local GDP in Mumbai was approximately twice that in Kolkata and Chennai. All three cities have a broad industrial base. Mumbai's economy largely depends textile mills and the seaport. Chennai's economy is also based on the IT technology and service sectors. In contrast, Kolkata hosts many heavy industrial factories, which may lead to pollutants emission. This study aims to 1) characterize the spatial trend of atmospheric PAHs in Indian cities and 2) estimate the potential human exposure risk in those areas.

2. Material and methods

2.1. Passive air sampling

The sampling campaign was conducted at urban, suburban and rural sites in three major Indian cities (Kolkata, Mumbai and Chennai) during the winter of 2006. Details on the sampling site can be found in previous work (Chakraborty et al., 2010). Polyurethane foam (PUF) disks (14.0 cm diameter; 1.30 cm thick; density, 0.0170 g cm^{-3}) were pre-extracted with a mixture (1:1, V/V) of acetone and dichloromethane (DCM) at 100 °C and 1500 psi for 15 min. Before sampling, all of the samples were wrapped with clean aluminium foil, sealed in Teflon bags and transported to the sampling sites. The deployment of PUF disks followed the standard procedures (Zhang et al., 2008). After 28-day sampling periods, the PUF disks were wrapped, sealed and transported again, and kept at −18 °C before analysis.

2.2. Analytical procedure

The samples were Soxhlet extracted with dichloromethane (DCM) for 18 h and spiked with deuterium-labelled PAHs (D8-Naphthalene, D10-Acenaphthene, D10-Phenanthrene, D12-Chrysene, and D12-Perylene) as surrogates. Excessive activated copper granules were added to the collection flask to remove elemental sulphur. Each sample extract was concentrated by a rotary evaporator and was solvent-exchanged into hexane. An 8 mm i.d. alumina/silica column containing, in order, anhydrous sodium sulphate (1 cm), neutral silica gel (3 cm, 3% deactivated) and neutral alumina (3 cm, 3% deactivated) was applied for purification. The target fractions were eluted with 20 ml of a mixture of dichloromethane and hexane (1:1, V/V). Finally, the eluent solvent was blown down to 25 μL under a gentle stream of nitrogen. Hexamethyl benzene was added as an internal standard before analysis.

The analytical procedure was conducted using an Agilent 7890 gas chromatograph equipped with a capillary column (DB-5MS, 30 m, 0.25 mm, 0.25 μm) and a mass spectrometric detector (MSD, Agilent 5975). Target compounds were identified by comparing the measured mass spectra and retention times to reference standards. The quantified PAHs were acenapthene (Ace), acenapthylene (Acey), fluorine (Flu), phenanthrene (Phe), anthracene (Ant), fluoranthene (Flur), pyrene (Pyr), benzo[a]anthracene (BaA), chrysene (Chr), benzo[b]fluoranthene and benzo[k]fluoranthene (BbkF), benzo[a]pyrene (BaP), dibenzo[a,h]anthracene (DahA), benzo[g,h,i]perylene (BghiP) and indeno[1,2,3-c,d]pyrene (Ind).

2.3. QA/QC

The instrumental stability was checked daily using PAH standards, and the deviation was less than ±15%. Isotope-labelled PAHs were added to samples and field blanks to monitor the analytical and sampling procedures. The recoveries were 69.3% ± 7.2%, 84.6% ± 9.8%, 95.0% ± 15.7% and 101.1% ± 19.1% for acenapthene-D10, phenanthrene-D10, chrysene-D12 and perylene-D12, respectively. For quality assurance and quality control (QA/QC), target compounds in field blanks were determined as the samples. Instrumental detection limits (IDLs) were integrated where the signal-to-noise ratio was equal to 3. The method detection limits (MDLs) were defined as the average concentrations of target compounds in field blanks plus three times the standard deviation. The MDLs of the target PAHs ranged from 1.0 to 9.0 ng $sample^{-1}$ at least one order of magnitude lower than the values in the samples, except Ind in the 3 samples from Chennai. None of the reported data in this study was recovery corrected.

3. Results and discussions

3.1. Concentrations and congener profiles

In this study, 15 PAHs were identified and quantified in passive atmospheric samples. Generally, the target compounds were above the MDL, with the exception of the three samples from Chennai. The mean values were calculated using the measured values if above the MDL or 1/2 MDL if below the MDL. The total PAH concentration ranged from 6480 to 54,800 ng $sample^{-1}$, with a median concentration of 19,300 ng $sample^{-1}$. The highest and lowest values were detected at the rural sites of Chennai (CN-06) and Kolkata (KL-16), respectively (Fig. 1). The concentration levels were higher than the PAS observed in Europe (Jaward et al., 2004) and other urban areas (Bohlin et al., 2008; Gouin et al., 2010) and comparable to some source regions in China (Liu et al., 2007). Chakraborty et al. (2010) used 7–10 days of AAS to represent the PAS of the 28-day deployment period. The estimated sampling rate (R) for similar AAS and PAS sites ranged from 2.2 to 7.8 m^3 d^{-1}. If following the average sampling rate of 3.6 m^3 obtained from the same project, the concentration of total PAHs in India ranged from 64 to 544 ng m^{-3}. These concentrations are within the highest range in the world, even though PAS may underestimate HMW (high molecular weight) PAHs. The concentrations in Indian air were dominated by the 3–4-ring PAHs, following the order Phe (~32%) > Pyr (~15%) > Chr (~12%)> Flur (~11%)>Flu (~10%), while the proportions of the other PAHs were <7%. Compared with the congener profiles in cities (Liu et al., 2007; Bohlin et al., 2008) and background sites (Halse et al., 2011), the Indian air was characterized by a relatively high proportion of HMW PAHs. An enrichment of HMW PAHs in the air was also recorded at a rural site in Chennai (Masih et al., 2012). Although there is a discrepancy in congener profiles between the PAS and AAS technique (Bohlin et al., 2009), this report still confirms that PUF–PAS captures a part of particle-associated HMW PAHs.

3.2. Spatial trend

The sampling sites can be categorized by geographical location, economic level, land use type, meteorological factors and so on. In

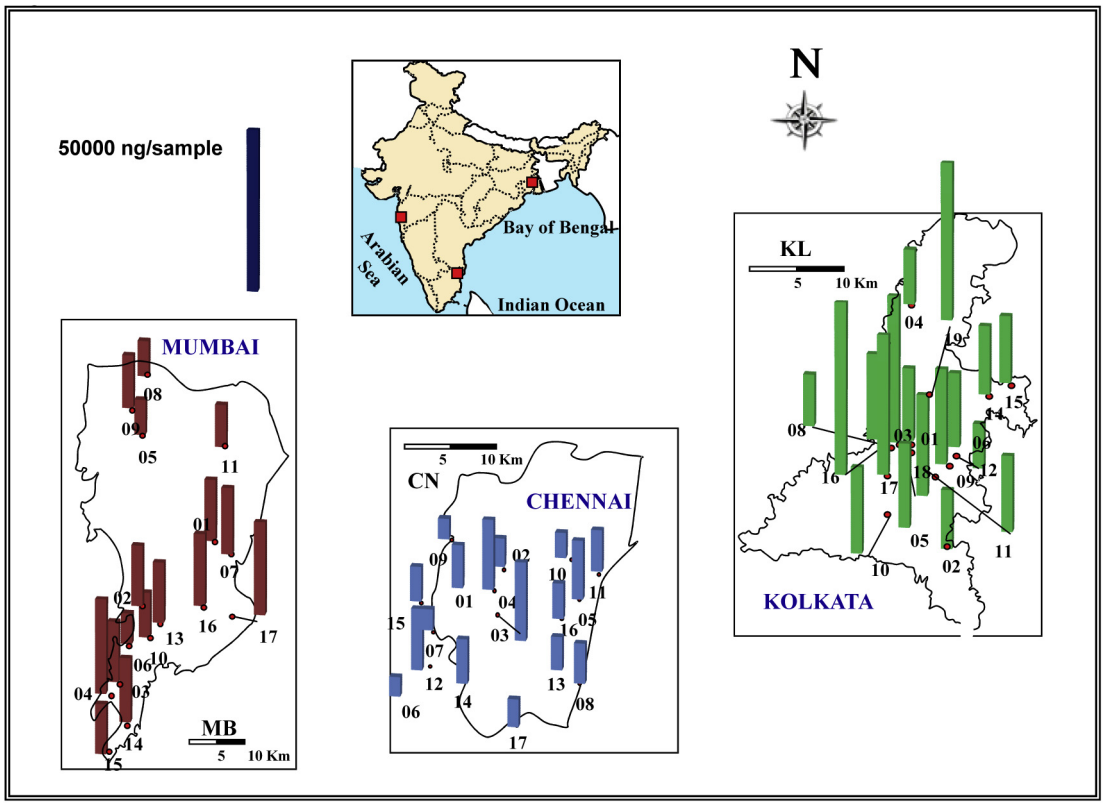

Fig. 1. Total PAH concentrations.

this study, the temperature ranged from 23 °C to 26 °C, so we assumed that the influence of meteorological factors was insignificant on spatial variations. What follows is a comparison made by statistical software (ANOVA by SPSS). Only statistically significant differences ($p < 0.05$) are discussed.

3.2.1. PAHs in Chennai, Kolkata and Mumbai

The total PAH concentrations (ng sample^{-1}) in the three cities are illustrated in Fig. 2. The median concentrations were 11,700, 26,200, and 19,300 ng sample^{-1}, in Chennai, Kolkata and Mumbai, respectively. Generally, most PAHs, excluding Acey, Anth, and DahA, exhibited a spatial trend with high values in Kolkata and low values in Chennai. The HMW PAHs (BaA, Chr, BbkF, BaP, Ind, DahA and BghiP) in Mumbai exceeded those in Chennai as well. The concentrations of some 3-4-ring PAHs, such as Anth, Pyrene, Flur and Chr, were high in Kolkata but low in Mumbai. Thus, the total PAH concentrations in Chennai were the lowest and those in Kolkata were the highest among the three cities. This observation contradicts previous studies, which suggests that the PAH pollution in Mumbai and Kolkata was lower than that in Chennai (Table 1). However, Mumbai is the most populous metropolitan area in India with a population more than double that of Kolkata and Chennai. The fossil fuel energy consumption in the three cities and their adjacent areas exhibited the following spatial trend: Kolkata > Mumbai > Chennai (Reddy and Venkataraman, 2002). It is possible that related PAH emissions in Chennai were relatively low.

On the other hand, the PAH composition profiles in each city were also different. Chennai was characterized by a relatively high proportion of low-molecular-weight (LMW) PAHs (Ace, Acey, Flu, Phe, Ant, Flur and Pyr) and a low proportion of HMW PAHs, while the reverse was observed for Mumbai and Kolkata (Fig. 3). Previous work on atmospheric PAHs in Kolkata and Chennai mainly focused on the particulate phase. The capture efficiency of fine particles by PASs may underestimate HMW PAHs; therefore, a comparison with other work in Kolkata and Chennai cannot be conducted directly. In particle samples collected in Kolkata, the concentrations of some HMW PAHs (BaA, BbkF and BaP) were comparable to Flur and Pyr (Karar and Gupta, 2006). In contrast, Mohanraj et al. (2010) reported that the predominant fine particulate phase PAHs in Chennai were Phe, Anth and BghiP, with maximum levels up to 771.5, 727.7 and 210 ng m^{-3}, respectively. Although the congener profiles were different from the present observation, a comparison between those reports suggests that the proportions of LMW PAHs (Phe and Anth) in Chennai were higher than in Kolkata, which is consistent with our observation to some extent. This pattern could be attributed to the economic and energy structure of the three cities. The major sources of PAHs in Indian cities are fuel combustion, vehicular emissions, industrial contributions and possibly wood burning (Kulkarni and Venkataraman, 2000). The economic base in Chennai is anchored by light and high-tech industry and health care and financial services, while Kolkata and Mumbai mainly rely on traditional industry. The energy consumption suggests that local coal consumption also follows the order Kolkata > Mumbai > Chennai (Garg et al., 2001). Based on the emission factors of diverse sources (Chakraborty et al., 2010), coal consumption and heavy industry introduced a relatively high proportion of HMW PAHs into the environment, thus possibly leading to the observed congener trend in this study.

3.2.2. Urban–rural trend

Another study illustrated an urban–rural trend of airborne pollutants in India, with a decline from urban to rural (Chakraborty et al., 2010). However, we did not find any statistically significant decrease of total PAH concentrations among urban, suburban and rural sites in this study. The urban–rural trend can only be

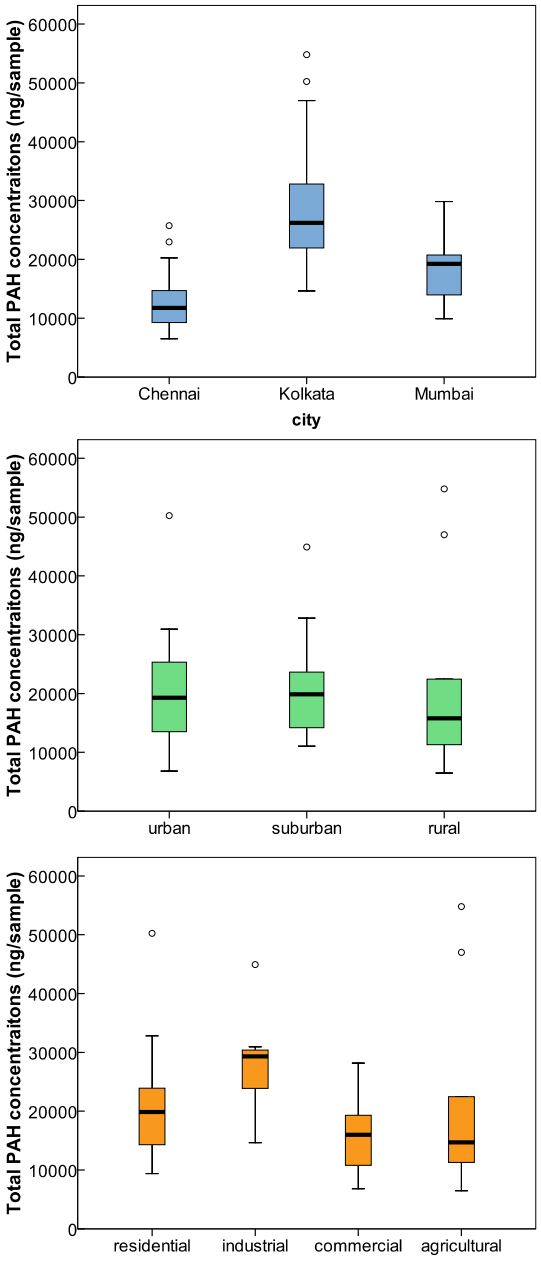

Fig. 2. Spatial pattern of total atmospheric PAHs in this study.

observed in the congener profiles, with an elevated proportion of Phe and decreased proportions of HMW PAHs (BaA, Ind and BghiP) in rural sites (Fig. 3). In India, vehicular traffic and industrial operations are mainly responsible for the atmospheric PAH pollution in the metropolitan areas. Traditional biomass resources are still used to meet an overwhelming proportion of the rural energy demand, possibly explaining the measured PAH concentration comparable to urban regions in rural site (Masih et al., 2012). The two types of sources exhibit specific isomeric ratios if the relative proportions of the species in question are assumed to be conserved between each emission source and the downwind point of measurement (Galarneau, 2008). The PAH source diagnostic ratio of Flur/(Flur + Pyr) ranged from 0.4 to 0.5, while that of Ind/(Ind + BghiP) ranged from 0.4 to 0.7 in this study, possibly indicating the coexistence of petroleum and biomass/coal combustion. The combined effect of these sources explains the composition difference between urban and rural sites and the lack of a significant decrease in the urban–rural concentration. This effect also indicates that the rural sources could be as important as the urban sources in India.

3.2.3. PAHs in residential, industrial, commercial and agricultural sites

The sampling sites in the present study included four types of land use in India: residential, industrial, commercial and agricultural. Statistically significant differences of total PAHs were only found between industrial and commercial areas (Fig. 2). The atmospheric concentrations of Phe and Flur in the industrial sites were significantly higher than those in commercial sites. Because the two compounds are major contributors to the atmospheric PAH burden, the total PAH concentrations exhibited a similar pattern. For other PAHs, such as BaA, Chr, BbkF, BaP, Ind and BghiP, the concentrations in the industrial sites exceeded those in residential, commercial and agricultural sites. Given that these compounds were minor components of the total PAHs, the influence on the total PAHs was insignificant. It is also worth noting that the PAHs in residential sites did not exhibit significantly lower levels than those in other zones. A major segment of the population gathers and lives in residential areas, so there is a potential health risk to the Indian population.

The differences in congener profiles among the four land use types are illustrated in Fig. 3. Industrial sites were deficient in LMW PAHs and enriched in HMW PAHs. The relative proportion of Flu was lower than that of the commercial sites, and that of Phe was lower than that of agricultural sites. Correspondingly, higher percentages of BaA, Chr, BaP, Ind and BghiP were observed at the industrial sites compared with the other sites. As noted earlier, industrial sources release a high proportion of HMW PAHs, confirming the above assumption that industrial emissions resulted in the difference in atmospheric PAH composition between Chennai

Table 1
A comparison of previous study on atmospheric PAHs in India.

Study area	Time	Media	Number of PAHs	Range (ng m^{-3})	Mean (ng m^{-3})	Reference
Mumbai	1996–1997	Particle	7	38.8–24.5		(Kulkarni and Venkataraman, 2000)
Mumbai	2001	Gas + particle	8	4.05–34.42	13	(Sahu et al., 2004)
Mumbai	2001	Gas + particle	7	4.1–30.4	17.4	(Pandit et al., 2006)
Delhi	2002–2003	Particle	12	176.7–1201.3	669.9	(Sharma et al., 2007)
Agra	2005–2006	Particle	18	8–97.9	42.3	(Masih et al., 2010)
Agra	2006–2007	Gas + particle	23	1212.37–3292.28		(Masih et al., 2012)
Agra	2006–2007	Gas + particle	23	879.13–2312.14		(Masih et al., 2012)
Chennai	2009–2010	Particle	11	121.1–1370.5	517.1	(Mohanraj et al., 2010)
Kolkata	2003–2004	Particle	5		96.7	(Karar and Gupta, 2006)
Tiruchirappalli	2009–2010	Particle	9	136–487.5	259.4	(Mohanraj et al., 2011)
Lucknow	2005	Particle	16	0.42–204.17	54.23 (median)	(Singh et al., 2008)

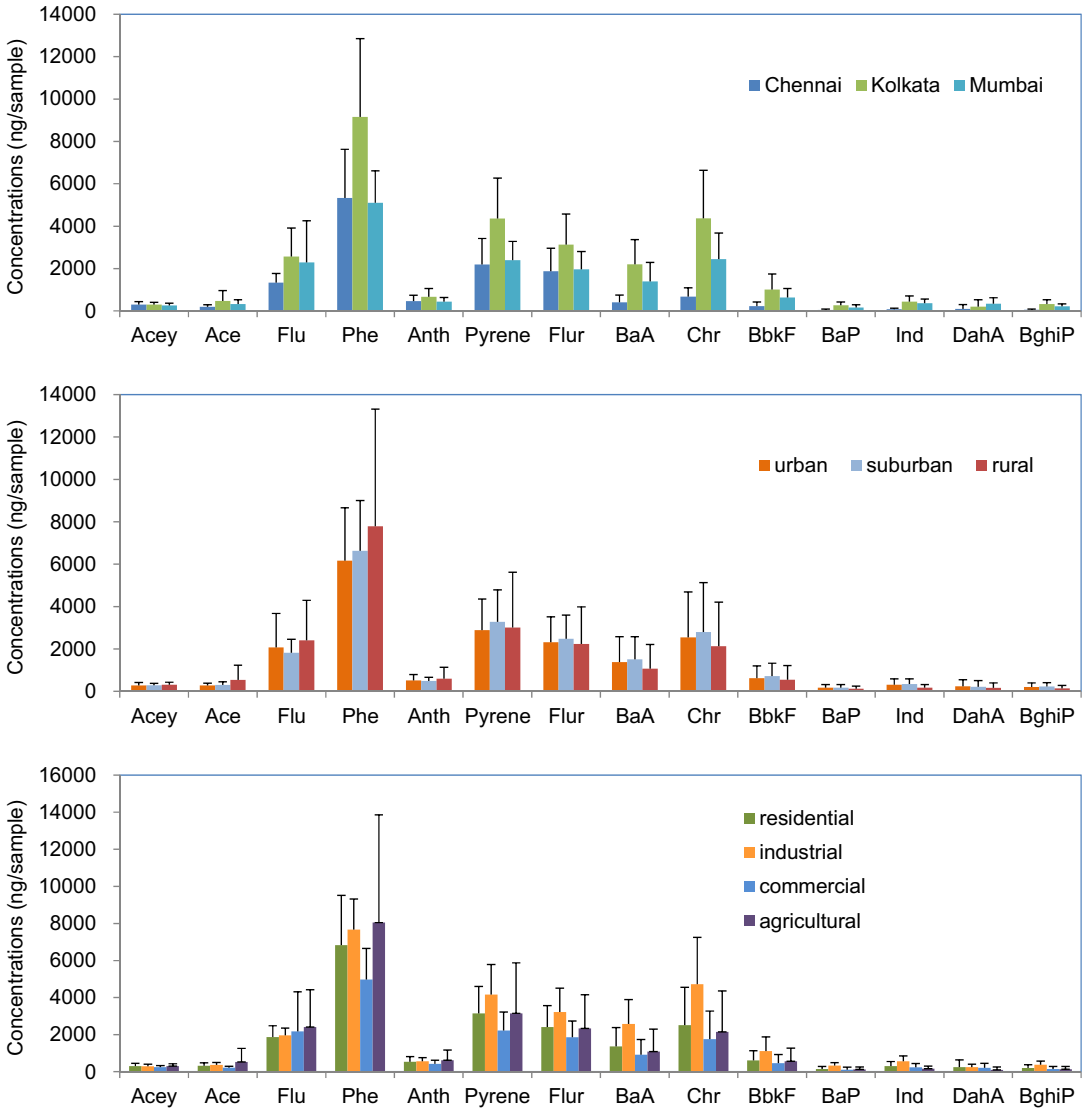

Fig. 3. Congener pattern of PAHs in this study.

and Kolkata and Mumbai. At the agricultural sites, the relative abundances of these HMW PAHs were low, which suggests low human exposure risk when the total PAH concentrations are the same.

3.3. Human exposure

PAS based on PUF samplers has been proved to be a potentially useful tool for evaluating human exposure to PAHs in the atmosphere (Bohlin et al., 2008). WHO (2000) stated that the corresponding concentrations for lifetime exposure to BaP producing excess lifetime cancer risk of 1/10,000 is approximately 1.2 ng m^{-3}. In the present study, the concentrations of BaP have already exceeded this threshold in more than 58% of the samples, even though only part of the HMW PAHs in the air can be captured by PUF disks (Chaemfa et al., 2009).

Given the carcinogenic potential of PAHs, the human risk in each Indian city was explored by multiplying the individual PAH concentrations by their toxic equivalence factors (TEFs). The TEFs values were obtained from Nisbet and Lagoy (1992). The calculated total BaP_{eq} values are presented in Fig. 4. Following the spatial trend of total PAH concentrations, the average BaP_{eq} exposure profile in Chennai was significantly lower ($p < 0.05$) than that in Kolkata and Mumbai. The only two sites with values lower than 1 ng m^{-3} in this study were also located in Chennai. The highest value was found at an urban residential site (KL-19) in Kolkata, with a value of

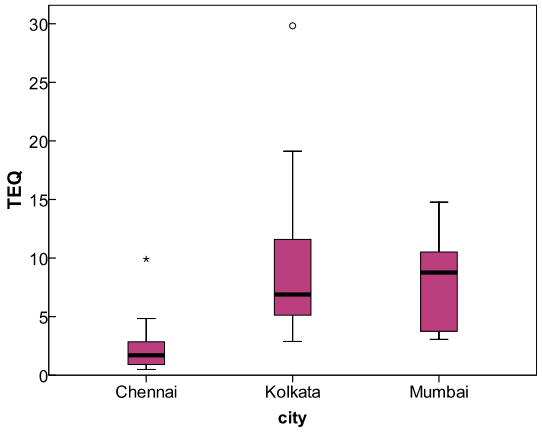

Fig. 4. Total BaP_{eq} values in Chennai, Kolkata and Mumbai.

29 ng m^{-3}. The human exposure risk was at the same level among the urban, suburban and rural sites. In contrast, the average BaP_{eq} concentrations in the industrial areas were significantly higher ($p < 0.05$) than those in the commercial or agricultural areas, highlighting the potential adverse effects from industrial sources in India.

Although LMW PAHs were the dominant in the air, their human exposure can be ignored according to this evaluation. BaA, BbkF, BaP and DahA were identified as the major atmospheric contributors, representing > 80% of the total BaP_{eq}. The BaP contribution was lower than the evaluation in Agra, India (Masih et al., 2012), which can be attributed to the spatial variation and the difference between AAS and PAS.

4. Conclusion

This study applied the PUF-PAS method to describe the spatial pattern of atmospheric PAHs in three Indian major cities during the wintertime of 2006. In general, it captured the spatial and compositional pattern of PAHs, even for some HMW PAHs, which are usually difficult to capture by PUF-PAS sampling. The measured concentration levels are within the highest range in the world. Spatially, the PAH concentrations in Chennai were the lowest and those in Kolkata were the highest among the three cities. Unlike other regions in the world, there is no significant decrease in PAH concentrations among urban, suburban and rural sites. The rural sources, associated with traditional biomass combustion, could be as important as the urban sources in India. If categorized by land use type, some HMW PAH concentrations in the industrial sites exceeded those in the residential, commercial and agricultural sites. The industrial sites were deficient in LMW PAHs and enriched in HMW PAHs. In this study, the air pollution caused by PAHs exceeded the criterion of 1 ng m^{-3} BaP_{eq} at >90% of the sites.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.atmosenv.2013.03.001.

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