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Polycyclic aromatic hydrocarbons on indoor/outdoor glass window surfaces in Guangzhou and Hong Kong, south China

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ABSTRACT

Organic films were collected from indoor and outdoor window surfaces in two large cities in southern China, Guangzhou and Hong Kong, and analyzed to quantify the polycyclic aromatic hydrocarbons (PAHs). In the glass films, the highest concentration of total PAHs, predominantly phenanthrene, fluorene, fluoranthene, and pyrene, was found to be l400 ng/m^2. The concentrations of PAHs in Guangzhou were usually higher than those in Hong Kong. In general, higher concentrations of PAHs on exterior window films in comparison with interior window films in both cities indicated that the outdoor air acted as a major source of pollution to the indoor environment. However, indoor air was a major source of some light-weight PAHs. Measurements made over time indicated that the growth rates of light-weight PAHs on window surfaces were fast at the beginning and then gradually reached a consistent level, whereas heavy-weight PAHs exhibited near-linear accumulation during the 40 days sampling period.

1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) of natural and anthropogenic origins are one of the major carcinogenic constituents of the atmosphere. They have been of concern for several decades due to their ubiquity in the environment and adverse effects on human beings. Atmospheric PAH concentrations vary widely in different locations. The highest levels often occur in densely populated and industrialized areas. The rapid socio-economic development in China during the past three decades has accelerated the urbanization process in many regions and produced a number of mega-cities. As one of the most developed regions in China, the Pearl River Delta (PRD) in the southern part of China, which includes many mega-cities such as Hong Kong, Guangzhou, and Shenzhen, has been affected by PAH pollution in the ambient air environment (Fu et al., 2003; Li et al., 2006; Liu et al., 2006).

Urban areas are characterized by a high proportion of impervious surfaces, such as windows, walls, and roadways. A thin organic film could develop on urban impervious surfaces from the condensation of air contaminants. The few existing studies on this topic show that organic films can range from 11 to 1000 nm in thickness and that the proportion of organic matter ranges from approximately 5–20% (Diamond et al., 2000; Liu et al., 2003b; Wu et al., 2008). Due to the high proportion of organic content of the film, the concentrations of semi-volatile organic compounds (SVOCs) are high on the artificial surfaces of urban areas (Butt et al., 2004; Gingrich et al., 2001; Liu et al., 2003a). SVOCs can accumulate in organic films through the exchange and partitioning of gas-phase chemicals with air and/or the direct deposition of particle-phase compounds (Diamond et al., 2000; Law and Diamond, 1998; Liu et al., 2003b). Thus, organic films could affect the surface-air exchange and the residence time of SVOCs in urban environments (Priemer and Diamond, 2002). An investigation of the amount and distribution of PAHs on urban impervious surfaces can help clarify the environmental behavior and conditions of PAHs in cities.

A recent study indicated that organic films could develop at a consistent rate on glass bead surfaces and that polychlorinated biphenyls (PCBs) also increased linearly at a rate similar to the surface films (Wu et al., 2008). Our previous study also showed that the growth rates of organic films on the window surfaces were fast at the beginning and then tended to be consistent and that the evolution rates ranged from 2.55 to 10.9 nm/day for “bulk film” and from 0.06 to 0.92 nm/day for “pure film” (Li et al., 2010a). On the other hand, organic film might provide a good adsorbent surface for

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the photo-degradation of pollutants (Butt et al., 2004). For example, the concentrations of polybrominated diphenyl ethers (PBDEs) on glass window surfaces did not increase with the number of growth days in a previous study (Li et al., 2010a). To evaluate the influence of photo-degradation on impervious surfaces on the chemical dynamics and mass balances of PAHs, it is important to understand how PAHs accumulate during the film evolution. The aims of the present study were the following: (1) to measure the concentrations and distributions of PAHs in glass window films in Guangzhou and Hong Kong, (2) to compare the concentrations and profiles of PAHs in indoor and outdoor films, and (3) to investigate the accumulation of PAHs throughout the film growth stage.

2. Materials and methods

2.1. Collection of organic films

The sampling sites and the window film sampling method have been described in a previous study (Li et al., 2010a). Briefly, from Dec 2007 to Jan 2008, six paired (indoor/outdoor glass film) samples in Guangzhou were collected from one office and one residential house on the Sun Yat-sen (Zhongshan) University campus, one office and one residential house on the campus of the South China University of Technology, and one office and one residential house at the Guangzhou Institute of Geochemistry; nine paired samples in Hong Kong were collected from three offices and one residential house on the University of Hong Kong campus, three offices on the Hong Kong Polytechnic University campus, and two residential buildings in Kowloon, Hong Kong. All of the sampling sites were located in urban areas. South-facing windows in the office buildings and living rooms of residential buildings between the 3rd and 6th floors were chosen. In the PRD region, the dry season lasts from late October to the following March, and the weather is relatively cool (average temperature: 16.5 ± 3.8 °C) and dry (average RH: 59.1 ± 13.1%). Therefore, sampling during this season limits the frequency of film wash-off by heavy rain. The windows selected for sampling had not been cleaned for at least three months.

To evaluate the accumulation of PAHs on the window glass surfaces, an additional field sampling campaign was conducted at the Guangzhou Institute of Geochemistry in downtown Guangzhou, where samples were collected from the same windows over a period of time. From a four-floor building, one row of windows facing south and another row of windows facing north were chosen. Before sampling, the chosen window glass surfaces were cleaned using distilled water, acetone and dichloromethane (DCM). Sampling was conducted on Day 2, 5, 9, 16, and 40. At each sampling time, duplicate samples from both the south and north window glass surfaces were collected (a total of 6 paired duplicate samples were analyzed for PAHs).

The window glass was sampled with clean, dry lab tissues (Kimwipes, Kimberly Clark, USA) and subsequently with DCM wetted lab tissues. Lab tissues were pre-cleaned by extraction with DCM, air-dried, and stored in glass jars. Windows were sampled to within 10 cm of the window edge to minimize contamination from exterior caulking or paint, and approximately 1–2 m^2 of window surface was sampled at each site. Field blank preparation was identical to the sample preparation. Field blank lab tissues did not make contact with the window surfaces. Samples and field blanks were immediately immersed in DCM in air-tight 40 mL amber vials after sampling. The total mass of material collected from the windows was determined by the duplicated weighing of desiccated paper tissues before and after sampling. To reduce the effect of humidity, the paper tissues were desiccated using 900 mL of desiccant for 24 h. All samples were then stored in a freezer at −20 °C.

2.2. Extraction and analysis

A mixture of surrogate standards of acenapthene-d_{10}, phenanthrene-d_{10}, chrysene-d_{12} and perylene-d_{12} were added to each sample prior to extraction. All the samples were sonicated 3 times with DCM. The three extracts were combined, concentrated and solvent-exchanged to hexane. A portion (20%) of each sample was accomplished by an 8 mm i.d. alumina/silica column in turn containing anhydrous sodium sulphate (1 cm), neutral silica gel (3 cm, 3% deactivated) and neutral alumina (3 cm, 3% deactivated). The fractions of PAHs were then eluted with 15 mL of a mixture of dichloromethane and hexane (1:1, V/V). The eluent solvent was blown down to a final volume of 25 μL in dodecane under a gentle stream of nitrogen. Prior to analysis, 1000 ng of hexamethyl benzene was added as an internal standard.

The PAHs were analyzed 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). Samples (1 μL) were injected under the splitless mode with a 10 min solvent delay time. High purity helium was used as a carrier gas with a flow velocity of 1.83 mL/min. The temperatures of the injector and transfer lines were 290 °C and 300 °C, respectively. The initial oven temperature was set at 60 °C for 1 min and raised to 290 °C at a rate of 3 °C/min and held for 20 min. Fifteen individual PAHs were quantified: acenapthene (Ace), acenapthylene (Acey), fluorene (Flu), phenanthrene (Phe), anthracene (Ant), fluoranthene (Flur), pyrene (Pyr), benzo[a]anthracene (BaA), chrysene (Chr), benzo[b]fluoranthene (BbF), benzo[k]fluoranthene (BkF), 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. Quality assurance and quality control (QA/QC)

Quality assurance and quality control were performed using method blanks and field blanks. The PAHs found in the field blanks were generally below the instrumental detection limits (IDLs) (the amount of analyte that would generate a signal-to-noise ratio of 3:1). The method detection limits (MDLs) were assigned to be 3 times the IDLs. The method blanks showed no detectable PAHs. The mean percent recoveries were 88 ± 9%, 92 ± 11%, 90 ± 9% and 88 ± 9% for acenapthene-d_{10}, phenanthrene-d_{10}, chrysene-d_{12} and perylene-d_{12}, respectively. The final concentrations of PAHs were not corrected for the recoveries.

3. Results and discussion

3.1. PAH concentrations and sources in Guangzhou and Hong Kong

The PAH concentrations in window films can be presented in two ways, either as an area-normalized or a mass-normalized concentration. Area-normalized concentrations determine the total concentrations of PAHs in the environment, whereas mass-normalization shows the PAH amount in relation to the organic window film available (Unger and Gustafsson, 2008).

The area-normalized concentrations of PAHs in the glass films in the two cities are summarized in Table 1 and plotted in Fig. 1. The concentrations of total PAHs in Guangzhou, which ranged from 410 to 1400 ng/m^2 with a GM value of 990 ng/m^2, were significantly higher ($p = 0.001$) than those in Hong Kong, which ranged from 71 to 630 ng/m^2 (GM: 230 ng/m^2). The area-normalized concentrations show that there are more PAHs in the air of Guangzhou than of Hong Kong. Similar distribution patterns of the PAHs in Guangzhou and Hong Kong were also observed in air (Guo et al., 2003; Li et al., 2006) and in atmospheric depositions (Li et al., 2010b).

Organic matter in the window films has formed through the condensation of gas-phase species and the deposition of carbonaceous particles. Generally, the SVOC concentrations in glass films were controlled by the organic matter content (Butt et al., 2004; Unger and Gustafsson, 2008). A detailed organic matter collection method and the organic matter concentrations were described in a previous study (Li et al., 2010a). Briefly, the concentrations of total organic matter (TOM) in Guangzhou ranged from 3600 to 18,200 μg/m^2 (GM: 7900 μg/m^2), which accounted for 0.76–2.91% (GM: 1.43%) of the film mass concentrations. In Hong Kong, the corresponding values ranged from 1240 to 10,600 μg/m^{-2} (GM: 5000 μg/m^2) and accounted for 0.65–9.51% (GM: 2.47%) of the film mass concentrations. Normalizing the PAH concentration to the amount of TOM (g_{TOM}) of the window films shows that the samples in Guangzhou varied between 78 and 220 μg/g_{TOM} with a GM of 120 μg/g_{TOM} and that the samples in Hong Kong varied between 17 and 180 μg/g_{TOM} with a GM of 45 μg/g_{TOM}. In ambient air, the concentrations of total PAHs in Guangzhou were also significantly higher ($p = 0.001$) than those in Hong Kong (Bi et al., 2003; Guo et al., 2003). However, the mass-normalized concentration ratios (120/45 = 2.66) between Guangzhou and Hong Kong were only half those of the area-normalized concentrations (990/230 = 4.30). This result suggested that the PAH concentrations in glass films were also influenced by the content of organic matter (Butt et al., 2004; Unger and Gustafsson, 2008).

Flu, Phe, Flur, and Pyr were the most abundant PAHs in all samples. In addition, the relative contribution of BghiP in Hong Kong was higher than that in Guangzhou (Fig. 2). BghiP is considered to be linked to vehicle emissions (Katsoyiannis et al., 2011;

Table 1
Geometric mean (GM) and concentration ranges of measured PAHs in glass window films (ng/m^2).

ng/m^2	Guangzhou				Hong Kong			
	Indoor		Outdoor		Indoor		Outdoor	
	GM	Range	GM	Range	GM	Range	GM	Range
Ace	18	10–25	27	20–32	4.0	1.6–6.6	6.8	4.1–11
Acey	26	13–36	30	20–38	5.0	2.5–9.8	7.4	3.9–15
Flu	110	62–140	130	100–160	19	9.8–37	24	12–47
Phe	280	160–380	380	320–440	55	32–100	88	57–140
Ant	23	12–32	32	27–36	4.9	2.3–9.8	8.1	5.1–13
Flur	53	28–81	140	88–200	11	5.9–28	48	23–110
Pyr	60	31–88	130	93–180	12	5.8–27	49	22–120
BaA	21	10–42	58	35–74	2.6	1.3–7.9	12	5.8–23
Chr	35	14–66	100	52–130	3.6	1.7–15	24	11–39
BbF	52	25–90	94	54–120	3.5	1.2–16	23	12–37
BkF	14	8.6–26	28	18–33	1.4	0.7–5.6	7.9	3.9–15
BaP	13	6.8–26	32	17–41	1.6	0.7–6.3	11	6.8–26
Ind	21	13–38	40	22–57	2.5	0.7–11	17	10–32
DahA	2.6	0.8–5.3	8.8	4.8–13	0.8	0.3–2.9	4.9	2.5–10
BghiP	19	12–35	41	20–62	3.1	0.8–14	34	19–77
Σ_{15}PAHs	760	410–1100	1300	1000–1400	140	71–260	380	210–630

Miguel and Pereira, 1989). In Hong Kong, the streets are generally narrow, and the buildings are close to the roadside; the higher contributions of BghiP suggest that the major source of PAHs on the window surfaces might be local vehicle emissions. Generally, the patterns of individual PAH concentrations in this study were consistent with those of previously reported film samples (Gingrich et al., 2001; Unger and Gustafsson, 2008). The concentration pattern at all sampling sites showed an increase in the predominantly light-weight PAHs (Flu, Phe, Flur, and Pyr) in comparison with urban air particulate samples as well as a greater abundance of heavy-weight PAHs, such as BbF, BkF, BaP, Ind, DahA, and BghiP, than in the total urban air (gas plus particle phases) (Li et al., 2006). Our previous study indicated that the organic film quickly accumulated from the beginning as a result of the condensation of gas-phase organic matter and then gradually grew as a result of particle deposition with a near-linear accumulation and continual gas-phase condensation on the window surface (Li et al., 2010a). Heavy-weight PAHs are mainly associated with the particulate phase. The particle-bound PAHs would accumulate on the window surface through deposition and adsorption, although some of them might be photo-degraded by sunlight. For the light-weight PAHs, only a small amount of them would accumulate by particle deposition, and most of them would remain in a balanced proportion between the organic film on the window surface and the air (Unger and Gustafsson, 2008). The proportion of particle-bound PAHs on the window surface would increase throughout the organic film growth process.

Comparing the PAH concentrations measured in this study with the limited observations of PAH levels in glass films, the area-normalized PAH concentrations in Hong Kong and Guangzhou were much lower than those reported in downtown Toronto, Canada (Gingrich et al., 2001). PAH concentrations in Guangzhou were found to be similar to those in the suburban areas of Toronto, and the concentrations in Hong Kong were similar to those in the rural areas of Toronto (Gingrich et al., 2001). Both the area-normalized and mass-normalized PAH concentrations in Hong Kong were comparable to those in the city center and suburban areas of Stockholm (Unger and Gustafsson, 2008).

3.2. Indoor/outdoor comparison

A comparison of PAHs in the interior and exterior films on a site-by-site basis is shown in Fig. 1. The concentrations of area-normalized PAHs in indoor and outdoor glass films in the two cities are summarized in Table 1. In Guangzhou, the concentrations of total PAHs on the indoor window surfaces ranged from

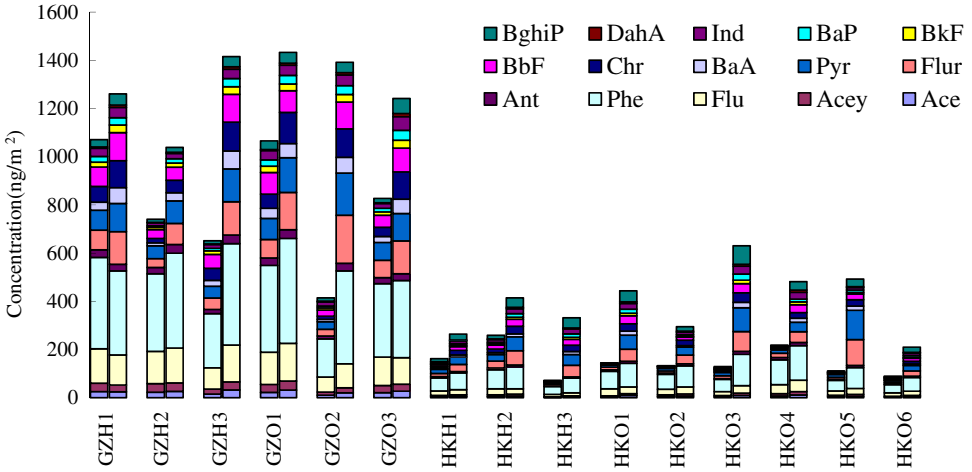

Fig. 1. PAH concentrations and compositions on the window glass surfaces in Guangzhou and Hong Kong. The left columns are indoors and the right columns are outdoors (GZH1 denotes Guangzhou-House 1; HKO1 denotes Hong Kong-Office 1).

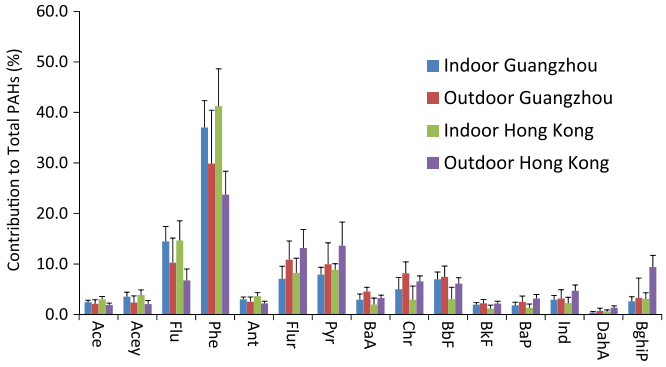

Fig. 2. PAH compositions on the exterior and interior window surfaces in Guangzhou and Hong Kong.

410 to 1100 ng/m^2 with a GM value of 760 ng/m^2, whereas those on the outdoor window surfaces were in the range of 1000–1400 ng/m^2 (GM: 1290). In Hong Kong, the concentrations of total PAHs on the indoor window surfaces ranged from 71 to 260 ng/m^2 with a GM value of 140 ng/m^2, whereas those on the outdoor window surfaces were in the range of 210–630 ng/m^2 (GM: 380). Clearly, the area-normalized total PAH concentrations in the interior films were generally lower than those in the exterior films in both cities (Fig. 1). However, the ratios of the interior/exterior concentrations varied for individual PAH compounds. For the light-weight PAHs, such as Acey, Flu, Phe, and Ant, the interior/exterior ratios were close to 1.0, and, in some cases, the ratios of these compounds were higher than 1.0. For the heavy-weight PAHs, the ratios were generally less than 1, especially in Hong Kong, with values of less than 0.5 (Fig. 3). The difference in the PAH concentrations and profiles between the interior and exterior films might be attributed to the different PAH composition patterns of the PAH sources in the indoor air and outdoor air environments. For example, a previous study indicated that the abundance of 3-ring PAHs could be related to the cooking practices in China (Zhu and Wang, 2003), and the heavy-weight PAHs are mainly associated with high-temperature combustion, such as vehicle exhaust emissions from the outdoors (Khalili et al., 1995). The building ventilation and physico-chemical properties of PAHs are other factors that control the PAH distribution patterns in indoor and outdoor glass films. For example, most buildings in Hong Kong were equipped with central air-conditioning systems and the air was filtered through the systems from outside, whereas, most building in Guangzhou were not equipped this system. Therefore, for the light-weight PAHs, exchanges between the indoor and outdoor air are not influenced by the filtering system. However, for the particle-associated (heavy-weight) PAHs, most of them might be filtered out by the central air-conditioning intake system. This may result in relatively lower concentrations of heavy-weight PAHs in the interior films than in the exterior films, such as those in Hong Kong.

A comparison of mass-normalized PAHs in indoor and outdoor glass films from the two cities is displayed in Fig. 3. In the most case, the indoor window surfaces had less heavy-weight PAHs than the outdoor windows, indicating the heavy-weight PAHs originated predominantly from the outdoor air. However, for the light-weight PAHs, such as Acey, Flu, Phe, and Ant, the interior/exterior ratios were greater than 1.0 in some cases. Based on the PAH concentrations in the window films and the partition coefficient of window film-air (Unger and Gustafsson, 2008), more light-weight PAHs appeared in the indoor air than in the outdoor air.

3.3. Accumulation of organic film and PAHs

The accumulation characteristics of organic films and PAHs on window glass were studied at one sampling location in Guangzhou. The time series of the growth of several PAH compounds and the total PAH growth on the window surface are displayed in Fig. 4. Generally, PAH growth patterns over time on the windows facing north and south showed a near-linear accumulation similar to that of PCBs in a previous study

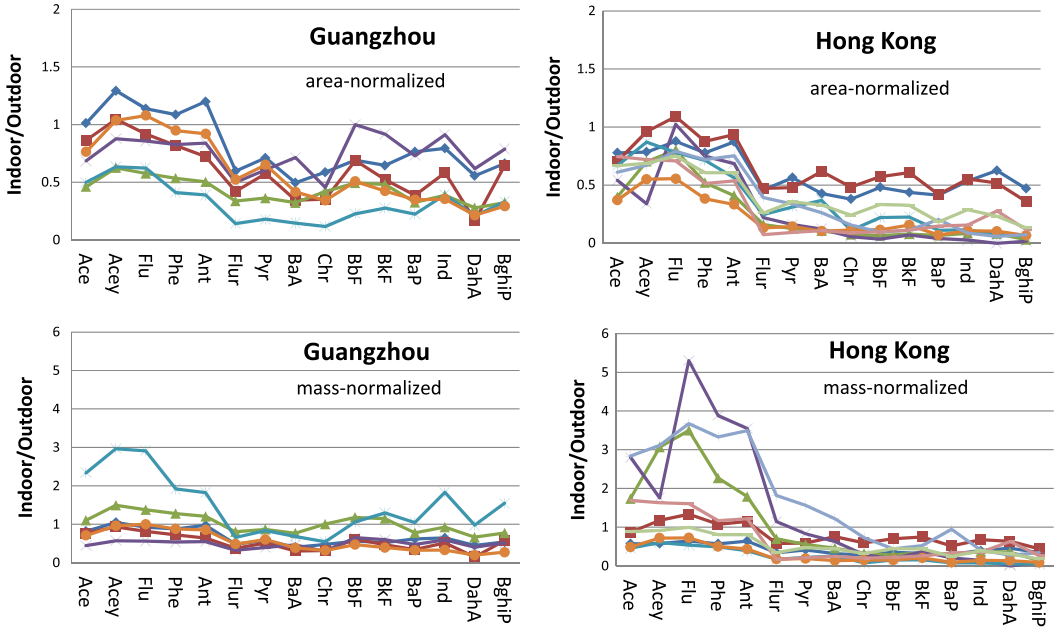

Fig. 3. The indoor/outdoor ratios of PAHs on the window glass surfaces in Guangzhou and Hong Kong (the different colour lines represent the different sample sites). Area-normalized concentrations determine the total concentrations of PAHs in the environment, whereas mass-normalization shows the PAH amount in relation to the organic window film available. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

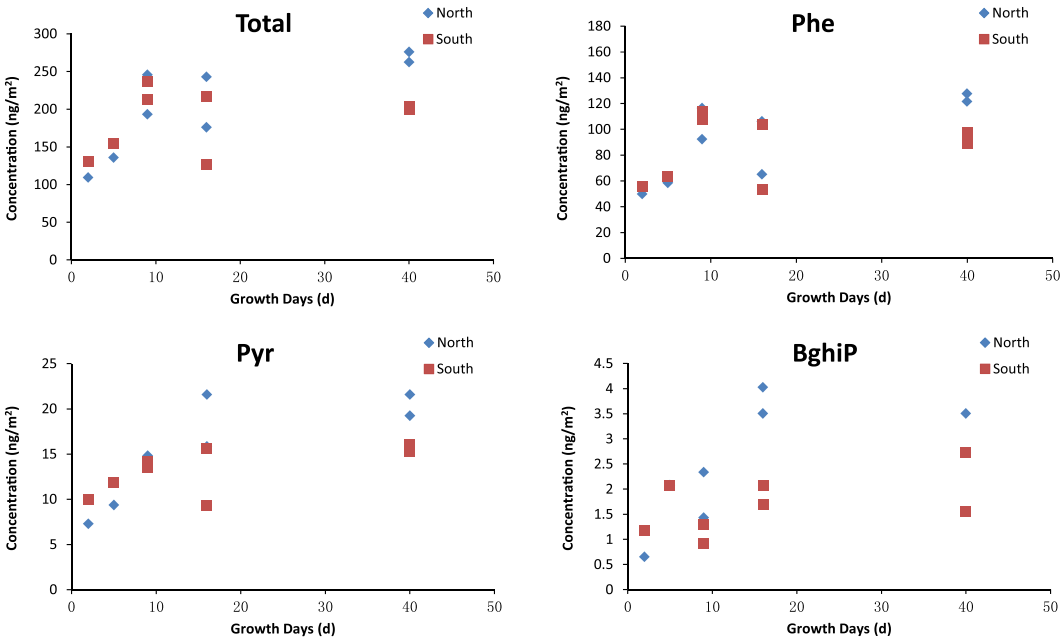

Fig. 4. Time series of PAHs on the south and north window surfaces in Guangzhou.

(Wu et al., 2008). For windows on both sides, the variation in accumulation characteristics of the individual PAH compounds was observed. The light-weight PAHs (Ace, Acey, Flu, Phe, and Ant) showed similar accumulation trends of fast growth at the beginning and then reaching a consistent level afterwards. Significant correlations ($r > 0.88$, $p < 0.004$) were found among those light-weight PAHs. The amount of light-weight PAHs in the organic films was mainly affected by the air concentrations and the organic matter mass. These compounds could reach equilibrium between the air and the organic matter on the window surfaces. The heavy-weight PAHs (BbF, BkF, BaP, Ind, and BghiP) correlated significantly ($r > 0.85$, $p < 0.05$) with each other. The accumulation trends of these compounds exhibited near-linear patterns initially. The window film is formed not only by the condensation of gas-phase organic matter, which acts as a passive partitioning medium for vapor-phase PAHs, but also by the accumulation of urban aerosols (Butt et al., 2004; Unger and Gustafsson, 2008). Urban aerosols contain combustion particles, including organic carbon (OC) and black carbon (BC). Heavy-weight PAHs are known to be efficiently adsorbed to BC (Chen et al., 2005). Therefore, the light-weight PAHs should have a positive correlation with the condensation of organic matter, whereas the heavy-weight PAHs should positively correlate with the BC on the window surfaces. The time series of BC exhibited a near-linear accumulation pattern throughout, whereas the growth of OC from gas-phase condensation (OC_{con}) showed a fast rate of increase initially and then increased at a gradual rate on the windows on both sides (Li et al., 2010a). There were no significant correlations between all PAHs and the organic matter on the windows. However, positive correlations between most of the PAH compounds and organic matter were observed. This result suggests that the accumulation of PAHs in the window films was partly controlled by the organic matter, but the window film might not be an ideal medium for the quantitative prediction of PAHs in the air (Unger and Gustafsson, 2008). The photo-degradation of PAHs on the window surface might be another possible factor. For example, DahA, which is more prone to photo-degradation than other heavy-weight PAHs, did not show any correlation with other heavy-weight PAHs.

4. Conclusions

High concentrations of PAHs were observed in organic films on interior and exterior window surfaces sampled in Guangzhou and Hong Kong. The concentrations of PAHs in Guangzhou were usually higher than those in Hong Kong. In general, higher concentrations of PAHs on exterior window films than on interior window films in both cities indicated that the outdoor air acted as a major source of pollution to the indoor air. However, for some light-weight PAHs, indoor air might be a potential pollution source. In addition, the building ventilation and the physico-chemical properties of PAHs might be other factors controlling the PAH distribution patterns in indoor and outdoor glass films. Measurements made over time indicated that the growth rates of light-weight PAHs on the window surfaces were initially fast and then gradually reached a consistent level, whereas the heavy-weight PAHs exhibited a near-linear accumulation pattern throughout the 40 days sampling time.

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