Assessment of airborne polycyclic aromatic hydrocarbons in a megacity of South China: Spatiotemporal variability, indoor-outdoor interplay and potential human health risk[☆]

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ABSTRACT

Although a number of studies have assessed the occurrence of atmospheric polycyclic aromatic hydrocarbons (PAHs) in indoor environment, few studies have systemically examined the indoor-outdoor interplay of size-dependent particulate PAHs and potential health risk based on daily lifestyles. In the present study, size-dependent particle and gaseous samples were collected both indoors and outdoors within selected schools, offices and residences located in three districts of Guangzhou, China with different urbanization levels during the dry and wet weather seasons. Results from measurements of PAHs showed that higher total PAH concentrations occurred in residential areas than in other settings and in indoor than in outdoor environments. Compositional profiles and size distribution patterns of particle-bound PAHs were similar indoors and outdoors, predominated by 4-and 5-ring PAHs and the 0.56−1.0 μm particle fraction. Statistical analyses indicated that outdoor sources may have contributed to 38−99% and 62−100% of the variations for indoor particle-bound and gaseous PAH concentrations, respectively. Incremental life cancer risk (*ILCR*) from human exposure to indoor and outdoor PAHs based on different lifestyles followed the order of adults > children > adolescents > seniors. All average *ILCR* values for four age groups were below the lower limit of the Safe Acceptable Range (10^{-6}). In addition, the *ILCR* value for adults (average: 7.2×10^{-7}; 95% CI: 5.4×10^{-8}–2.5×10^{-6}), estimated from outdoor air PAH levels with 24-h exposure time, was significantly higher than our assessment results (average: 5.9×10^{-7}; 95% CI: 6.3×10^{-8}–1.9×10^{-6}), suggesting the significance of assessing human inhalation exposure risks of indoor and outdoor PAHs in urban air based on daily lifestyles.

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1. Introduction

Environmental quality has been deteriorating during the last few decades, partly due to the manufacture and consumption of increased amounts of chemicals and inappropriate disposal of obsolete consumer products. Growing concerns have been raised about the levels of hazardous contaminants constantly emitted from human activities and related harmful effects in daily life (Destaillats et al., 2008; Rivas et al., 2014; Williams et al., 2000). A great example is air pollution within indoor settings, as the general population spends approximately 90% of the time indoors and therefore is more exposed to pollution indoors than outdoors (Klepeis et al., 2001). The situation is even worse for certain vulnerable groups such as the elderly, children and the sick. Another issue is how much outdoor pollution contributes to indoor loadings and vice versa, which has not been adequately addressed.

Although indoor pollutants are complex and diversified, polycyclic aromatic hydrocarbons (PAHs) have remained the most abundant and therefore are most relevant for human health effects (Kim et al., 2013). They are mainly generated from incomplete

combustion of carbonaceous materials from natural and anthropogenic sources, including industrial, traffic and residential emissions (Kim et al., 2012; Liu et al., 2001). The amount of PAHs emitted from China was 106 Gg in 2007, accounting for 21% of the total global amount, of which 62% was from residential activities (Chen et al., 2017; Shen et al., 2013). Atmospheric PAHs are often a complex mixture distributed between the particle and gaseous phases and can cause bronchitis, asthma, lung cancer and atherosclerosis in human through inhalation exposure (Farmer et al., 2014).

Despite numerous efforts in evaluating indoor and outdoor airborne PAHs and associated human health risks, there is a paucity of the systemic assessment with daily life scenarios in current datasets. We compiled 457 publications (data from Web of Science) on the occurrence of and human exposure to indoor atmospheric PAHs from 2007 to 2017, 20% of which focused on residential air PAHs levels. Besides, only one study was conducted to determine atmospheric PAH concentrations in multiple indoor microenvironments such as schools, homes, offices, hospitals, cars and buses (Romagnoli et al., 2014). As the event time duration in a specific exposure scenario is a key input parameter, assessments based on a single indoor microenvironment may underestimate or overestimate the potential health risk. Although particle-bound PAHs are the major contributor to inhalation exposure to airborne PAHs in indoor environment (Delgado-Saborit et al., 2011). Size-dependent exposure to particle-bound PAHs has not been well characterized because total particles were often collected by high-volume air samplers (Ellickson et al., 2017; Guo et al., 2003; Shi et al., 2003). To our best knowledge, only our previous study (Zhang et al., 2012a) has evaluated inhalation exposure to size-fractionated particle-bound PAHs in indoor environments in Guangzhou, but the sampling size was limited.

Amid the above-mentioned knowledge gap, the present study was conducted to simultaneously evaluate the occurrence of indoor and outdoor airborne PAHs and human health risk from inhalation exposure to PAHs. Three typical types of urban settings (school, office and residence) within three districts with different urbanization levels in Guangzhou of China were selected for field sampling. The spatial and temporal distribution of airborne PAHs, infiltration of outdoor pollutants to indoor loadings, particle size distribution patterns and human health risk from inhalation exposure to PAHs were examined. We also explored the influences of human activities, such as cooking and smoking, and haze conditions on the distribution patterns and levels of indoor PAHs. The results are expected to provide updated information for the public and regulatory agencies in implementing better measures to improve personal environments.

2. Materials and methods

2.1. Materials

A standard solution of 16 PAHs, including acenaphthene (Ace), fluorene (Flo), phenanthrene (Phe), anthracene (Ant), fluoranthene (Fla), pyrene (Pyr), benz[a]anthracene (B[a]A), chrysene (Chr), benzo[b]fluoranthene (B[b]F), benzo[k]fluoranthene (B[k]F), benzo[e]pyrene (B[e]P), benzo[a]pyrene (B[a]P), indeno[1,2,3-cd]pyrene (IcdP), dibenzo[a,h]anthracene (DahA), benzo[ghi]perylene (BghiP) and coronene (Cor), was obtained from AccuStandard (New Haven, CT, USA). Acenaphthene-d_{10}, phenanthrene-d_{10}, chrysene-d_{12}, perylene-d_{12}, benzo[ghi]perylene-d_{12} and coronene-d_{12} used as surrogate standards were obtained from Dr. Ehrenstorfer GmbH (Augsburg, Germany). Fluorene-d_{10} and pyrene-d_{10} were purchased from AccuStandard (New Haven, CT, USA) and dibenzo[a,h]anthracene-d_{14} was from Cambridge Isotope Laboratories (Andover, MA, USA); they were used as internal standards.

2.2. Sample collection

Nine school, office and residence sites, located at the districts of Tianhe, Luogang and Zengcheng representative of substantial, moderate and light urbanization, respectively, were selected for sample collection. Detailed procedures about the sample collection are presented in Text S1 ("S" indicates texts, tables and figures in the Supplementary material afterwards). A total of 550 particles samples (50 samples containing 11 size fractions) and 50 gaseous samples, including duplicate samples, were collected with a Micro-Orifice Uniform Deposit Impactor (MOUDI; MSP Corporation, Shoreview, MN, USA) connected to a stainless steel unit containing polyurethane foam (PUF). Gaseous samples (PUFs) were sealed consecutively with plastic zipper closed bags and lightly vacuum plastic bags, and particle samples were kept in membrane cell holders immediately after sampling. All samples were cooled with ice during transport to the laboratory and were stored at −20 °C until analysis.

2.3. Sample extraction and instrumental analysis

Both glass fiber filters (47 mm diameter and 1.6 μm aperture) and PUFs loaded with particulate and gaseous PAHs, respectively, were spiked with the surrogate standards before extraction. Loaded filters were sonicated three times with 20 mL of hexane, dichloromethane and acetone mixture (2:2:1 in volume) and PUFs were Soxhlet-extracted for 48 h with 200 mL of the same mixed solvent. Each extract was concentrated, solvent-exchanged to hexane and further concentrated with a Zymark TurboVap 500 (Hopkinton, MA, USA). The gaseous sample extracts were purified consecutively with a glass column (with an internal diameter at 0.8 cm) packed with 5 cm neutral silica gel and 1 cm anhydrous sodium sulfate from bottom to top. The fraction containing target PAHs was eluted with 15 mL of hexane, and the extract was concentrated to 1 mL and spiked with the internal standards before instrumental analysis.

All samples were analyzed with a Shimadzu gas chromatograph coupled to a mass spectrometer (GCMS-2010 Plus). An HP-5MS capillary column (30 m × 0.25 mm i.d. with a 0.25 μm film thickness) was used for chromatographic separation. The column temperature was programmed from 60 °C (initially held for 1 min), elevated to 250 °C at 20 °C min^{-1} (held for 2 min), ramped to 280 °C at 5 °C min^{-1} (held for 15 min) and further increased to 300 °C at 20 °C min^{-1} (held for 20 min). All samples were automatically injected (1 μL each) in a programmed temperature vaporizer with an initial temperature of 60 °C and then elevated to 300 °C at 400 °C min^{-1} (held for 10 min). Ultrahigh-purity helium was used as carrier gas at a flow rate of 1 mL min^{-1}. Mass spectra were acquired in the full scan mode with an electron impact energy of 70 eV.

2.4. Quality assurance and quality control

One procedural blank and one field blank sample were analyzed for every batch of 22 particle samples and 12 gaseous samples, respectively. The recoveries of the surrogate standards, i.e., acenaphthene-d_{10}, phenanthrene-d_{10}, chrysene-d_{12}, perylene-d_{12}, benzo[ghi]perylene-d_{12} and coronene-d_{12}, for various samples are represented in Table S1. The recoveries of individual PAH compounds in the spiked samples ranged from 82% to 114%. Concentrations of PAHs in all field samples were corrected by those detected in the corresponding procedural blanks within the same batch but not corrected for the surrogate standard recoveries. The lowest calibration concentrations divided by the actual sample volumes were defined as the reporting limits for the target compounds. The reporting limits for particle-bound and gaseous PAHs

are represented in Table S2, calculated with the air volumes of $30.0-169.2$ m^3 for both particle and gaseous samples. In addition, 66 particle samples (six samples containing 11 size fractions) and six gaseous samples were collected as duplicate samples. Duplicate samples with mean concentrations and standard deviations of target PAHs are presented in Table S3.

2.5. Data analysis

The free sampling approach used in the present study was able to capture the levels of atmospheric PAHs closely associated with human daily life. The sampling scenarios were organized into four types i.e., normal (without unexpected indoor and outdoor sources, including the school sites in Tianhe, Luogang and Zengcheng districts during the dry and wet weather seasons, the office site in Tianhe district during the wet weather season and the office sites in Luogang and Zengcheng districts during the dry and wet weather seasons), cooking (with indoor cooking emission, including the residence sites in Tianhe and Luogang districts during the dry and wet weather seasons), smoking (with indoor smoking emission, including the residence site in Zengcheng district during the dry weather season) and haze episode (with an outdoor haze episode, including the office site in Tianhe district during the dry weather season).

All statistical analyses were performed with PASW Statistics 18 (Chicago, IL, USA). The independent-samples t-test was performed to identify any variability in concentrations of particle-bound and gaseous PAHs between the dry and wet weather seasons and among different urban settings. The relationships between the indoor and outdoor PAH concentrations were examined using the indoor-to-outdoor (I/O) ratio of individual PAHs. An I/O ratio greater than 1 means positive contribution from indoor sources to outdoor PAHs air concentrations and vice versa (Naumova et al., 2002). Linear regression was conducted between indoor and outdoor concentrations of total and individual PAHs for each sampling site without indoor source emission.

Chrysene was used as a tracer to estimate the infiltration factor (F_{inf}) for assessing the contributions of outdoor-generated PAHs to indoor loadings (Choi and Spengler, 2014; Mohammed et al., 2016). In the absence of any significant indoor source (cooking and smoking in the present study), F_{inf} is calculated by standardizing the I/O ratio of each target compound with the concentration of Chr as (Choi and Spengler, 2014)

$$F_{inf} = \frac{PAH_{in}}{Chr_{in}} \times \frac{Chr_{out}}{PAH_{out}} \tag{1}$$

where PAH_{in} and PAH_{out} are the indoor and outdoor concentrations of a PAH compound and Chr_{in} and Chr_{out} are the concentrations of indoor and outdoor Chr. The value of F_{inf} greater than unity means a higher indoor concentration of the target analyte than that from outdoor infiltration only (Choi and Spengler, 2014; Mohammed et al., 2016).

The deposition fractions and concentrations of inhaled particle-bound PAHs in the human respiratory tract were calculated with the simplified equations from the International Commission on Radiological Protection (ICRP) model (International Commission on Radiological Protection, 1994). The model calculates the deposition fractions of inhaled particles in three main regions of the respiratory tract, i.e., head airway (HA, which includes nose, mouth, pharynx and larynx), tracheobronchial region (TB) and alveolar region (AR) (Hinds, 1982; International Commission on Radiological Protection, 1994). More details about the model are provided in Text S2.

Daily intake (DI) and incremental lifetime cancer risk ($ILCR$) are

often used to assess exposure risk via inhalation (Guo and Kannan, 2011; United States Environmental Protection Agency, 1991, 2009)

$$DI = \frac{C \times IR \times ET}{BW} \tag{2}$$

$$ILCR = \frac{C \times IR \times ET \times ED \times EF \times CSF}{BW \times AT} \times 10^{-6} \tag{3}$$

where C is the sum of deposition concentrations of size-fractionated particle-bound and gaseous PAHs with B[a]P equivalent (B[a]P$_{eq}$) concentration in the human respiratory system (ng m^{-3}); IR is the rate of inhalation (m^3 h^{-1}); ET is the event time, i.e., the time duration for a day with exposure (h d^{-1}); BW is the average body weight (kg), ED is the lifetime exposure duration (year), EF is the exposure frequency (365 d year^{-1}), AT is the average time for carcinogenic effects set at 70 years × 365 d yr^{-1} (day) and CSF is the cancer slope factor ((mg kg^{-1} bw day^{-1})$^{-1}$). Because body weights and air inhalation rates vary by age, DI and $ILCR$ of PAHs were calculated for four age groups, i.e., children (<6 years), adolescents (6—17 years), adults (18—60 years) and seniors (>60 years). These are corresponding to four integrated scenarios, i.e., residence and outdoors for children, school, residence and outdoors for adolescents, office, residence and outdoors for adults and residence and outdoors for seniors (Table S4). It should be noted that adolescents are defined as 6—17 years old, because the school sampling sites in the present study are located in primary and middle schools. A Monte Carlo simulation was conducted to assess the uncertainty in variability of daily inhalation intake and $ILCR$ by Crystal Ball software (version 2000.2, Decisioneering, Denver, CO, USA). Although the event time vary with age groups (Table S4), the total indoor and outdoor exposure time is 24 h at the Monte Carlo simulation. The uncertainty, sensitivity and 95% confidence interval (CI) for all calculation were reported with 5000 iterations of simulation.

Due to instrumental errors, some data were lost from individual sampling sites, i.e., concentrations in both indoor and outdoor samples at the residential site in Zengcheng during the wet weather season. Particles collected with MOUDI were divided into three diameter groups corresponding to larger than 1.8 μm, between 0.1 and 1.8 μm and smaller than 0.1 μm, which are defined as coarse, accumulation mode and ultrafine particles, respectively. The sum of accumulation mode and ultrafine particles is defined as fine particles.

3. Results and discussion

3.1. Temporal and spatial variability of particle-bound and gaseous target PAHs

Of 16 PAHs, individual analytes not detected or with concentrations below the reporting limits in most samples will not be further discussed herein. The remaining analytes are designed as $\sum_p$ PAH (sum of particle-bound Ace, Flo, Phe, Fla, Pyr, B[a]A, Chr, B[$b+k$]F, B[e]P, B[a]P, IcdP, DahA, BghiP and Cor), $\sum_g$ PAH (sum of gaseous Ace, Flo, Phe, Ant, Fla, PYR, B[a]A and Chr) and $\sum_{(p+g)}$ PAH (sum of $\sum_p$ PAH and $\sum_g$ PAH). Statistical data are summarized in Table 1.

The concentrations of particle-bound PAHs were significantly higher in the dry weather season than in the wet weather season (t-test and $p < 0.05$), while those of gaseous PAHs were only slightly higher in the dry weather season than in the wet weather season (t-test and $p > 0.05$). Such variability may partially be explained by wet deposition, which would remove a much larger portion of particle-bound PAHs in the wet weather season than in the dry

Table 1
Concentrations (Mean, Median, Minimum and Maximum; ng m^{-3}) of airborne PAHs in Guangzhou, China.

	Σ_p PAH[a]				Σ_g PAH[b]				$\Sigma_{(p+g)}$ PAH[c]			
	Mean ± SD[d]	Median	Min	Max	Mean ± SD	Median	Min	Max	Mean ± SD	Median	Min	Max
Seasonality												
Dry Weather Season	5.06 ± 2.50	4.79	2.35	11.9	383 ± 339	309	40.2	1280	388 ± 341	314	42.5	1290
Wet Weather Season	3.07 ± 0.96	3.16	1.71	5.30	268 ± 136	239	59.8	524	271 ± 136	244	63.8	527
District												
Tianhe	3.90 ± 1.42	3.32	2.35	5.94	237 ± 155	213	40.2	524	241 ± 156	218	42.5	527
Luogang	3.41 ± 1.43	2.93	1.71	5.64	276 ± 120	313	59.8	419	279 ± 120	318	63.8	422
Zengcheng	5.55 ± 3.31	3.88	2.98	11.9	548 ± 424	400	167	1280	553 ± 427	403	171	1290
Urban Setting												
School	3.50 ± 1.08	3.16	2.35	5.64	214 ± 156	171	40.2	530	217 ± 156	174	42.5	534
Office	3.76 ± 1.45	3.30	1.83	5.94	295 ± 119	316	74.8	420	299 ± 118	318	80.6	423
Residence	5.25 ± 3.23	4.61	1.71	11.9	501 ± 386	341	117	1280	506 ± 389	345	119	1290
Sampling Site												
Indoors	3.78 ± 2.37	3.06	1.75	11.9	334 ± 274	318	40.2	1280	338 ± 276	322	42.5	1290
Outdoors	4.52 ± 1.89	4.13	1.71	9.50	323 ± 267	274	59.8	1140	327 ± 268	280	63.8	1150

[a] Σ_p PAH is sum of particle-bound Ace, Flo, Phe, Fla, Pyr, B[a]A, Chr, B[b+k]F, B[e]P, B[a]P, IcdP, DahA, BghiP and Cor.
[b] Σ_g PAH is sum of gaseous Ace, Flo, Phe, Ant, Fla, Pyr, B[a]A and Chr.
[c] $\Sigma_{(p+g)}$ PAH is sum of Σ_p PAH and Σ_g PAH.
[d] Standard deviation.

weather season.

Both particulate and gaseous concentrations of PAHs were not consistent with the level of urbanization in the administrative districts under investigation, which follows the sequence of Tianhe > Luogang > Zengcheng, with population densities of 15400, 1000 and 650 km^{-2} and GDP of 280, 190 and 90 billion RMB (Elivecity Laboratory, 2013; Guangzhou Statistics, 2013). In urban areas, PAHs are generally derived from incomplete combustion of fossil fuels (Zhang et al., 2007), especially traffic emissions (Nielsen, 1996). Transport of atmospheric PAHs would result in spreading of PAHs from urban centers to adjacent regions, i.e., rapid urbanization can aggravate environmental pollution (Shao et al., 2006). Besides traffic emissions, human daily activities such as cooking and smoking may also add to the pre-existing contaminant loads, which is perhaps why total PAH concentrations varied widely at the sampling sites, i.e., 42.5–527 ng m^{-3} in Tianhe, 63.8–422 ng m^{-3} in Luogang and 171–1290 ng m^{-3} in Zengcheng (Table 1). These results were corroborated by the distribution of indoor PAH concentrations (particle + gas) among three urban settings, i.e., higher in residence (506 ng m^{-3}) than in office (299 ng m^{-3}) and school (217 ng m^{-3}) (Table 1).

The average indoor and outdoor PAH concentrations were 258 ng m^{-3} (range: 42.5–404 ng m^{-3}) and 268 ng m^{-3} (range: 63.8–534 ng m^{-3}), respectively (Table S5). The I/O ratio of particle-bound PAHs varied within 0.50–0.97 with an average of 0.73 (Fig. 1), suggesting that particle-bound PAHs are mainly derived from outdoor sources. On the other hand, the average I/O ratio of gaseous PAHs was 1.38 (range: 0.30–6.08). Specifically, I/O ratios of low molecular weight (LMW) (2–3-ring) gaseous PAHs were all greater than 1 (Fig. 1), consistent with previously reported findings (Li and Ro, 2000; Naumova et al., 2002), i.e., LMW PAHs were largely generated from indoor sources. Consumer products were found to emit LMW PAHs (Zhang and Tao, 2009), e.g., heated plastic materials, chips and other computer components could emit Phe (Destaillats et al., 2008).

Multiple studies have documented that levels of indoor PAHs are influenced by endogenous sources (Abdullahi et al., 2013; Romagnoli et al., 2014). In residence environment with cooking in the present study, indoor and outdoor PAH concentrations (particle + gas) were 385 and 272 ng m^{-3} (Table S5), higher than 164 ng m^{-3} (indoors) and 36.7 ng m^{-3} (outdoors) from another cooking scenario (Pandit et al., 2001). In the indoor smoking scenario, the indoor and outdoor PAH concentrations (particle + gas)

were 1290 and 1150 ng m^{-3}, 19 times higher than that (66.7 ng m^{-3}) in another tobacco smoking scenario within residential indoor settings in Porto, Portugal (Castro et al., 2011). Such variation may be affected by different emission processes, or even by indoor ventilation efficiency.

A short haze episode occurred from the evening of October 25 to early morning of October 26, 2014 during one of the sampling events at the office site in Tianhe district. The outdoor particle-bound PAHs concentration in the sample collected during this event was 5.94 ng m^{-3}, higher than the average value (4.24 ng m^{-3}) measured during the non-haze periods. A previous study suggested that concentrations of particle-bound PAHs were considerably enhanced under stagnant haze conditions, i.e., air masses were transported to Guangzhou from northeast on October 24, adding to local emissions as the main contributors to PAHs (Tan et al., 2011). The concentration of indoor particle-bound PAHs also increased to 5.59 ng m^{-3} during the haze episode, higher than average value (3.10 ng m^{-3}). Although the data during the haze episode is limited, the derived result still indicated that indoor environment in Guangzhou is incessantly affected by outdoor sources.

3.2. Profiles of particle-bound and gaseous PAHs

The composition profiles of particle-bound and gaseous PAHs by ring size in indoor and outdoor settings varied with four sampling scenarios. Physicochemical properties are an important factor in dictating the gas–particle partitioning of PAHs (Kim et al., 2013), e.g., concentrations of 2–4-ring particle-bound and gaseous PAHs were comparable, while 5–7-ring PAHs occurred in the particulate phase only (Bi et al., 2003). The profiles of particle-bound PAHs in both indoor and outdoor environments were dominated by 4- and 5-ring PAHs, accounting for 27–41% and 25–29% of total concentrations, respectively (Fig. 2). Gaseous PAH profiles were substantially different from those of particle-bound PAHs, with 3-ring PAHs accounting for 81–84% of total levels (Fig. S1). The composition profiles of particle-bound and gaseous PAHs were not significantly different between indoor and outdoor environments (t-test and $p > 0.05$). In contrast, the profiles of indoor LMW PAHs were different with and without indoor emissions. For example, 2–4-ring PAHs accounted for 53% and 54% of indoor particle-bound PAHs under cooking and smoking scenarios, respectively, which is higher than those under normal scenario (43%) (Fig. 2). During the haze event, the contributions of 5–7-ring PAHs to particle-bound
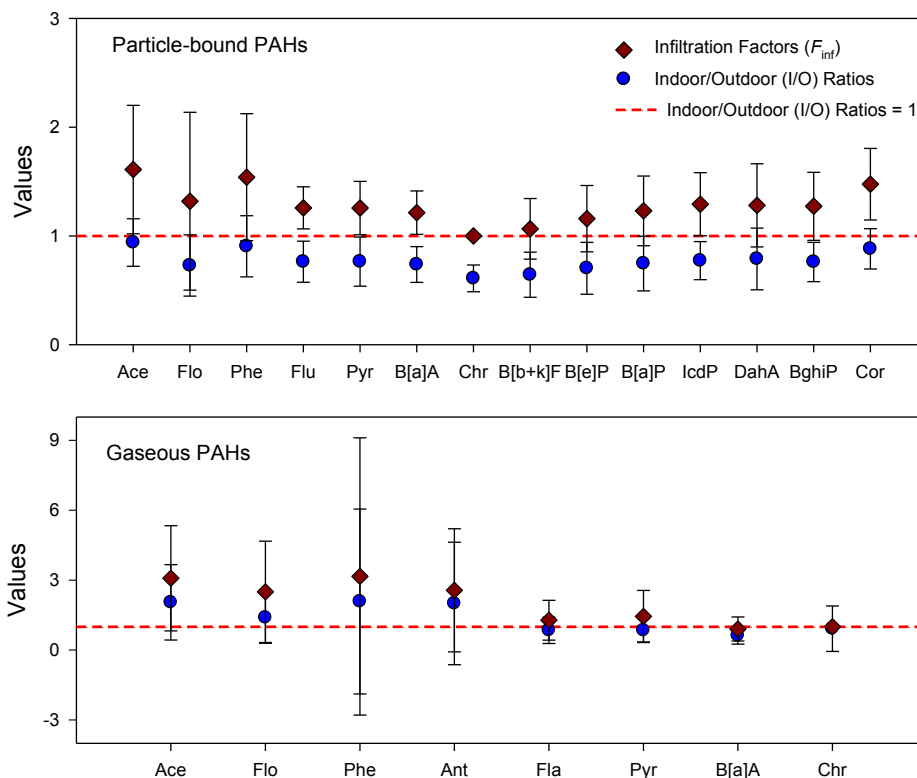
Fig. 1. Estimated infiltration factors (F_{inf}) and indoor/outdoor (I/O) values of particle-bound and gaseous PAHs.

concentrations (indoor: 50%; outdoor: 48%) were greater than those in indoor emission scenarios with cooking and smoking. Tan et al. (2011) also revealed that a haze episode with air masses from north and south of Guangzhou contributed abundant 5–6-ring PAHs (55–75%) to particle-bound PAHs.

3.3. Particle size distribution of PAHs with four sampling scenarios

The particle size distributions (dC/C dlog D_p versus D_p) of individual and total PAHs in both indoor and outdoor settings for four sampling scenarios (normal, cooking, smoking and haze episode) were unimodal, with a sharp peak in the 0.56–1.0 μm fraction (Figs. S2–S5), except for outdoor samples collected during the haze episode (0.32–1.8 μm). While the predominance of PAHs in fine particles has been widely observed in previous studies (Kawanaka et al., 2009; Zhang et al., 2012a, 2012b), the size distribution patterns in the present study were different from a previous finding where total PAHs peaked at 0.32–0.56 μm in summer of Guangzhou (Zhang et al., 2012a). The peak shift toward larger size fraction in the present study may have resulted from prolonged aging processes for accumulation mode PAHs, as discussed by Vaeck and Cauwenberghe (1985).

No obvious peaks were observed for 2-ring PAHs in both indoor and outdoor samples (Figs. S2–S5). The size distributions of 3–7-ring PAHs were essentially unimodal peaking at 0.56–1.0 μm size. In particular, the size profiles of 5–7-ring PAHs were unimodal with a sharp peak at 0.56–1.0 μm (Figs. S3–S4) with indoor cooking and smoking. Several previous studies indicated that particle-bound PAHs emitted freshly from combustion are dominantly distributed in the ultrafine fractions (Miguel et al., 1998; Phuleria et al., 2006), but unimodal size distributions of PAHs peaking at larger particle sizes have been observed from cooking and smoking in the present study than those in previous studies. For example, Saito

et al. (2014) showed that almost all PAHs were concentrated in particles with diameters < 0.43 μm, regardless of food types. Kleeman et al. (2008) reported the greater concentration of PAHs in ultrafine particles (size < 100 nm) than coarse particles from char grilling. Moreover, the particle-bound PAHs emitting from different types of incense smokes exhibited bimodal distributions at < 0.18 and 0.32–0.56 μm (Yang et al., 2007). Different from the previous study, the present results may point to the tendency of PAHs to migrate towards large particles in ambient environment. In particular, it may take longer for indoor particles within closed environments to undergo aging process than fresh particle-bound PAHs emitted from combustion sources (Venkataraman et al., 1994). The similar size-dependent distributions of particle-bound PAHs in indoor and outdoor environments suggested an efficient air exchange that should not be neglected in assessing air quality in indoor environments.

During the haze episode, the size distributions of PAHs remained unimodal, but the peak spread to larger size (0.32–1.8 μm) for 3–4-ring PAHs both indoors and outdoors (Fig. S5). For 5–7-ring PAHs, a major peak occurred around 0.32–1.0 μm indoors and a sharp peak at 0.56–1.0 μm outdoors. The outdoor particle-bound PAHs peak at 1.0–1.8 μm was more prominent than the indoor peak (Fig. S5). During the haze episode, some coarse particles were generated from the transformation of accumulation mode aerosols with prolonged suspension, enhancing the affiliation of PAHs with large particles (Tan et al., 2011), which was probably due to weak infiltration of PAHs affiliated with outdoor large particles into indoor settings.

3.4. Relationship between indoor and outdoor PAH concentrations

Simultaneous measurements of indoor and outdoor PAHs allows the estimation of the infiltration of outdoor-generated PAHs into
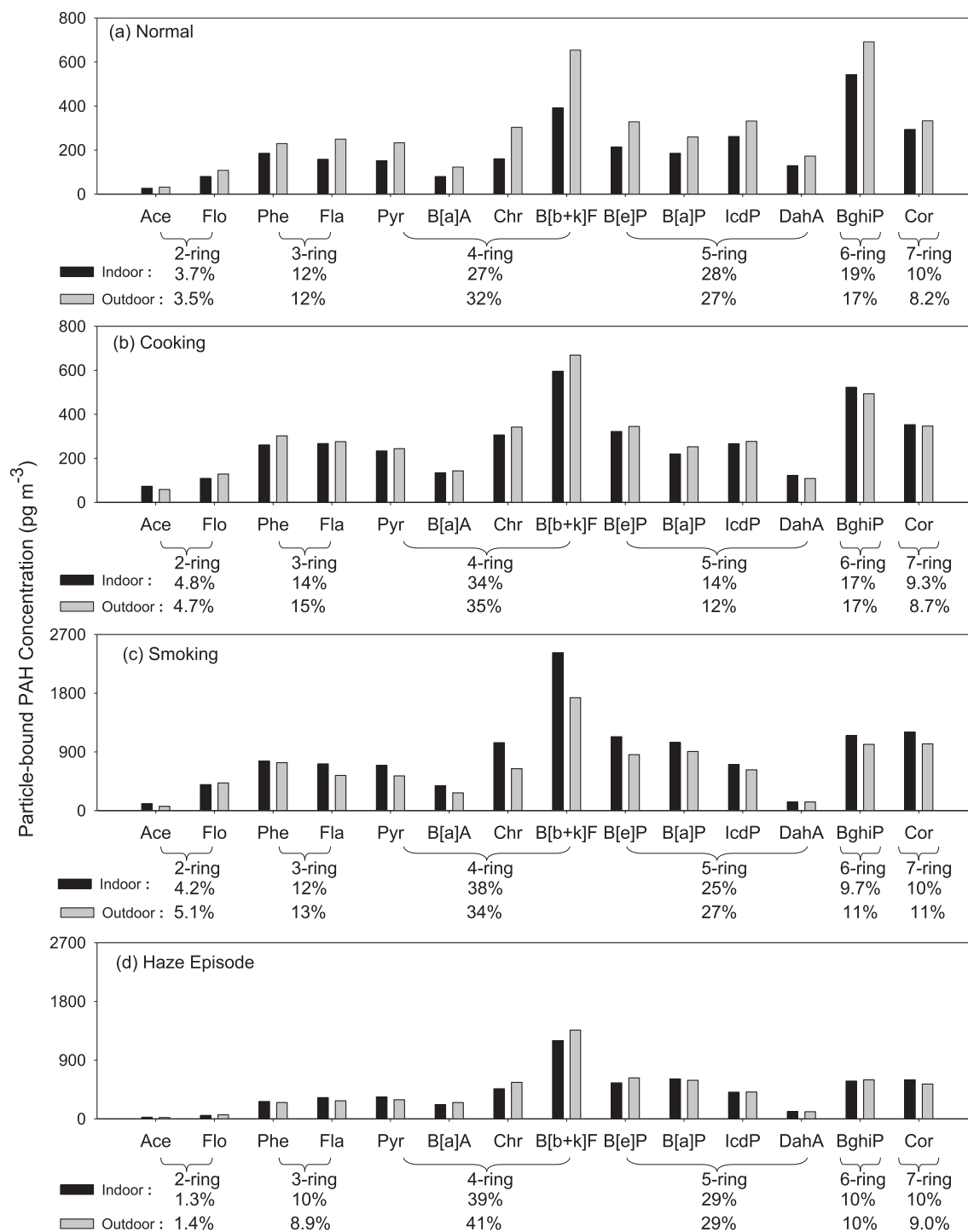
Fig. 2. Composition profiles of particle-bound PAHs under four sampling scenarios (normal, cooking, smoking and haze) indoors and outdoors in Guangzhou, China.

indoor settings (Mohammed et al., 2016). All paired indoor and outdoor samples were organized into 17 groups. The indoor (C_{indoor}) and outdoor ($C_{outdoor}$) concentrations of particle-bound and gaseous PAHs for each group were linearly regressed by

$$C_{indoor} = a\, C_{outdoor} + b \qquad (4)$$

where a and b are the fitting parameters. Significant correlation between C_{indoor} and $C_{outdoor}$ was found for total PAHs both in particle-bound and gaseous samples (Table S6). The coefficients of

determination, r^2, for all groups with significant correlations ranged from 0.38 to 0.99 (average $r^2 = 0.77$) for particle-bound PAHs and 0.62 to 1.0 (average $r^2 = 0.97$) for gaseous PAHs. This implied that 38–99% (or 77% on average) and 62–100% (or 97% on average) of the variations for the indoor concentrations of particle-bound and gaseous PAHs were explained by the outdoor concentrations. This was particularly true for 5–7-ring particle-bound PAHs which exhibited a highly significant correlation ($p < 0.01$). A previous study also obtained r^2 values of 0.64–0.99 for 5–7-ring PAHs, attributed to the predominance of outdoor sources over

indoor respirable 5–7-ring PAHs (Naumova et al., 2002).

The indoor/outdoor relationships for individual PAHs were quite different. Largest variability in I/O ratios (0.11–14) and F_{inf} (0.17–9.3) occurred with 2–4-ring PAHs predominantly present in the gas phase (Table S6). Apparently higher volatile PAHs were more likely affected by indoor sources than outdoors. On the other hand, in the absence of indoor emission sources, good correlation between the indoor and outdoor concentrations of particle-bound 5–7-ring PAHs and their high F_{inf} values (1.2–1.5 greater than 1; Fig. 1) indicated that they were mainly from outdoor-to-indoor exchange. Obviously, atmospheric PAHs exchange between indoor and outdoor environments was efficient and synergistically influenced human inhalation exposure to atmospheric PAHs in ambient environment.

3.5. Regional deposition and size dependence of particle-bound PAHs in human respiratory tract

The inhalable, thoracic and respirable fractions are defined by the International Standards Organization and American Conference of Governmental Industrial Hygienists (Hinds, 1982). The calculated inhalable, thoracic and respirable fractions of PAH concentrations were 91–98%, 80–98% and 68–97% indoors and 89–97%, 78–98% and 63–97% outdoors (Table S7). By comparison, only 30–55% and 32–54% of PAHs indoors and outdoors could deposit in the human respiratory tract as determined from the ICRP model.

The deposition concentrations of particle-bound PAHs were 0.62–4.4 ng m^{-3} indoors and 0.71–3.2 ng m^{-3} outdoors among all sampling sites (Table S8), with the combined mean values (indoors + outdoors) following the sequence of residence (4.1 ng m^{-3}) > office (2.8 ng m^{-3}) > school (2.5 ng m^{-3}). The HA received the most abundant fraction of PAHs, followed by the AR and TB. The regional deposition efficiencies of individual inhaled PAHs in HA, TB and AR were 19–43%, 1.8–2.8% and 8.1–10% indoors and 21–44%, 1.9–2.8% and 7.7–9.9% outdoors, respectively. Apparently, the deposition efficiencies of indoor and outdoor particle-bound PAHs in human respiratory tract were similar, although their deposition concentrations were slightly different.

The deposition concentrations of particle-bound PAHs in different respiratory tract regions were size-dependent (Fig. 3). Coarse particles-affiliated PAHs predominantly deposited in the HA (38–47%), while PAHs in fine particles mostly occurred in the AR (68–75%) and TB (87–90%) (Table S9). The size-dependent deposition fractions of particle-bound PAHs were different among all sampling sites (Table S9), and also from other previous studies. For example, Zhang et al. (2012a) reported the efficiencies of 87.1–96.3%, 26.4–35.1% and 27.0% for PAHs in coarse, accumulation and ultrafine particles to deposit in the respiratory tract in Guangzhou urban environment. In a study conducted at an e-waste recycling zone (Qingyuan) and urban Guangzhou, PAHs in coarse particles were found to prevalently deposit in the HA (69–91%), while PAHs in fine particles were dominant in the AR (62–80%) and TB (44–68%) (Luo et al., 2015).

3.6. Risk assessment of human inhalation exposure to airborne PAHs

Based on the relative Potency Factor Approach (Flowers et al., 2002), the average values of the total carcinogenic potency B[a]P$_{eq}$ concentrations of particle-bound and gaseous PAHs were 1.2 ± 0.6, 1.4 ± 0.6 and 1.5 ± 0.6 ng m^{-3} indoors at school, office and residence, respectively, which are slightly higher than the European Union's annual average B[a]P$_{eq}$ standard (1 ng m^{-3}) (European Union, 2012). Amid the spatial variability of PAHs in indoor settings, different age groups with different prevailing exposure
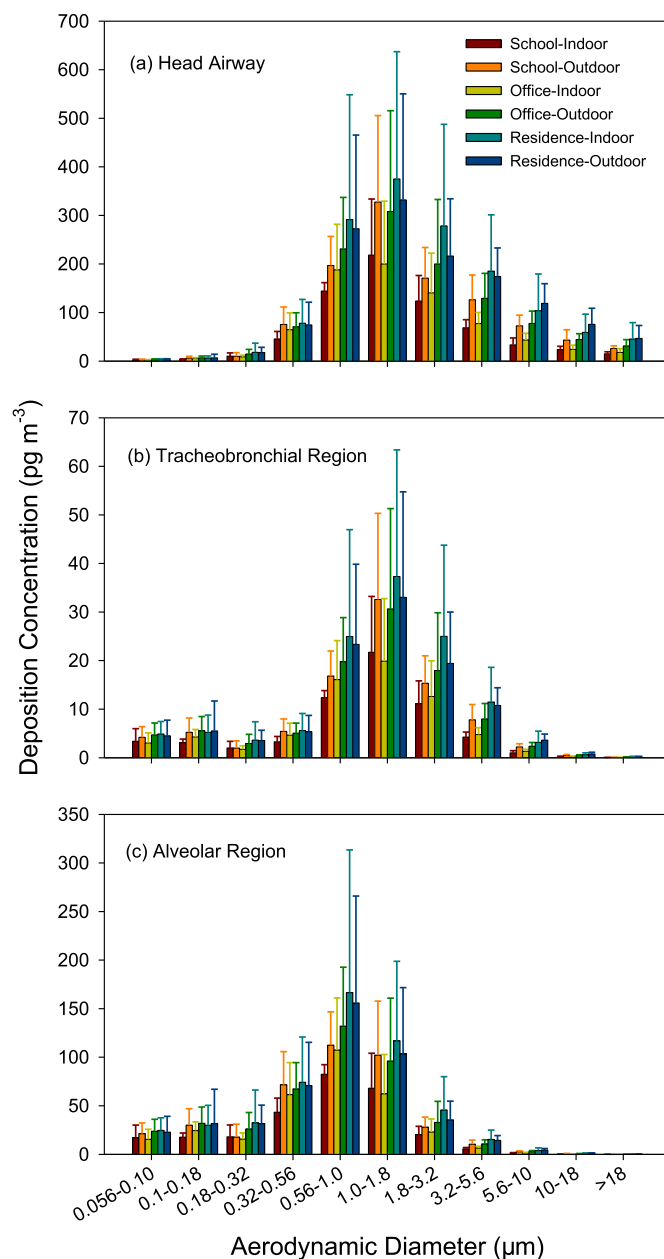

Fig. 3. Concentrations of particle size-fractionated PAHs deposited in different regions of the human respiratory tract both indoors and outdoors at school, office, and residence.

scenarios should be carefully considered in risk assessment of human inhalation exposure to airborne PAHs. Estimated average daily inhalation intake of airborne PAHs for four age groups based on B[a]P$_{eq}$ deposition concentration followed the sequence of children (1.1 ng kg^{-1} bw d^{-1}) > adolescents (0.51 ng kg^{-1} bw d^{-1}) > adults (0.33 ng kg^{-1} bw d^{-1}) > seniors (0.19 ng kg^{-1} bw d^{-1}) (Table 2). For all four age groups, daily inhalation intake through indoor air exposure to PAHs was 3–9 times greater than that through outdoor exposure (Table 2), suggesting higher health risk in indoor environments than in outdoor environments for general population except outdoor workers in urbanized megacities of China such as Guangzhou.

Inhalation *ILCRs* from exposure to indoor and outdoor airborne PAHs for all four age groups ranged from 8.8×10^{-8} (95% CI: 4.9×10^{-9}–3.3×10^{-7}) for seniors to 5.9×10^{-7} (95% CI: 6.3×10^{-8}–

Table 2
Daily inhalation intakes (*DI*; ng kg^{-1} bw d^{-1}, B[a]P equivalent (B[a]P$_{eq}$)) and incremental lifetime cancer risk (*ILCR*, 10^{-6}) upon exposure to PAHs for children, adolescents, adults and seniors in indoor and outdoor settings.

	DI			ILCR		
	Indoor	Outdoor	Total	Indoor	Outdoor	Total
Children (<6 yr)	0.84 (0.13–2.5)[a]	0.25 (0.02–0.88)	1.1 (0.17–3.2)	0.24 (0.02–0.79)	0.07 (0.003–0.27)	0.30 (0.03–1.0)
Adolescents (6–17 yr)	0.36 (0.07–1.0)	0.13 (0.01–0.46)	0.51 (0.10–1.4)	0.19 (0.03–0.59)	0.06 (0.004–0.23)	0.26 (0.04–0.77)
Adults (18–60 yr)	0.28 (0.03–0.90)	0.03 (0.002–0.12)	0.33 (0.04–1.0)	0.53 (0.06–1.8)	0.06 (0.03–0.23)	0.59 (0.06–1.9)
Seniors (>60 yr)	0.14 (0.01–0.54)	0.04 (0.002–0.16)	0.19 (0.01–0.67)	0.07 (0.003–0.24)	0.02 (0.001–0.06)	0.09 (0.005–0.33)

[a] A (B–C) represents the median A and a 95% confidence interval.
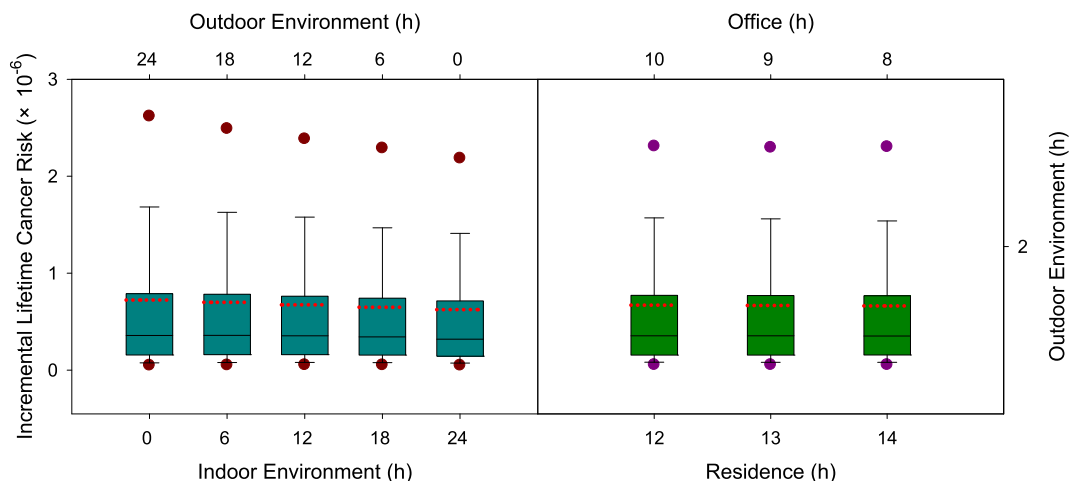
Fig. 4. Incremental lifetime cancer risk induced by inhalation exposure to indoor and outdoor PAHs for adults under different exposure scenarios. Results for the indoor environment were obtained based on the concentrations of PAHs for the residence only.

1.9×10^{-6}) for adults (Table 2). All average *ILCR* values for residents were lower than the risk threshold (1.0×10^{-6}; Table 2) (United States Environmental Protection Agency, 1989), suggesting lower cancer risk for exposure to indoor and outdoor air PAHs in Guangzhou. The calculated values are generally lower than and comparable to the *ILCR* values obtained in other urban areas of China. For example, the median *ILCR* of PAHs for urban citizens in Tianjin was 2.2×10^{-4} (Bai et al., 2009). The geometric mean inhalation *ILCRs* for children and adults in Taiyuan were 1.76×10^{-6} and 7.95×10^{-6}, respectively (Xia et al., 2013). Zhang et al. (2009) reported the arithmetic mean for general residents in 2003 as 1.79×10^{-4} in Shanxi Province of China and 6.5×10^{-6} for all of China. In addition, the indoor inhalation *ILCRs* for general population from exposure to PAHs (0.02×10^{-6}–2.7×10^{-6}) were higher than outdoor ones (0.008×10^{-6}–1.0×10^{-6}; Table 2), further suggesting the urgency to minimize indoor exposure to airborne PAHs. Furthermore, it is noteworthy that the *ILCR* value for adults (average: 6.2×10^{-7}; 95% CI: 4.9×10^{-8}–2.1×10^{-6}), estimated from indoor air PAH levels only in residence with 24-h exposure time, was comparable to our assessment results (average: 5.9×10^{-7}; 95% CI: 6.3×10^{-8}–1.9×10^{-6}), while the risk value derived from outdoor concentrations (average: 7.3×10^{-7}; 95% CI: 5.2×10^{-8}–2.6×10^{-6}) with 24-h exposure time was significantly (t-test; $p < 0.01$) higher than our estimation (Fig. 4). This was obviously attributed to the higher B[a]P$_{eq}$ concentrations of particle-bound 5–6-ring PAHs outdoors than indoors (Table S8). Apparently, the extent of overestimation for exposure risk could become greater if the total concentration of particle-bound PAHs other than the deposition concentration was included in the assessment. The findings also suggested that the respiratory risk

assessment should be conducted with specific details, e.g., both indoor and outdoor environments within different sampling scenarios to better reflect human daily life.

4. Conclusions

The present study has demonstrated that the concentrations of indoor airborne PAHs were elevated by common contributions from indoor source emission and outdoor infiltration with outdoor-to-indoor exchange. The profiles and size distributions of particle-bound PAHs were related to various lifestyles, such as cooking and smoking. Particle-bound PAHs with accumulation mode contributed the majority of PAHs that may deposit in the human respiratory system. All average *ILCR* values for four age groups were below the lower limit of the Safe Acceptable Range (10^{-6}). Collectively, the health risks of exposure to indoor and outdoor PAHs via inhalation were low for residents with different age groups in Guangzhou, China.

Appendix A. Supplementary data

Supplementary data related to this article can be found at https://doi.org/10.1016/j.envpol.2018.03.040.

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