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# Exposure and health risk assessment of $PM_{2.5}$-bound polycyclic aromatic hydrocarbons during winter at residential homes: A case study in four Chinese cities

Zhuozhi Zhang[1], Qi Yuan[1], Meng Wang[1], Tafeng Hu[2], Yu Huang[2], Guangli Xiu[3], Senchao Lai[4], Yuan Gao[5], Shun Cheng Lee[1*]

[1] Department of Civil and Environmental Engineering, The Hong Kong Polytechnic University, Hung Hom, Hong Kong, China

[2] State Key Laboratory of Loess and Quaternary Geology (SKLLQG), Key Laboratory of Aerosol Chemistry and Physics, Institute of Earth Environment, Chinese Academy of Sciences (IEECAS), Xi'an 710061, China

[3] School of Resources and Environmental Engineering, East China University of Science and Technology (ECUST), Shanghai 200237, China

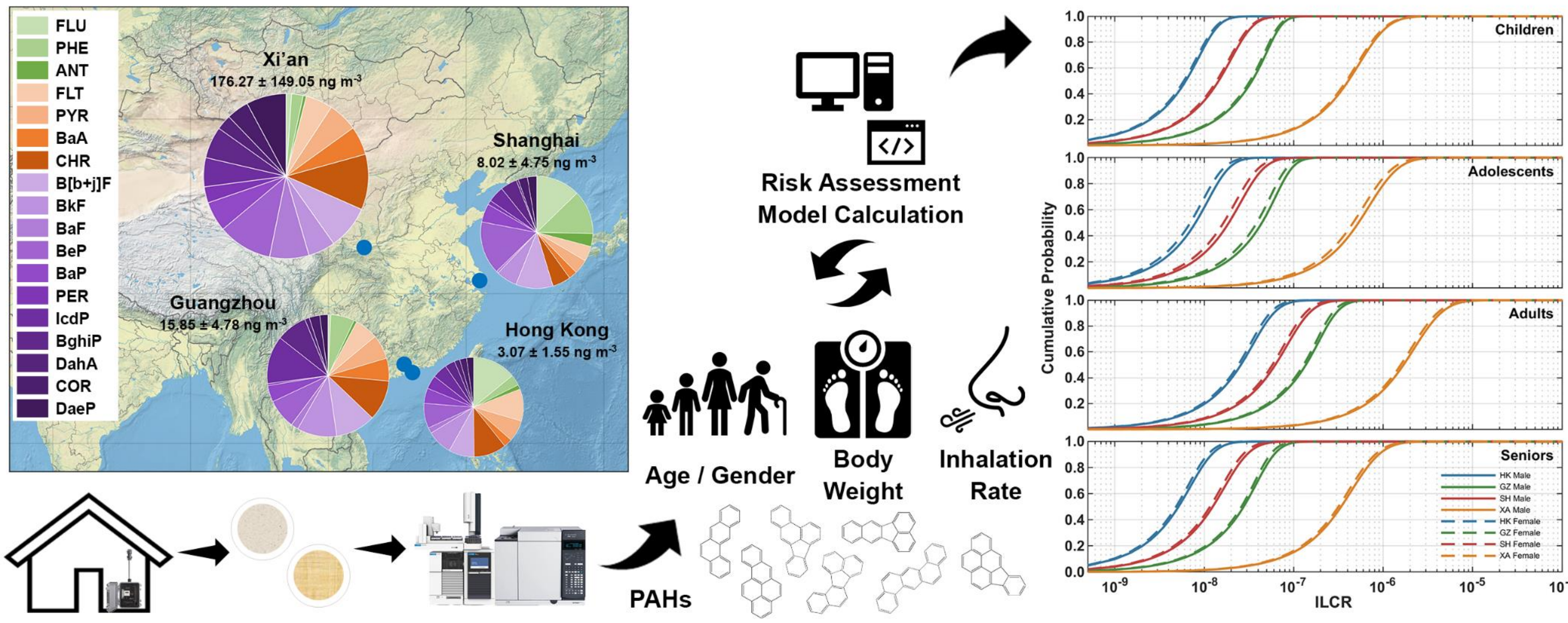
[4] The Key Lab of Pollution Control and Ecosystem Restoration in Industry Clusters, Ministry of Education, School of Environment and Energy, South China University of Technology (SCUT), Guangzhou 510006, China

[5] Instrumentation and Service Center for Science and Technology, Beijing Normal University, Zhuhai 519087, China

[*] Corresponding author. Department of Civil and Environmental Engineering, The Hong Kong Polytechnic University, Hung Hom, Hong Kong, China.

Email address: shun-cheng.lee@polyu.edu.hk (S.C. Lee)

Graphical Abstract



Highlights

- Indoor $PM_{2.5}$-bound PAHs showed city-specific characteristics.
- Influence from outdoor infiltration was significant for indoor $PM_{2.5}$-bound PAHs.
- Inhalation cancer risk in Xi'an was 1-2 magnitudes higher than other cities.
- Early-age inhalation cancer risk may require special attention for Xi'an residents.

**Abstract**

Residential indoor $PM_{2.5}$ were concurrently collected in Hong Kong, Guangzhou, Shanghai, and Xi'an during the winter and early spring seasons of 2016–2017, for updating the current knowledge of the spatial variation of indoor air pollution and the potential health risks in China. $PM_{2.5}$-bound polycyclic aromatic hydrocarbons (PAHs) were characterized, and the associated inhalation cancer risks were assessed by a probabilistic approach. Higher levels of indoor PAHs were identified in Xi'an residences (averaged at 176.27 ng $m^{-3}$) with those of other cities ranging from 3.07 to 15.85 ng $m^{-3}$. Traffic-related fuel combustion was identified as a common contributor to indoor PAHs through outdoor infiltration for all investigated cities. Indoor PAHs profiles showed city-specific differences, while distinctions between profiles based on indoor activities or ambient air quality were limited. Similar with the total PAHs concentrations, the estimated toxic equivalencies (*TEQ*) with reference to benzo[a]pyrene in Xi'an residences (median at 18.05 ng $m^{-3}$) were above the recommended value of 1 ng $m^{-3}$ and were magnitudes higher than the other investigated cities with estimated median *TEQ* ranging from 0.27 to 1.55 ng $m^{-3}$. Incremental lifetime cancer risk (ILCR) due to PAHs inhalation exposure was identified with a descending order of adult (median at $8.42 \times 10^{-8}$) > adolescent ($2.77 \times 10^{-8}$) > children ($2.20 \times 10^{-8}$) > senior ($1.72 \times 10^{-8}$) for different age groups. Considering the lifetime exposure-associated cancer risk (LCR), potential risks were identified for residents in Xi'an as an LCR level over $1 \times 10^{-6}$ was identified for half of the adolescent group (median at $8.96 \times 10^{-7}$), and exceedances were identified for about 90% of the groups of adults (10th percentile at $8.29 \times 10^{-7}$) and seniors (10th percentile at $1.02 \times 10^{-6}$). The associated LCR estimated for other cities were relatively insignificant.

**Keywords**

Fine particulate matter; PAH; Residence; Indoor activities; Inhalation cancer risk; Probabilistic risk assessment

## 1. Introduction

Numerous adverse health effects were found to be associated with inhalation of particulate matters (PMs) (Pope et al., 2002; Pope and Dockery, 2006; Chen et al., 2013; Heo et al., 2014). The World Health Organization (WHO) pointed out that approximately seven million premature deaths are related to air quality issues annually, and about 99% of the world population is exposed to air pollutants exceeding the WHO guideline values (World Health Organization, 2022). PMs as carriers in the atmosphere participate in various chemical processes, hence the constituents of it could be complex and highly toxic (Kelly and Fussell, 2012). By categorizing the size of PMs, the fine suspended particulates with an aerodynamic diameter less than 2.5 μm ($PM_{2.5}$, or FSP), which can penetrate into the human respiratory system, are the most concerned class of PMs owing to their potential health effects. Besides the laboratory-based *in-vitro* studies, the assessment of health effects associated with PMs was mostly based on ambient PM samples (Kim et al., 2015; Chen and Hoek, 2020). However, people tend to spend more time in indoors, that Klepeis et al. (2001) reported for nearly 10,000 survey subjects, approximately 90% of the time was spent in indoors, among which 80% was spent in residences. In addition, some recent studies have demonstrated that health effects associated with indoor air pollution could be even more significant than those related to ambient air pollution (Fan et al., 2018; Wu et al., 2018; Chi et al., 2019). Investigation of indoor PMs is necessary for comprehensive assessment of the health impacts induced by PMs inhalation.

Polycyclic aromatic hydrocarbon (PAH), which is a group of hydrocarbons composed of multiple aromatic rings, is one of the often-concerned groups of species in PMs, due to its carcinogenicity and genotoxicity (Bostrom et al., 2002; Kim et al., 2013). Benzo[a]pyrene, which is the most representative PAH congener in terms of carcinogenic toxicity, has been classified as Group 1 carcinogen (definitely carcinogenic to humans) by the International Agency for Research of Cancer (IARC) (International Agency for Research on Cancer, 2022). Some other PAH congeners were categorized as Group 2A and 2B carcinogens, which are likely to be carcinogenic to humans (International Agency for Research on Cancer, 2022). It was estimated that a working life (40 years) exposure to PAHs mixture with a benzo[a]pyrene equivalent concentration of 0.25–2.5 μg $m^{-3}$ could be associated with a 50% increase in lung cancer (Armstrong et al., 1994). Airborne PAHs are mostly associated with direct emission (i.e., petrogenic sources, from petroleum, oil products, etc.) or combustion processes (i.e., pyrogenic sources) (Yunker et al., 2002). It was concluded that, in Asia, indoor PAHs are mainly from infiltrated traffic emissions, cooking processes, and biomass burning (Ma and Harrad, 2015). Recent study reported a global range of 2.23 to 14,300 ng $m^{-3}$ with a weighted median concentration of 369 ng $m^{-3}$ for indoor ΣPAHs through a data-mining approach from previous related studies and concluded that the indoor PAHs are still posing carcinogenic risks to occupants (Wang et al., 2021).

Potential chronic health impacts induced by PM were usually evaluated by cancer risk assessment. Assessment models standardized in USEPA documents (USEPA, 1991; USEPA, 2005) were widely adopted in past studies (Morawska et al., 2013). Point estimation of cancer risks based on either average or maximum observed values of pollutant concentrations in ambient air were presented in most previous studies (Leung et al., 2014; Hong et al., 2016; Li et al., 2016; Yan et al., 2019). Wang et al. (2021) concluded that 2.6 times higher indoor ΣPAHs level compared with that of outdoor from more than 70 previous studies reviewed, indicating a non-negligible contribution from indoor exposures to the health impacts. Some studies have estimated the indoor or outdoor PAH levels utilizing indoor to outdoor (I/O) ratios for the subsequent risk assessment (Chen and Liao, 2006; Taghvaee et al., 2018). Meanwhile, the point estimation may not be comprehensive enough as the variations of parameters in the model were not considered. Probabilistic risk assessment (PRA) approach, which treats variables in the risk assessment model as probabilistic distributions, provides information on the variability in risk (USEPA, 2001).

In this study, we conducted a joint $PM_{2.5}$ sampling campaign in four Chinese cities with different environmental and cultural contexts, i.e., Hong Kong (HK), Guangzhou (GZ), Shanghai (SH), and Xi'an (XA), in both residential indoors and corresponding outdoors during the same period from November 2016 to mid-April 2017. Particle-bound PAHs were examined for characterizing the spatial variations of indoor and outdoor PAH pollution, identifying the possible PAHs sources, and estimating the particle-bound PAHs inhalation cancer risks through a probabilistic approach.

## 2. Material and Methods

### 2.1 Sampling Sites

Locations of the investigated cities are shown in **Fig. 1**. Brief introduction of the investigated cities is provided in **Text S1**. In brief, the investigated cities are with different environmental and cultural contexts, representing different typical regions of China in terms of status of air pollution and lifestyles of the residents, and these may lead to spatial variations of the residential indoor air pollution and the associated inhalation health risks. Information of sampling periods and site characteristics is summarized in **Table S1**. As described in our previous study (Zhang et al., 2020) and illustrated in **Fig. S1**, the investigated residential homes are separately located in urban and suburban residential areas of the cities. In addition, multiple indoor activities in normal families including cooking, tobacco smoking, and incense burning were identified for the investigated residential homes. Hence, the investigated sites were considered being capable to represent the general condition of the four cities.

## 2.2 Filter Sampling and Handling

Detailed information on sampling sites and sampling methods has been reported in our previous study (Zhang et al., 2020), and is described in **Text S1**. In brief, indoor $PM_{2.5}$ samples were collected at a sampling height of about 1.5 m above the ground level in investigated residential homes indoor. Samples were collected on 47 mm Quartz-fibre filters (PALL, USA), which were pre-fired at 900 ℃ for 3 hours before sampling. Clean filters were stored in petri slides (Millipore, USA) in sealed zip bags for minimizing contamination. MiniVol Air Samplers (Airmetrics, USA) were used for sample collection at a flow rate of 5 L $min^{-1}$. A 24-hour sampling duration was adopted for each sample, with a total sampled air volume of 7.2 $m^3$. Two consecutive 24-hour samplings were performed at most of the residences. Sample filters were immediately stored in freezer below 4 °C until laboratory analysis. A total of 40, 14, 40, and 42 indoor particle samples were collected in Hong Kong, Guangzhou, Shanghai, and Xi’an, respectively. Outdoor samples were also collected at balconies of the corresponding residences, except for those in Hong Kong, as there are fewer homes with balconies in Hong Kong. Due to the issue of availability of samplers, outdoor samples were collected on separated days to the indoor samples and were not collected at all residences. Hence, the outdoor samples will only be used for estimating the health risks in this study.

## 2.3 Sample Analysis

Analysis of $PM_{2.5}$-bound PAHs was performed via thermal desorption-gas chromatography/ mass spectrometry (TD-GC/MS) method (Ho and Yu, 2004; Ho et al., 2008), of which the detailed analytical method was stated in **Text S2**. In brief, a portion of the sample filter was spiked with known amounts of internal standards [$^2H_{10}$] phenanthrene (PHE-$d_{10}$, ≥98%, Supelco, USA) and [$^2H_{12}$] chrysene (CHR-$d_{12}$, ≥98%, Supelco, USA) in dichloromethane. The sample portion was then divided into strips and loaded in a pre-baked TD tube for subsequent thermal desorption analysis in the injector port of a 6890 GC/5795 MSD (Agilent, USA) system. An HP-5MS capillary column (30 m × 0.25 mm × 0.25 μm, 5% diphenyl/ 95% dimethylsiloxane, Agilent, USA) was used for GC separation, and ultra-high purity Helium at constant flow rate of 1.0 mL $min^{-1}$ was used as carrier gas. The operation parameters and temperature programs of the GC/MS system were described in **Text S2**. A total of 18 species of PAHs were analyzed in this study: fluorene (FLU), phenanthrene (PHE), anthracene (ANT), fluoranthene (FLT), pyrene (PYR), benzo[a]anthracene (BaA), chrysene (CHR), benzo[b]fluoranthene & benzo[j]fluoranthene (B[b+j]F), benzo[k]fluoranthene (BkF), benzo[a]fluoranthene (BaF), benzo[e]pyrene (BeP), benzo[a]pyrene (BaP), perylene (PER), indeno[1,2,3-cd]pyrene (IcdP), benzo[ghi]perylene (BghiP), dibenz[a,h]anthracene (DahA), coronene (COR), and dibenzo[a,e]pyrene (DaeP), respectively. Detailed information on the PAHs was summarized in **Table S2**.

## 2.4 Inhalation Cancer Risk Estimation

The inhalation-induced cancer risk is related to multiple varying factors. We referred to Xia et al. (2013) to estimate the incremental lifetime cancer risk (ILCR) with a probabilistic approach. Compared with other cancer risk assessment models (e.g., widely adopted model based on unit risk of BaP ($UR_{BaP}$)), the model adopted in Xia et al. (2013) includes the contributions from human exposure parameters as well as the exposure durations, enabling the differentiation of estimated risk levels between scenarios or between individuals. The estimation of ILCR was based on a life expectancy of 70 years with different age groups categorized, i.e., children (0–6 years), adolescents (7–18 years), adults (19–60 years), and seniors (61–70 years).

The cancer risk induced by inhalation of $PM_{2.5}$-bound carcinogenic PAHs (CPAHs) was primarily estimated by the toxic equivalencies with reference to BaP (*TEQ*, in ng $m^{-3}$), using **Equation (1)**:

$$TEQ = \sum_{i=1}^{n} (C_i \times TEF_i) \tag{1}$$

where the $C_i$ is the concentration of the $i^{th}$ PAH congener in ng $m^{-3}$; and $TEF_i$ is the toxic equivalency factor of the corresponding $i^{th}$ PAHs congener, which were referred to Nisbet and LaGoy (1992) and Malcom and Dobson (1994). The corresponding *TEF* values were listed in **Table S3**. The $C_i$ was treated as lognormal distributed in **Equation (1)**.

Exposures in different microenvironments, i.e., indoor and outdoor specifically for this study, were both considered in the risk estimation. The daily inhalation exposure (*E*, in ng $day^{-1}$) of $PM_{2.5}$-bound CPAHs in terms of *TEQ* was estimated using **Equation (2)**:

$$E = \sum_{j} \left(TEQ_j \times IR \times T_j\right) \tag{2}$$

where *IR* is the inhalation rate ($m^3$ $day^{-1}$), and $T_j$ is the proportion of exposure time in the $j^{th}$ environment (i.e., indoor or outdoor). Both parameters are varied by individual differences, and were treated as normal distributions. They were extracted and processed from a recent Chinese national investigation of human exposure parameters (Ministry of Environmental Protection of the People's Republic of China, 2013; Duan et al., 2015; Duan et al., 2016), which were more in line with the actual situations of the subject in our study, when comparing with exposure parameters from other sources. The derived parameter *TEQ* from **Equation (1)** was approximated as a lognormal distribution.

Based on the risk assessment model according to USEPA (1991), the ILCR due to inhalation of $PM_{2.5}$-bound CPAHs were estimated by **Equation (3)**:

$$ILCR = (CSF \times E \times ED \times EF \times cf)/(BW \times AT) \tag{3}$$

Where *CSF* is the inhalation cancer slope factor of BaP (mg $kg^{-1}$ $day^{-1})^{-1}$, and the recommended value is 3.9 (mg $kg^{-1}$ $day^{-1})^{-1}$ (Chen and Liao, 2006; OEHHA, 2011); *ED* is the exposure duration (year) according to age groups; *EF* is the exposure frequency (day $year^{-1}$), of which the value is set as 90 day $year^{-1}$ as only winter exposure (from December to February) was considered in this study; *BW* is the body weight (kg); *AT* is the averaging time of carcinogenic exposure (25550 days); and *cf* is the conversion factor ($10^{-6}$ mg $ng^{-1}$). *BW* is another human exposure parameter which can be referred to the Chinese national investigation (Ministry of Environmental Protection of the People's Republic of China, 2013; Duan et al., 2015; Duan et al., 2016), and was treated as a normal distribution. *ED* was treated as uniformly distributed within the total exposure duration of the specific age group (the values are 6, 12, 42, and 10 years for children, adolescents, adults, and seniors, respectively). The method of treatment for the human exposure parameters (i.e., *IR*, *T*, and *BW*) was described in **Text S3**, and the derived human exposure parameters were listed in **Table S4**. The derived parameter *E* from **Equation (2)** was approximated as a lognormal distribution.

## 2.5 Quality Assurance and Control (QA/QC)

Measures have been undertaken for ensuring the quality of presentable results. For sample collection, regular calibration of the flow rate of air samplers were conducted throughout the sampling campaign. Field blank samples were collected by placing a filter in the impactor on-site for around five minutes every twenty samples for examining the levels of background contamination. As described in **Section 2.3** and **Text S2**, internal standards were added during sample analysis for quantification of targeted compounds. Besides, five-point calibration curves were established for each targeted PAH congener based on standard mixtures (Wisconsin State Laboratory of Hygiene), and of which the correlation coefficients for linear regressions were all >0.99. Replicate analyses were conducted for each twenty samples, and the resulting relative standard deviations for replicates were <10%. All PAH congeners detected on the field blank samples were close to or lower than the method limit of detection (LOD), therefore subtraction of background level was not conducted for other filter samples.

## 3. Results and Discussion

3.1 Overview of $PM_{2.5}$-bound PAHs Concentrations

**Table 1** shows the city-dependent statistical summary of indoor $PM_{2.5}$-bound PAHs in residences. Relatively low levels of 3.07 ± 1.55 (0.74–9.76) and 8.02 ± 4.75 (2.35–21.79) ng $m^{-3}$ of indoor total detected $PM_{2.5}$-bound PAHs (Σ18PAHs) were identified in Hong Kong and Shanghai residential indoors, while a higher level of 15.85 ± 4.78 (9.59–26.41) ng $m^{-3}$ was found for Guangzhou residences. A distinguishably high Σ18PAHs level averaging at 176.27 ± 149.50 ng $m^{-3}$ was identified for residences in Xi'an with a range of 12.32–600.44 ng $m^{-3}$. The indoor Σ18PAHs level in Hong Kong residences was comparable to those of previous studies (ranging from 1.6 to 2.6 ng $m^{-3}$ during the period of 2010–2017) (Wang et al., 2013; Tong et al., 2019; Chen et al., 2020; Chen et al., 2022), regardless of the sampling period, while the result obtained from Guangzhou residences was found relatively low when compared to those of previous studies (ranged from 18.7 to 41.5 ng $m^{-3}$ during the period of 2002–2019) (Li et al., 2005; Wang et al., 2013; Luo et al., 2021), There are limited studies on residential indoor PM-bound PAHs in Shanghai and Xi'an, whilst studies on different indoor environments in Xi'an were found as references. Without indoor combustion activities, the average indoor PM-bound PAHs levels in March and May 2012 were found at 79.9 ng $m^{-3}$ (Wang et al., 2017) and 53.2 ng $m^{-3}$ (Xu et al., 2015) in an occupied school classroom in Xi'an, which were obviously lower than that of our study. Another study that took place in December 2015 focused on office IAQ in Xi'an reported an average Σ16PAHs in $PM_{2.5}$ of 374.11 ng $m^{-3}$ (Qiu et al., 2021), suggesting seasonal variation of indoor $PM_{2.5}$-bound PAHs levels in Xi'an, despite the type of indoor environments. With relatively limited differences in indoor $PM_{2.5}$ levels between cities (**Table 1**), the Σ18PAHs/PM mass ratios (ΣPAH/PM) for residential indoor also showed a city-dependent trend of Xi'an (0.228 ± 0.119%) > Guangzhou (0.034 ± 0.001%) > Shanghai (0.015 ± 0.001%) > Hong Kong (0.009 ± 0.000%). Unlike the relationships of between indoor and outdoor $PM_{2.5}$ levels (Zhang et al., 2020), the differences between indoor and outdoor Σ18PAHs levels (listed in **Table S5**) were more significant with general low levels of indoor-to-outdoor ratios (ranging from 0.54 for Xi'an to 0.72 for Guangzhou residences).

The detected PAHs were classified according to the molecular weights (MW) and the number of rings into Low Molecular Weight PAHs (LMW), Medium Molecular Weight PAHs (MMW), and High Molecular Weight (HMW), which can be referred to **Table S2**. LMW-PAHs are generally considered as combustion PAHs from various sectors (industrial, transportation, domestic sectors, etc.) as they are associated with the combustion processes of fuels including coal, wood, petroleum, natural gas, etc. Whilst MMW- and HMW-PAHs are more likely to be generated from the production processes of crude oil and its products, which are considered petrogenic PAHs mainly associated with industrial sources (Lima et al., 2005; Wolska et al., 2012). As shown in **Fig. S2**, residences in Shanghai and Hong Kong were with high proportions of LMW-PAHs, with average percentages of 29.1% and 19.1%, respectively, whilst the values of Guangzhou and Xi'an residences were 7.8% and 4.0%. MMW-PAHs contributed around 30% of the total PM-bound PAHs for residences in Hong Kong, Guangzhou, and Xi'an, while for Shanghai residences MMW-PAHs took up a lower proportion of 16.2%. The HMW-PAHs fractions contrast with LMW-PAHs fraction distribution, that high contributions of over 60% were identified in Guangzhou (62.9%) and Xi'an (68.4%) samples and relatively low proportions were found for Hong Kong (50.2%) and Shanghai (54.7%) residences. The MW-based PAHs group proportions of outdoor samples shared similar patterns with those of indoors (except for Hong Kong), as illustrated as triangle markers in **Fig. S2**. These suggest that the distinctions between different cities in terms of the characteristics of indoor PM-bound PAHs may mainly attribute to the differences in outdoor-originated PAHs, which are likely to be associated with the local energy-related emissions and industrial activities.

PAHs from sources may exhibit different patterns, allowing one to differentiate sources based on individual PAHs as source markers (Ravindra et al., 2008a). For instance, CHR and BkF were reported to dominate in coal combustion emissions (Khalili et al., 1995; Ravindra et al., 2008b). Ravindra et al. (2006) reported that BghiP, COR, and PHE presented high levels in vehicle emissions, while incineration emissions were found to be associated with reasonably high levels of PYR, FLT, and PHE, and high concentration of volatile PAHs including FLU, FLT, and PHE along with moderate levels of B[b+j]F and IcdP were found in oil combustion emissions. **Fig. 2** shows the city-dependent average proportions of individual PAH congeners in Σ18PAHs of indoor and outdoor $PM_{2.5}$, and **Table S5** summarizes the outdoor levels of individual PAHs during the study period. Notable discrepancies can be observed between the indoor PAHs profiles of different cities. Samples of Hong Kong residences were found with high levels of FLU (14.1/8.7% for indoor/outdoor, respectively), FLT (10.4/7.4%), and B[b+j]F (8.3/14.7%), which may be attributed to the emissions from diesel-fueled heavy-duty vehicles (HDVs). It was reported that coal accounted for 48% of the electricity generation fuel mix in 2015 (Environment Bureau - The Government of the Hong Kong Special Administrative Region, 2017), which may be associated with the high CHR (10.6/9.2%) and BkF (8.2/10.7%) levels in Hong Kong samples. These two species (10.7/10.8% for CHR, and 10.5/10.9% for BkF) were also at high levels in Guangzhou samples, implying the influences from coal-powered electricity generation plants. Another major contributor for Σ18PAHs in Guangzhou may be the traffic emission, that both B[b+j]F (10.6/10.2%) and IcdP (12.7/12.6%) were found of high levels. For Shanghai residences, the fractions of FLU (12.7/9.2%), B[b+j]F (10.7/10.8%), and PHE (12.6/11.4%) were of high levels compared to other species, which may be related to fuel combustion from traffic and industrial sources, as well as the emissions from incinerators. Samples collected from Xi'an, however, exhibited a more uniform pattern compared with other cities. Considering the need for centralized heating services and domestic heating, the fuel consumption for heat and electricity generation may be raised during winter in Xi'an, which increased the portions of fuel combustion-related species, i.e., CHR (10.9/10.5%) and B[b+j]F (8.6/9.1%), in the particles collected in Xi'an. Besides, high levels of BaF (7.7/10.8%) and DaeP (7.8/6.4%) were found in Xi'an samples, of which the levels were limited in other cities (0.6–2.0% for BaF, and 2.2–4.2% for DaeP). BeP is emitted from multiple sources including vehicle exhaust, industrial emissions, and tobacco smoke (Lima et al., 2005; Lu and Zhu, 2007; Niu et al., 2020), and it contributed high portions in Σ18PAHs of all cities (from 7.8% for Guangzhou indoors to 15.3% for Shanghai indoors), while ANT (0.5–3.8%), PER (0.5–4.4%), DahA (0.6–3.0%), and COR (2.2–4.4%) accounted for fewer proportions in Σ18PAHs.

Similar to our previous study (Zhang et al., 2020), the residences were categorized into three types according to the indoor activities to better characterize the influence induced by indoor combustion. The categories were Type A (without indoor combustion, n = 5), Type B (with indoor cooking, n = 39), and Type C (with indoor cooking and smoking/incense burning, n = 22), respectively. **Table S6** summarizes the concentrations of PAHs in homes with different levels of indoor activities on a city-basis, and **Fig. S3** shows the average Σ18PAHs concentrations as well as the distributions of individual PAH species in different types of indoors collectively for all cities. Considering the relatively less differed indoor $PM_{2.5}$ concentrations, the Σ18PAHs were found to differ greatly between types of homes, especially for Type A homes ($3.00 \pm 2.56$ ng $m^{-3}$, compared with those of Type B and Type C homes). The profiles of MW-based and individual PAHs in indoors with different activities were found less varied when compared with indoors in different cities, especially for Type B and Type C homes, as illustrated in **Fig. S2** and **Fig. S3**.

We also categorized the ambient air pollution into four levels according to $PM_{2.5}$ concentrations, i.e., *Clear* (< 35.0 μg $m^{-3}$), *Light Pollution* (35.0–75.0 μg $m^{-3}$), *Medium Pollution* (75.1–115.0 μg $m^{-3}$), and *Heavy Pollution* (> 115.0 μg $m^{-3}$), respectively, according to the Chinese guidelines (Ministry of Environmental Protection of the People's Republic of China, 2012) in our previous study (Zhang et al., 2020). Referring to **Table S7** and **Fig. S3**, a noticeable trend of increasing indoor PAHs with the deteriorating ambient air quality was found, with the mean Σ18PAHs concentrations increasing from 4.45 ± 3.22 ng $m^{-3}$ for *Clear* cases to 360 ± 161 ng $m^{-3}$ for *Heavy Pollution* cases. However, the highest Σ18PAHs concentration among *Light Pollution* cases (294.44 ng $m^{-3}$) was found higher than that of *Medium Pollution* cases (244.41 ng $m^{-3}$), indicating the possibly significant influence of indoor PAHs sources. As shown in **Fig. S2**, the distributions of MW-based PAHs groups could not be distinguished based on different levels of ambient air pollution cases, and for the distribution of PAH congeners shown in **Fig. S3**, there were fewer differences in profile patterns for *Light*, *Medium*, and *Heavy Pollution* cases.

## 3.2 Source Identification of $PM_{2.5}$-bound PAHs

Diagnostic ratios (DRs) exhibiting intra-source variability and inter-source similarity (Galarneau, 2008) have been used for differentiating PAHs from different sources or processes (Tobiszewski and Namieśnik, 2012; Famiyeh et al., 2021). Four DRs were selected for characterizing the plausible sources of the indoor PAHs in this study (**Table S8**). With relatively large differences in thermodynamic stability between isomers, FLT/(FLT+PYR) and IcdP/(IcdP+BghiP) ratios were reported as the most definitive groups of PAHs for source interpretation, and they were selected for differentiating the combustion processes (Yunker et al., 2002). Some recent combustion emission studies (Zhang et al., 2021a; Zhang et al., 2021b) pointed out that these two DRs are capable to differentiate coal combustion and biomass burning sources. BaP/(BaP+BeP) is photosensitive and was used to indicate the degree of particle aging (Tobiszewski and Namieśnik, 2012; Oliveira et al., 2015). BaP/BghiP was used to characterize whether the particles are related to traffic sources (Katsoyiannis et al., 2007). Tobiszewski and Namieśnik (2012) summarized the influence of the aging process on the diagnostic ratios, based on the studies of PAH congener half-lives on different media including fly ash, carbon black, soot, etc. PYR on most of the media was found consumed slightly faster than FLT in aging processes (Behymer and Hites, 1985; Niu et al., 2007; Kim et al., 2009), while BghiP was found to decay faster than IcdP on soot (Kim et al., 2009). The differences in decay rate for these PAH congeners lead to a slightly higher FLT/(FLT+PYR) and a moderately higher IcdP/(IcdP+BghiP) ratio during the particle aging processes. Degradation rates of BaP and BghiP are similar, and thus photoreactions tend to have less influence on the ratio of BaP/BghiP (Kim et al., 2009).

**Fig. 3** shows the distributions of the DRs of $PM_{2.5}$-bound PAHs on a city basis. For Hong Kong residences, the FLT/(FLT+PYR), IcdP/(IcdP+BghiP), and BaP/BghiP ratios were 0.60 ± 0.03, 0.60 ± 0.04, and 1.63 ± 0.23, respectively, which were all at high levels in comparison with those of other cities. Considering the plausible aging of the particles, which was indicated by the BaP/(BaP+BeP) ratio of 0.38 ± 0.02, the indoor particulate PAHs in Hong Kong may be mainly originated from outdoor infiltration of the traffic emission and coal combustion-related processes, with another certain portion possibly attributed to indoor smoking and incense burning. **Fig. S4** summarizes the DRs of previous indoor particle-phase PAHs studies. The ranges of FLT/(FLT+PYR), IcdP/(IcdP+BghiP), and BaP/BghiP ratios in Hong Kong indoors reported in previous studies were 0.44–0.71, 0.35–0.51, and 0.45–0.89, respectively (Chao et al., 2002; Wang et al., 2013; Tong et al., 2019; Chen et al., 2022), suggesting a mixture of sources including petroleum combustion and biomass or coal combustion. The highest indoor average IcdP/(IcdP+BghiP) ratio (0.61 ± 0.03) was found for Guangzhou residences, indicative of considerable contribution from biomass or coal combustion. With a relatively high level of BghiP (8.2% of $\Sigma$18PAHs), the BaP/BghiP ratio was found abnormally low (0.54 ± 0.07) for Guangzhou indoors. Interestingly, the BaP/BghiP ratios of Guangzhou indoors reported in previous studies were also at low levels (0.45 – 0.62) among cities (Li et al., 2005; Wang et al., 2013; Luo et al., 2021). Though with low BaP/BghiP ratios, the contribution from traffic emission to Guangzhou indoor PM-bound PAHs may be ineligible as indicated by the ratios of FLT/(FLT+PYR) and relatively high levels of traffic-related PAH congeners of B[b+j]F and IcdP. The differences between indoor and outdoor for all selected DRs were found insignificant for Guangzhou indoors. Lowest BaP/(BaP+BeP) ratio (0.26 ± 0.02) was found for Shanghai indoors among the four cities, which may imply a high level of particle aging. In line with this, the FLT/(FLT+PYR), IcdP/(IcdP+BghiP), and BaP/BghiP ratios (0.51 ± 0.03, 0.49 ± 0.03, and 1.12 ± 0.12, respectively) for Shanghai indoors may indicate the dominance of infiltrated traffic-related petroleum combustion sources. The ratios of FLT/(FLT+PYR) (0.48 ± 0.02) and IcdP/(IcdP+BghiP) (0.48 ± 0.03) of Xi'an residences were both the lowest among the four cities and were comparable to those values recorded in classrooms (0.52–0.54 and 0.50–0.54, respectively) from previous studies (Wang et al., 2017; Qiu et al., 2021), implying the strong influence from petroleum combustion sources infiltrated from ambient. The high levels of CHR and B[b+j]F, as well as the BaP/BghiP ratio (0.94 ± 0.18) over 0.6 found for Xi'an indoors also support the finding of the strong influence of petroleum combustion. The difference between indoor and outdoor DRs was found limited for Xi'an residences. A recent study shared similar DRs for Xi'an samples with our study reported that biomass burning accounts for significant portions of ambient PM-bound PAHs in Xi'an due to the massive use of biomass fuel for residential heating (Sun et al., 2022), based on the analysis of more specific tracers and source apportionment approach.

### 3.3 Health Risk Estimation of Exposure to Indoor $PM_{2.5}$-bound PAHs

Most of the PAH congeners investigated in this study are carcinogens. These congeners were marked as CPAHs and the collective carcinogenicities were estimated by *TEQ*. The CPAHs accounted for over 95% of Σ18PAHs in indoor $PM_{2.5}$ of Hong Kong, Guangzhou, and Shanghai residences, and a relatively low value of 84.8% was found for Xi'an homes. The values in outdoor $PM_{2.5}$ samples were found slightly lower than those of indoors for all cities. Decreasing trends of CPAHs proportions were identified for both indoor activities (Type A (96.5%) > Type B (85.5%) > Type C (85.1%)) and ambient air pollution levels (*Clear* (96.3%) > *Light Pollution* (88.1%) > *Medium Pollution* (84.9%) > *Heavy Pollution* (84.2%)). In terms of the contributions from PAH congeners to the *TEQ*, the major contributor was found to be BaP (accounted for 44–62%/46–68% of *TEQ* of indoor/outdoor $PM_{2.5}$) for households in all cities, as shown in **Fig. S5**. With *TEF* equal to BaP, the contributions from DahA ranged from 7–26%/6–23%, which in some cases were less than some PAH congeners with *TEF* of 0.1 (e.g., IcdP (12.8%/12.4%) > DahA (12.3%/11.8%) for Guangzhou samples, and B[b+j]F (12.4%/9.8%) and BkF (7.0%/5.4%) > DahA (6.8%/6.5%) for Shanghai samples). Contributions from these congeners with *TEF* values of 1.0 (BaP and DahA) and 0.1 (IcdP, B[b+j]F, and BkF) were over 90% of total *TEQ* for all samples.

The statistical characteristics of *TEQ*s of indoor $PM_{2.5}$-bound CPAHs were summarized in **Table 2**. Consistent with the indoor ΣPAHs concentrations, the *TEQ* in Xi'an residences (median at 18.05 ng $m^{-3}$) was much higher than those of other cities (median at 0.27, 1.55, and 0.64 ng $m^{-3}$ for Hong Kong, Guangzhou, and Shanghai, respectively). The World Health Organization (WHO) non-mandatory guide value of *TEQ* is 1 ng $m^{-3}$ (Ravindra et al., 2008a).

In view of this, most of the residents in Hong Kong were identified safe from indoor $PM_{2.5}$-bound CPAHs exposure, as for 95% of the cases the values were lower than 0.46 ng $m^{-3}$. Shanghai residences also shared low *TEQ* levels, as approximately 90% of the cases were with a *TEQ* level lower than the WHO recommended value. Most of the Xi'an residences were with *TEQ* levels (6.27 ng $m^{-3}$ at 1st percentile) at least 6-folds higher than the recommended value, implying severe indoor PAHs pollution and potentially high health risk due to inhalation exposure of indoor $PM_{2.5}$ for Xi'an residents. In terms of different indoor activities, a trend of Type C (median at 4.42 ng $m^{-3}$) > Type B (3.56 ng $m^{-3}$) > Type A (0.24 ng $m^{-3}$) could be observed. Standard deviations for Type B and Type C homes were found over 10 ng $m^{-3}$, which shall be induced by the variation of *TEQ* between cities. We calculated the *TEQ*s for indoor PM from previous studies with the same *TEF*s adopted from Nisbet and LaGoy (1992) and Malcom and Dobson (1994), results are shown in **Table S9**. Studies on Hong Kong residential indoors across over a decade showed relatively low levels and a decreasing trend of the indoor particulate *TEQ* (Chao et al., 2002; Wang et al., 2013; Tong et al., 2019; Chen et al., 2022), from 0.60 ng $m^{-3}$ in 2000 (Chao et al., 2002) to 0.11 ng $m^{-3}$ in 2014–16 (Chen et al., 2022). *TEQ* decreasing trend was also observed in Guangzhou residences, from 7.08 ng $m^{-3}$ in 2002 (Li et al., 2005) to 0.79 ng $m^{-3}$ in 2019 (Luo et al., 2021). For Xi'an indoors, the *TEQ* levels were all over 10 ng $m^{-3}$ in previous and our studies (Wang et al., 2017; Qiu et al., 2021).

The probabilistic distributions of daily inhalation exposures (combining both indoor and outdoor exposures) of $PM_{2.5}$-bound CPAHs in terms of *TEQ* for different age groups of occupants in the four cities are shown in **Fig. 4** and the related statistical summary is shown in **Table 2**. Clear trends of Xi'an (median at 261.5 ng $day^{-1}$) > Guangzhou (20.2 ng $day^{-1}$) > Shanghai (9.5 ng $day^{-1}$) > Hong Kong (3.7 ng $day^{-1}$), males (15.0 ng $day^{-1}$) > females (12.5 ng $day^{-1}$), and adults (20.8 ng $day^{-1}$) > seniors (16.8 ng $day^{-1}$) > adolescents (14.7 ng $day^{-1}$) > children (8.5 ng $day^{-1}$) for daily inhalation exposures could be identified. Variations in the inhalation rate were the origin of differences in exposure level between age groups and gender. The difference between gender was less significant for children and seniors as the gaps in inhalation rate between genders were relatively small. Categorized by different indoor activities, the probabilistic distributions of daily exposure are shown in **Fig. S6**. Similar trends could be identified for genders and age groups. With a limited sample size, the exposure level of Type A residents (2.4 ng $day^{-1}$) was found to be significantly lower than those of Type B (35.6 ng $day^{-1}$) and Type C (44.3 ng $day^{-1}$) residents with indoor combustion activities.

It was widely accepted that an incremental cancer risk over a 70-year assessment period above $1.0 \times 10^{-6}$ indicates potential cancer risk, and a level over $1.0 \times 10^{-4}$ may imply considerable carcinogenic effects that shall pay more attention (Xia et al., 2013; Wang et al., 2020). Inhalation cancer risks associated with $PM_{2.5}$ were identified at $10^{-6}$ to $10^{-5}$ levels in Hong Kong, based on previous studies focused on various exposure modes and different risk assessment models (i.e., based on the unit risk of BaP ($UR_{BaP}$)) (Wang et al., 2013; Leung et al., 2014; Chen et al., 2022). Due to different assessment models and exposure parameters adopted, Wang et al. (2013) reported an average lifetime cancer risk of $1.6 \times 10^{-4}$ for Guangzhou residential indoors, which was higher than that of Xi'an ambient ($8.4 \times 10^{-5}$) with higher *TEQ* level (9.6 ng $m^{-3}$ in Xi'an vs. 2.0 ng $m^{-3}$ in Guangzhou) reported by Leung et al. (2014). Average lifetime cancer risks related to airborne PAHs exposures reported in other regions varied from $10^{-14}$ to $10^{-3}$ levels. A similar probabilistic assessment model was adopted in this study, to estimate the ILCR with as many variables as possible considered. The estimated inhalation ILCR of winter particulate CPAHs for residents in the four cities is shown in **Fig. 5** and the related statistical summary is shown in **Table 2**. Noted that for different age groups the exposure durations were independent, this estimation approach enables the differentiation of the potential risks between age groups based on their exposure parameters, i.e., *IR*, *T*, and *BW*, despite the possible accumulation of risk. Clear trends of Xi'an (median at $5.10 \times 10^{-7}$) > Guangzhou ($4.09 \times 10^{-}$

$^{8}$) > Shanghai ($1.80 \times 10^{-8}$) > Hong Kong ($7.59 \times 10^{-9}$) can be observed, which was consistent with the trend of *TEQ* and daily exposure levels. The modeled ILCR for Hong Kong and Shanghai residents were mostly under $10^{-7}$ level, that at the 95% percentiles of the ILCR datasets for Hong Kong and Shanghai residents were $4.45 \times 10^{-8}$ and $1.09 \times 10^{-7}$, respectively. Different from the distributions of daily inhalation exposure, the trend of ILCR of age groups was adults ($8.46 \times 10^{-8}$) > adolescents ($2.52 \times 10^{-8}$) > children ($2.00 \times 10^{-8}$) > seniors ($1.70 \times 10^{-8}$). Xia et al. (2013) identified a higher risk for the children group compared with adolescent and senior groups, which could be attributed to a different age range defined for the children group, thus the related exposure parameters may varied, leading to differences in exposure parameters and calculated ILCRs. Sensitivity analysis was conducted for evaluating the influences of each parameter to the total ILCR variances. We used the ranges of input parameters including *TEQ* of indoor ($TEQ_{in}$) and outdoor ($TEQ_{ou}$), *IR*, *T* in indoor ($T_{in}$) and outdoor ($T_{ou}$), *ED*, and *BW* for different genders, age groups, and cities in the analysis. **Fig. 6** shows the combined sensitivity analysis results for different age groups. *ED*, which is directly related to the intake amounts accounted for the most contributions (48–74%) to the total ILCR variances. $TEQ_{in}$ and *BW* were also found influential to ILCR, for which the contribution ranges were 3–24% and 5–8%. Influence from indoor exposures in terms of *TEQ* was more significant for Xi'an residences (>20%) than for other cities. **Fig. S7–S10** show the detailed sensitivity analysis results for different age groups in the four cities. Contributions from variance differed between age groups, for example, $TEQ_{in}$ showed more significant influence on the cancer risk for children and adolescent groups than adult and senior groups, while contrast relationships could be identified for $TEQ_{ou}$, $T_{in}$, and $T_{ou}$. Noted the differences on cancer risks between genders were mainly attributed to *IR*, and the influences were more significant for adult and senior groups. Differences on *BW*, on the other hand, were less influential to the inequalities of cancer risks between genders for all age groups.

The accumulated lifetime cancer risks (LCR) due to lifetime exposure were also estimated by adding up the estimated ILCR results of different age groups, assuming the *TEQ* levels remain relatively stable during the assessed lifetime period. The results are presented in **Fig. S11** with the statistical summary shown in **Table 2**. The LCR estimated for males was higher than that for females for all age groups. Insignificant risks were identified for Hong Kong (mostly at $10^{-8}$ level for the senior group), Guangzhou, and Shanghai (both ranged from $10^{-8}$ to $10^{-7}$ levels) residents. These results were lower than the lifetime cancer risks assessed in previous studies (Wang et al., 2013; Leung et al., 2014; Li et al., 2016). Potential risks could be identified for the adolescent group in Xi'an, that around half of this group was found with cancer risk over $10^{-6}$ level. And for the senior group in Xi'an over 90% were with potential risk above $10^{-6}$ level with a median value of $3.19 \times 10^{-6}$, which was also lower than the previous result of ambient PAHs (Leung et al., 2014). ILCR related to different indoor activities were also studied, of which the results are shown in **Fig. S12**. Trends for age group and gender difference were similar to those of city-dependent ILCR results. The overall ILCR of Type C homes residents (median at $9.10 \times 10^{-8}$) was slightly higher than that of Type B residents ($7.33 \times 10^{-8}$) and were both much higher than that of Type A residents ($4.83 \times 10^{-9}$).

It was acknowledged that there were limitations in this work. One of the scopes of this work is to understand the variations of indoor particle-phase PAHs levels and the associated health risks between different regions in China. In this study, four cities with different environmental and cultural contexts were selected as representatives, yet studies in more cities with diverse features are needed. In addition, due to the limited number of samples obtained, increased uncertainty would occur during the risk assessment for Guangzhou residents. From the aspect of cancer risk estimation, we made a few assumptions according to our data availability, such as the PAH levels in other indoor microenvironments (offices, schools, and other public indoors) were assumed equal to the residential indoor levels. Though we applied a probabilistic approach to expand the ranges of parameters involved in the cancer risk model, the lack of information for pollution levels in other indoor environments may induce bias to the risk estimation results. In light of the constraints of the current study, there are a number of points to be addressed in our future study, including expanding the range and number of microenvironments to be investigated, carrying out long-term investigations, and so forth, to improve the representativeness of the dataset and accuracy of risk estimation results.

## 4. Conclusions

Spatial variations for level and characteristics of indoor $PM_{2.5}$-bound PAHs, and the associated inhalation cancer risks were investigated in four Chinese cities. Average indoor $PM_{2.5}$-bound Σ18PAH levels showed differences of magnitudes between cities, from 3.07 ng $m^{-3}$ in Hong Kong residences, to 176.27 ng $m^{-3}$ in Xi'an residences. Traffic emission was identified as a common source contributed to the indoor $PM_{2.5}$-bound PAHs levels for all cities, implied the significance of outdoor infiltration. City-specific sources including coal-fired power plant emission for Hong Kong and Guangzhou, industrial emission for Shanghai, and fuel consumption for domestic heating for Xi'an were also identified. Estimated risk levels for male were generally higher than those for female for all age groups, which would be mainly attributed to the differences on inhalation rates, as suggested by the results of sensitivity analysis. Besides inhalation rate, the influential factors towards cancer risk in our model were the exposure duration, indoor *TEQ*, and body weight, respectively. Residents in Xi'an were found exposed to considerable levels of risk in terms of both *TEQ* referencing to BaP and ILCR, which were beyond recommended levels. Due to the improving ambient air quality under recent policies on air quality management, indoor air quality which is strongly influenced by outdoor infiltration is also expected to be improved in recent future, and the inhalation exposure risks for particulate-associated PAHs are expected to be continuously reduced. However, for extreme cases such as the exposure levels for Xi'an residents in this study, it is recommended to employ air purifying measures such as air cleaners for reducing indoor inhalation risks during heavy pollution periods.

**CRediT authorship contribution statement**

**Zhuozhi Zhang**: Conceptualization, Methodology, Software, Formal Analysis, Investigation, Writing – Original Draft, Visualization. **Qi Yuan**: Formal Analysis, Writing – Review & Editing. **Meng Wang**: Formal Analysis, Writing – Review & Editing. **Tafeng Hu**: Investigation, Writing – Review & Editing. **Yu Huang**: Investigation. **Guangli Xiu**: Investigation. **Senchao Lai**: Investigation. **Yuan Gao**: Writing – Review & Editing. **Shun Cheng Lee**: Conceptualization, Funding Acquisition, Project Administration, Supervision, Methodology, Formal Analysis, Resources, Writing – Review & Editing.

**Data availability**

The authors confirm that the data supporting the findings of this study are available within the article and its supplementary material. Raw data that support the findings of this study are available from the corresponding author, upon reasonable request.

**Declaration of competing interest**

The authors declare that they have no known competing financial interest or personal relationship that could have appeared to influence the work reported in this article.

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# Tables

## Exposure and health risk assessment of $PM_{2.5}$-bound polycyclic aromatic hydrocarbons during winter at residential homes: A case study in four Chinese cities

Zhuozhi Zhang[1], Qi Yuan[1], Meng Wang[1], Tafeng Hu[2], Yu Huang[2], Guangli Xiu[3], Senchao Lai[4], Yuan Gao[5], Shun Cheng Lee[1*]

[1] Department of Civil and Environmental Engineering, The Hong Kong Polytechnic University, Hung Hom, Hong Kong, China

[2] State Key Laboratory of Loess and Quaternary Geology (SKLLQG), Key Laboratory of Aerosol Chemistry and Physics, Institute of Earth Environment, Chinese Academy of Sciences (IEECAS), Xi'an 710061, China

[3] School of Resources and Environmental Engineering, East China University of Science and Technology (ECUST), Shanghai 200237, China

[4] The Key Lab of Pollution Control and Ecosystem Restoration in Industry Clusters, Ministry of Education, School of Environment and Energy, South China University of Technology (SCUT), Guangzhou 510006, China

[5] Instrumentation and Service Center for Science and Technology, Beijing Normal University, Zhuhai 519087, China

[*] Corresponding author. Department of Civil and Environmental Engineering, The Hong Kong Polytechnic University, Hung Hom, Hong Kong, China.

Email address: shun-cheng.lee@polyu.edu.hk (S.C. Lee)

**Table 1**. Indoor $PM_{2.5}$ and $PM_{2.5}$-bound PAHs concentrations (in ng $m^{-3}$) in four cities.

| | Hong Kong | | | | Guangzhou | | | | Shanghai | | | | Xi'an | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | **Avg.** | **S.D.** | **Min** | **Max** | **Avg.** | **S.D.** | **Min** | **Max** | **Avg.** | **S.D.** | **Min** | **Max** | **Avg.** | **S.D.** | **Min** | **Max** |
| **$PM_{2.5}$** (in µg $m^{-3}$) | 34.0 | 14.6 | 10.0 | 76.8 | 47.2 | 5.43 | 39.6 | 55.4 | 50.3 | 17.9 | 18.9 | 139 | 78.7 | 49.3 | 31.0 | 225 |
| **FLU** | 0.43 | 0.18 | 0.11 | 0.80 | 0.12 | 0.03 | 0.07 | 0.16 | 1.02 | 0.66 | 0.29 | 2.97 | 1.90 | 1.74 | 0.13 | 7.99 |
| **PHE** | 0.11 | 0.04 | 0.03 | 0.21 | 0.99 | 0.27 | 0.56 | 1.33 | 1.01 | 0.58 | 0.28 | 2.89 | 3.99 | 3.36 | 0.32 | 13.2 |
| **ANT** | 0.04 | 0.02 | 0.01 | 0.16 | 0.13 | 0.04 | 0.08 | 0.21 | 0.30 | 0.20 | 0.09 | 1.01 | 1.12 | 0.94 | 0.08 | 3.82 |
| **FLT** | 0.32 | 0.13 | 0.08 | 0.65 | 0.97 | 0.27 | 0.56 | 1.45 | 0.35 | 0.21 | 0.10 | 0.94 | 9.28 | 7.77 | 0.61 | 29.3 |
| **PYR** | 0.21 | 0.09 | 0.05 | 0.41 | 1.02 | 0.32 | 0.61 | 1.67 | 0.34 | 0.20 | 0.10 | 0.98 | 10.3 | 8.71 | 0.71 | 34.0 |
| **BaA** | 0.09 | 0.04 | 0.02 | 0.19 | 0.96 | 0.38 | 0.53 | 2.08 | 0.20 | 0.12 | 0.06 | 0.56 | 9.95 | 8.42 | 0.69 | 33.4 |
| **CHR** | 0.33 | 0.15 | 0.07 | 0.89 | 1.70 | 0.50 | 1.01 | 2.69 | 0.42 | 0.25 | 0.12 | 1.05 | 19.2 | 16.5 | 1.43 | 63.0 |
| **B[b+j]F** | 0.25 | 0.24 | 0.06 | 1.60 | 1.68 | 0.60 | 1.05 | 2.99 | 0.86 | 0.50 | 0.26 | 2.12 | 15.2 | 12.9 | 1.21 | 51.0 |
| **BkF** | 0.25 | 0.16 | 0.06 | 1.09 | 1.67 | 0.45 | 1.00 | 2.53 | 0.49 | 0.28 | 0.16 | 1.19 | 9.36 | 7.74 | 0.61 | 31.7 |
| **BaF** | 0.06 | 0.03 | 0.01 | 0.15 | 0.31 | 0.09 | 0.18 | 0.47 | 0.05 | 0.03 | 0.01 | 0.12 | 13.6 | 12.1 | 0.86 | 57.1 |
| **BeP** | 0.25 | 0.15 | 0.06 | 1.00 | 1.24 | 0.54 | 0.70 | 2.81 | 1.22 | 0.72 | 0.35 | 3.03 | 18.8 | 17.2 | 1.18 | 67.7 |
| **BaP** | 0.15 | 0.09 | 0.03 | 0.54 | 0.69 | 0.19 | 0.39 | 0.95 | 0.43 | 0.25 | 0.13 | 1.11 | 10.4 | 9.11 | 0.75 | 34.5 |
| **PER** | 0.14 | 0.05 | 0.04 | 0.23 | 0.08 | 0.03 | 0.05 | 0.15 | 0.10 | 0.05 | 0.03 | 0.25 | 5.58 | 4.64 | 0.42 | 17.8 |
| **IcdP** | 0.15 | 0.08 | 0.03 | 0.54 | 2.01 | 0.64 | 1.16 | 3.41 | 0.37 | 0.24 | 0.11 | 1.27 | 10.1 | 8.38 | 0.65 | 32.3 |
| **BghiP** | 0.10 | 0.09 | 0.02 | 0.63 | 1.29 | 0.39 | 0.71 | 2.12 | 0.38 | 0.23 | 0.12 | 1.04 | 11.0 | 9.05 | 0.69 | 36.4 |
| **DahA** | 0.05 | 0.03 | 0.01 | 0.18 | 0.19 | 0.06 | 0.11 | 0.29 | 0.05 | 0.03 | 0.02 | 0.13 | 5.22 | 4.35 | 0.34 | 17.7 |
| **COR** | 0.07 | 0.04 | 0.02 | 0.29 | 0.44 | 0.13 | 0.24 | 0.66 | 0.22 | 0.14 | 0.06 | 0.60 | 7.68 | 6.38 | 0.49 | 24.8 |
| **DaeP** | 0.07 | 0.06 | 0.02 | 0.43 | 0.35 | 0.10 | 0.22 | 0.52 | 0.22 | 0.13 | 0.07 | 0.60 | 13.8 | 12.3 | 1.00 | 50.2 |
| ***Σ18PAHs*** [1] | 3.07 | 1.55 | 0.74 | 9.76 | 15.9 | 4.78 | 9.59 | 26.4 | 8.02 | 4.75 | 2.35 | 21.8 | 176 | 150 | 12.3 | 600 |
| ***ΣLMWPAHs*** [2] | 0.59 | 0.24 | 0.15 | 1.10 | 1.24 | 0.33 | 0.71 | 1.69 | 2.33 | 1.43 | 0.66 | 6.88 | 7.00 | 5.86 | 0.54 | 22.4 |
| ***ΣMMWPAHs*** [2] | 0.94 | 0.39 | 0.23 | 2.14 | 4.65 | 1.39 | 2.85 | 7.88 | 1.30 | 0.77 | 0.37 | 3.53 | 48.7 | 41.1 | 3.46 | 157 |
| ***ΣHMWPAHs*** [2] | 1.54 | 0.98 | 0.36 | 6.52 | 9.97 | 3.09 | 6.03 | 16.8 | 4.38 | 2.57 | 1.31 | 11.4 | 121 | 103 | 8.31 | 421 |
| ***ΣCPAHs*** [3] | 2.94 | 1.46 | 0.71 | 9.18 | 15.2 | 4.59 | 9.19 | 25.4 | 7.75 | 4.60 | 2.27 | 21.1 | 149 | 126 | 10.5 | 493 |
| ***ΣNCPAHs*** [3] | 0.13 | 0.09 | 0.03 | 0.58 | 0.66 | 0.19 | 0.40 | 1.00 | 0.27 | 0.16 | 0.08 | 0.71 | 27.4 | 24.2 | 1.86 | 107 |
| ***ΣComPAHs*** [4] | 1.85 | 1.00 | 0.43 | 6.54 | 12.0 | 3.57 | 7.15 | 19.9 | 3.83 | 2.25 | 1.15 | 10.3 | 105 | 87.8 | 7.40 | 343 |

[1]: Total detected PAHs.

[2]: Total PAHs by molecular weight groups: low molecular weight (LMW), medium molecular weight (MMW), and high molecular weight (HMW).
[3]: Total carcinogenic PAHs (CPAHs) and non-carcinogenic PAHs (NCPAHs) (Nisbet and LaGoy, 1992).
[4]: Total combustion-related PAHs (Ravindra et al., 2008a).

**Table 2**. Statistical summary of the indoor *TEQ*, daily exposures, ILCR, and LCR of indoor $PM_{2.5}$-bound CPAHs exposures.

| | | 5th [1] | Median | 95th [1] | Mean | S.D. [2] |
|---|---|---|---|---|---|---|
| ***TEQ*** (ng $m^{-3}$) | Hong Kong | 0.175 | 0.273 | 0.461 | 0.289 | 0.091 |
| | Guangzhou | 1.22 | 1.55 | 2.00 | 1.57 | 0.241 |
| | Shanghai | 0.378 | 0.637 | 1.21 | 0.695 | 0.270 |
| | Xi'an | 8.34 | 18.1 | 47.0 | 21.6 | 13.4 |
| | Type A | 0.121 | 0.235 | 0.591 | 0.280 | 0.165 |
| | Type B | 0.818 | 3.56 | 24.5 | 7.20 | 12.3 |
| | Type C | 1.10 | 4.42 | 27.5 | 8.34 | 13.1 |
| ***Daily Exposure*** (ng $day^{-1}$) | Hong Kong | 1.58 | 3.71 | 7.92 | 4.08 | 2.04 |
| | Guangzhou | 9.31 | 20.2 | 37.4 | 21.1 | 8.91 |
| | Shanghai | 3.99 | 9.54 | 21.2 | 10.7 | 5.57 |
| | Xi'an | 95.2 | 261 | 688 | 310 | 200 |
| | Type A | 0.955 | 2.39 | 6.66 | 2.92 | 2.02 |
| | Type B | 7.28 | 35.6 | 261 | 75.1 | 137 |
| | Type C | 9.73 | 44.3 | 294 | 86.9 | 146 |
| ***ILCR*** ($\times 10^{-8}$) | Hong Kong | 0.0736 | 0.759 | 4.45 | 1.24 | 1.50 |
| | Guangzhou | 0.403 | 4.09 | 22.7 | 6.42 | 7.34 |
| | Shanghai | 0.174 | 1.80 | 10.9 | 3.03 | 3.73 |
| | Xi'an | 4.74 | 51.0 | 316 | 88.5 | 114 |
| | Type A | 0.0434 | 0.483 | 2.77 | 0.813 | 1.02 |
| | Type B | 0.458 | 7.33 | 74.2 | 19.1 | 39.0 |
| | Type C | 0.596 | 9.10 | 86.0 | 22.4 | 43.5 |
| ***LCR*** ($\times 10^{-8}$) | Hong Kong | 0.225 | 2.10 | 7.73 | 2.89 | 2.49 |
| | Guangzhou | 1.24 | 11.1 | 38.9 | 15.0 | 12.4 |
| | Shanghai | 0.522 | 5.06 | 19.0 | 7.01 | 6.17 |
| | Xi'an | 14.4 | 150 | 572 | 207 | 188 |
| | Type A | 0.145 | 1.49 | 5.28 | 1.96 | 1.73 |
| | Type B | 1.59 | 27.0 | 152 | 45.9 | 61.1 |
| | Type C | 2.05 | 32.9 | 174 | 53.9 | 68.4 |

[1]: $i^{th}$ percentile of the dataset.

[2]: Standard deviation.

**Figures**

**Exposure and health risk assessment of $PM_{2.5}$-bound polycyclic aromatic hydrocarbons during winter at residential homes: A case study in four Chinese cities**

Zhuozhi Zhang[1], Qi Yuan[1], Meng Wang[1], Tafeng Hu[2], Yu Huang[2], Guangli Xiu[3], Senchao Lai[4], Yuan Gao[5], Shun Cheng Lee[1*]

[1] Department of Civil and Environmental Engineering, The Hong Kong Polytechnic University, Hung Hom, Hong Kong, China

[2] State Key Laboratory of Loess and Quaternary Geology (SKLLQG), Key Laboratory of Aerosol Chemistry and Physics, Institute of Earth Environment, Chinese Academy of Sciences (IEECAS), Xi'an 710061, China

[3] School of Resources and Environmental Engineering, East China University of Science and Technology (ECUST), Shanghai 200237, China

[4] The Key Lab of Pollution Control and Ecosystem Restoration in Industry Clusters, Ministry of Education, School of Environment and Energy, South China University of Technology (SCUT), Guangzhou 510006, China

[5] Instrumentation and Service Center for Science and Technology, Beijing Normal University, Zhuhai 519087, China

[*] Corresponding author. Department of Civil and Environmental Engineering, The Hong Kong Polytechnic University, Hung Hom, Hong Kong, China.

Email address: shun-cheng.lee@polyu.edu.hk (S.C. Lee)

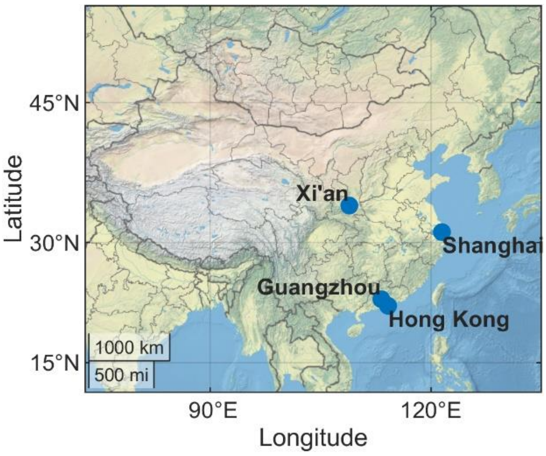


**Fig. 1**. Locations of investigated four cities.

(Fig. 1 is a one-column figure)

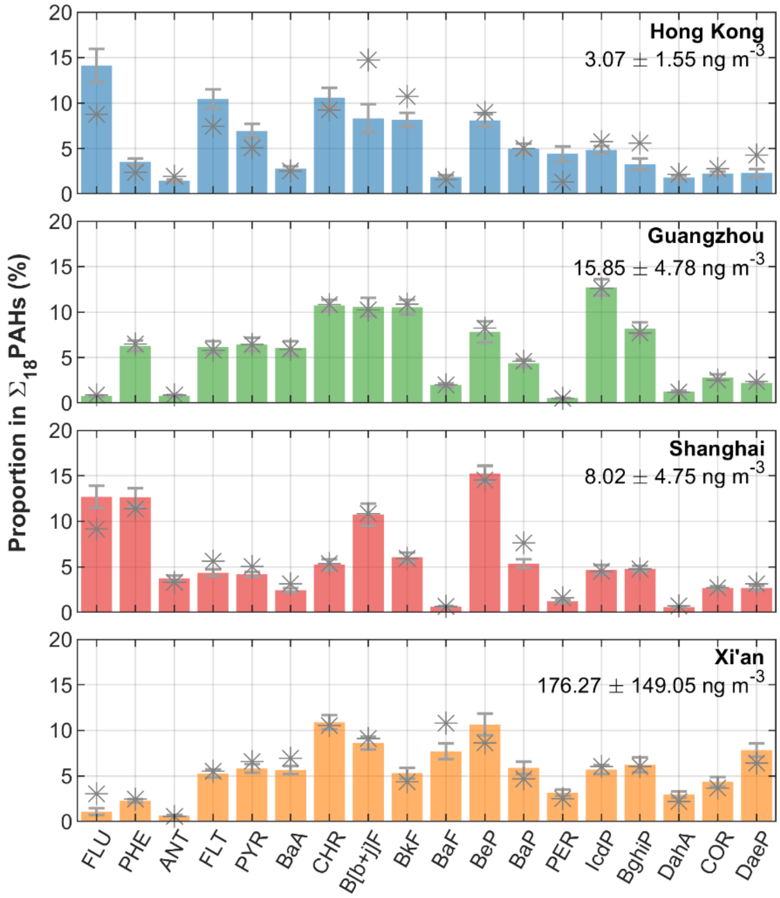


**Fig. 2**. Profiles of individual PAH species contributions to $\Sigma_{18}$PAHs in $PM_{2.5}$ (bars, error bars, and concentrations are for PAHs in indoor $PM_{2.5}$, and ✳ marks represent the PAHs in outdoor $PM_{2.5}$)

(Fig. 2 is a 1.5 column figure)

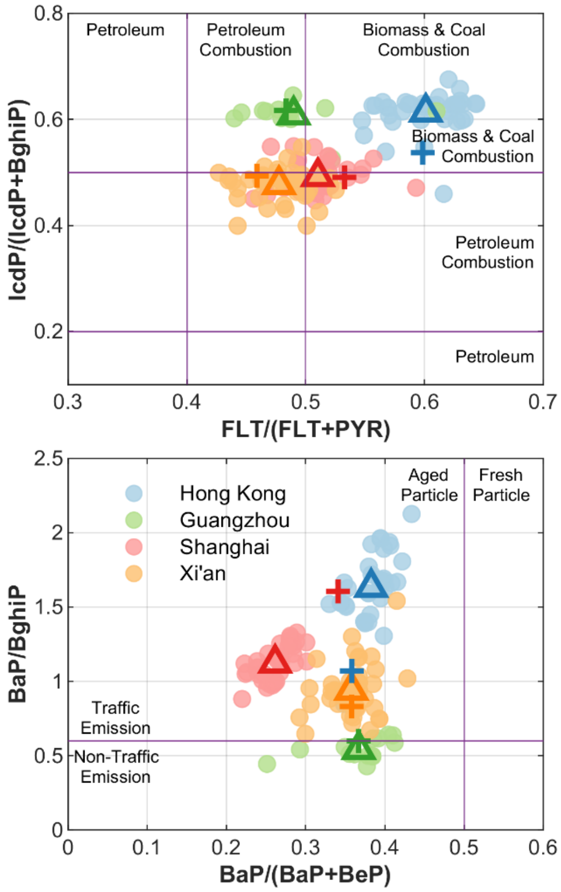


**Fig. 3**. Molecular diagnostic ratios of $PM_{2.5}$-bound PAHs.

(Fig. 3 is a one-column figure)

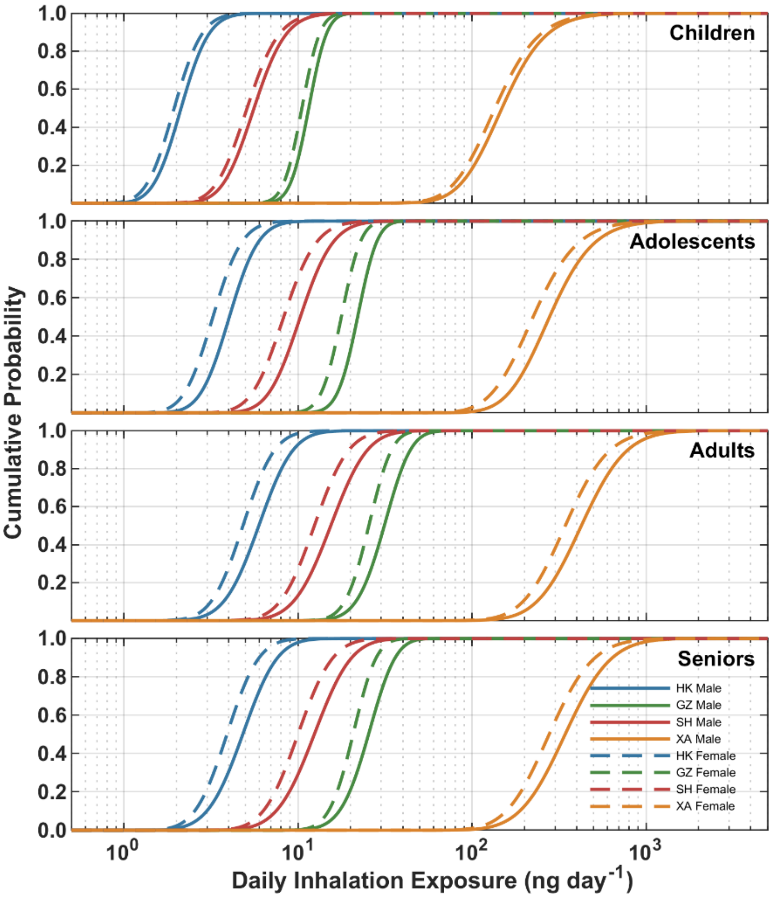


**Fig. 4**. Probability distribution of the daily inhalation exposure of $PM_{2.5}$-bound CPAHs in terms of *TEQ* for different age groups.

(Fig. 4 is a 1.5 column figure)

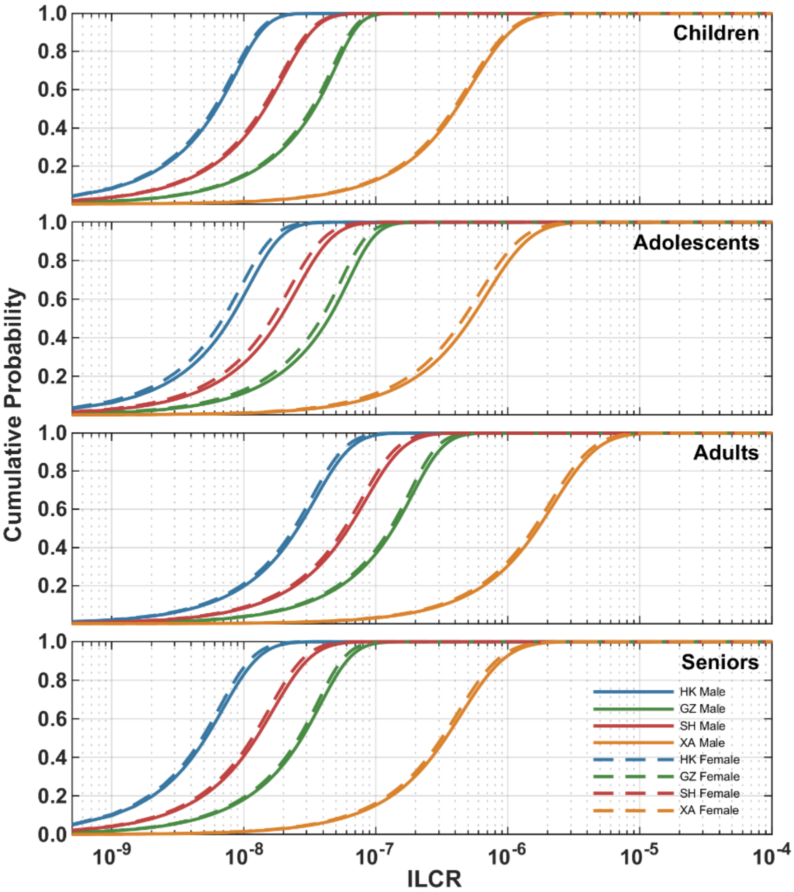


**Fig. 5**. Probability distribution of the ILCR related to inhalation exposure of ΣCPAHs in $PM_{2.5}$ of different age groups.

(Fig. 5 is a 1.5 column figure)

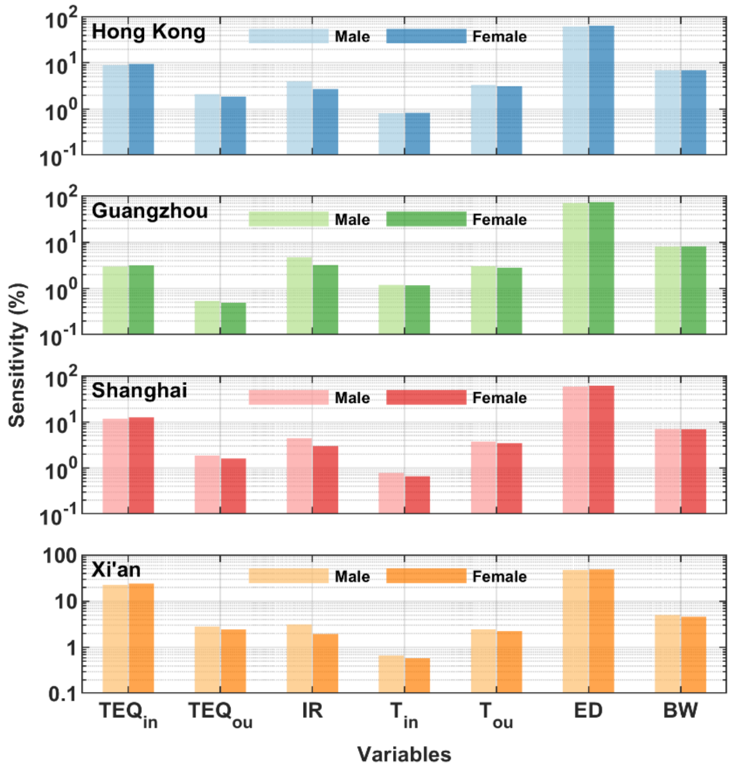


**Fig. 6**. Sensitivity analysis results on ILCR related to inhalation exposure of ΣCPAHs in $PM_{2.5}$ (subscripts *in* and *ou* represent the exposure levels (*TEQ*) or time spent (*T*) in indoor and outdoor respectively; *IR* represents inhalation rate; *ED* represents exposure duration; and *BW* represents body weight, respectively).

(Fig. 6 is a 1.5 column figure)

## CRediT authorship contribution statement

**Zhuozhi Zhang**: Conceptualization, Methodology, Software, Formal Analysis, Investigation, Writing – Original Draft, Visualization. **Qi Yuan**: Formal Analysis, Writing – Review & Editing. **Meng Wang**: Formal Analysis, Writing – Review & Editing. **Tafeng Hu**: Investigation, Writing – Review & Editing. **Yu Huang**: Investigation. **Guangli Xiu**: Investigation. **Senchao Lai**: Investigation. **Yuan Gao**: Writing – Review & Editing. **Shun Cheng Lee**: Conceptualization, Funding Acquisition, Project Administration, Supervision, Methodology, Formal Analysis, Resources, Writing – Review & Editing.