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Age- and sex-specific dermal exposure of polycyclic aromatic hydrocarbons in the general population of a city in south China☆

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A B S T R A C T

This study assessed the dermal exposure of population to polycyclic aromatic hydrocarbons (PAHs) in a South China city. Skin wipe samples of the face, hand, forearm, and shank were collected from 120 volunteers (50% male and 50% female) belonging to different age groups (preschooler, thresholder, middle-aged, and elderly). Concentrations of PAHs in the skin wipe samples varied from 18 to 27000 ng/m^2 in the order of face > hand > forearm > shank, regardless of age and gender. The PAH concentrations of bare skin locations were significantly higher in females than in males, while no significant differences were observed for clothing-covered skin locations between genders. The PAH concentrations for faces were significantly higher in the elderly compared to the other groups. The PAH composition was distinct between the four age groups. The dermal exposure levels of total PAHs and total BaP equivalent concentration (BaP$_{eq}$) varied from 25.6 to 620 and 0.093–37.4 ng/kg body weight/d, respectively. The dermal exposure levels of total PAHs were significantly higher in females than in males in all age groups except for the middle-aged group. The hand-mouth exposure doses were significantly higher in the preschoolers than in the other age groups. The values of the carcinogenic risk caused by dermal PAH exposure were between 3.5×10^{-6} and 1.4×10^{-3} with 29% of the population (35/120) having risk values exceeding significant levels (1×10^{-4}). The thresholder group exhibited the highest risk for PAH dermal exposure among all groups of the population. This study provides a comprehensive evaluation of the age- and gender-related risk of PAH through dermal exposure.

1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) have been recognized as one of the primary organic pollutants affecting human health due to their genetic toxicity, mutagenicity, and carcinogenicity (Yang et al., 2015; Matos et al., 2021; Dobaradaran et al., 2020; Dat and Chang, 2017), including damage to the respiratory, circulatory, and nervous system (Nam et al., 2021). Due to the threat to public health, the European Environment Agency and the US Environmental Protection Agency (EPA) have designated PAHs as priority pollutants (Ramirez et al., 2011). Polycyclic aromatic hydrocarbons are ubiquitous in various environmental media, including the atmosphere, soil, water, and sediments (Sei et al., 2021; Dong et al., 2016; Krzyszczak and Czech, 2021).

Human exposure to PAH occurs mainly by inhalation, diet, and dermal contact. Although inhalation has been assumed to be the primary route of exposure to environmental PAHs (Raymond, 1998), exposure through dermal contact is increasingly being considered (Tsai et al., 2001; Cao et al., 2020; Lao et al., 2018). Lao et al. (2018) reported dermal absorption as a more important pathway than inhalation for intake of low molecular-weight PAHs. Several studies also reported the dermal uptake from airborne organic particles such as low molecular

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weight plasticizers and flame retardants as an important route of human exposure to pollutants (Wu et al., 2016; Abdallah et al., 2015; Abdallah et al., 2016; Tang et al., 2021). These results highlight the importance of assessing the dermal absorption of organic pollutants as a significant exposure route.

Recently, several studies were conducted to assess the risk of human exposure to PAHs via dermal contact (Sjöström et al., 2019; Wang et al., 2021; Stec et al., 2018). Skin site-specific exposure was investigated in these studies. However, all these studies were conducted in populations that included firefighters and steel and iron manufacturing industry workers, where occupational exposure likely occurred. To the best of our knowledge, limited information is available for skin site-specific dermal absorption of PAH among the general population. Moreover, no information on the dermal absorption and health risk of PAH with respect to different age groups is available. The work location, lifestyle, and hygienic habits are all age specific, and are expected to influence the degree of exposure. Previous studies mostly focused on the difference between children and adults; therefore, a knowledge gap exists for re-assessing the PAH exposure and risk for general populations based on these factors.

In the present study, skin wipe samples from different body locations (the face, hand, forearm, and shank) were collected from individuals in four different age groups (preschooler: 5–6-year-olds, thresholder: 17–23-year-olds, middle-aged: 32–50-year-olds, and elderly: 70–100-year-olds). The PAH concentrations in the abovementioned skin locations were determined and the dermal uptake and health risk were assessed. The present study aims to explore the skin site-specific dermal uptake of PAHs among a population of different age groups and genders.

2. Materials and methods

2.1. Sampling

The samples were collected in Mao-Ming, China, from December 8 to 21, 2020. A total of 120 volunteers (60 males, 60 females) belonging to different age groups (preschooler, thresholder, middle-aged, and elderly) were considered. The preschoolers were from a kindergarten, the thresholder group and middle-aged group comprised students and staff at Guangdong University of Petrochemical Technology and the elderly were from a nursing home in Maoming city. Skin wipes were collected from the face, hands, forearm, and shank of each participant. A short questionnaire, covering age, gender, daily activities, and details of any skincare product used, was completed by each participant. All participants were required not to wash the skin sampling locations at least 2 h prior to sampling. All participants were required to give informed consent prior to sampling, and ethical approval for this investigation was obtained from the Research Ethics Committee of Guangzhou Institute of Geochemistry.

2.2. Sample analysis

All skin wipe samples were spiked with internal standards (Nap-d8, Acp-d10, Phe-d10, Chr-d12, and Per-d12, each with 100 ng). Then the samples were placed in a 20 mL solvent solution (n-hexane and acetone, 3:1, v/v) and were extracted in an ultrasonic bath for 20 min. The extraction was repeated 3 times. The extract was then concentrated to ~1 mL using a rotary evaporator. The extracts were quantitatively transferred to silica solid-phase cartridges (Poly-Sery SPE, 1 g weight, 6 mL, ANPEL Laboratory Technologies (Shanghai) Inc.), pre-washed with 10 mL hexane, and then eluted with 12 mL of hexane/dichloromethane (1:1, v/v). The volume of the extracts was condensed to 1 mL, and the extracts were evaporated to dryness under nitrogen before being reconstituted with 100 μL iso-octane. Prior to instrumental analysis, 100 ng of the recovery standards (2-Fluorobiphenyl and p-Terphenyl-d14) were added to the final solution.

The content of sixteen PAH compounds (Table 1) was determined using a gas chromatograph (GC) with a mass spectrometer (MS) (SHIMADZU GC-MS-QP2020NX) operated in electron ionization mode. This GC/MS was equipped with a Rtx-5 MS capillary column (30 m × 0.25 mm × 0.25 μm , SHIMADZU). The injection volume was 1 μL (split less mode). The injection and the ion source temperatures were 290 °C and 230 °C, respectively. The temperature program for the GC system was 80 °C for 5 min, after which it was increased to 310 °C at a rate of 4 °C/min. It was then maintained at this temperature for 15 min. The mass of primary and secondary ions, used for quantitative and qualitative analyses of PAHs, was determined using selected ion monitoring (SIM).

2.3. Quality assurance and quality control

Analysis of the field blank, including wipes and silica cartridges showed no traces of targets. Spiked recovery values were obtained by spiking 20 ng standards of 16 PAHs into soaked gauze wipes (3 replicates) and solvent (3 replicates). The analysis of the wipes was done using the same procedure used for the samples. The recoveries of PAHs

Table 1
PAH concentrations (median and range) in 4 selected body surface areas.

	Preschooler (5–6 years old)		Thresholder (17–23 years old)		Middle-aged (32–50 years old)		Elderly (70–100 years old)	
	Male	Female	Male	Female	Male	Female	Male	Female
PAH concentration in selected skin (ng/m^2)								
Face	3000 (720–11000)	82,000 (22,000-21000)	5000 (2800–27000)	10,000 (5400–20000)	6100 (2100–10000)	5300 (3300–24000)	5900 (1900–13000)	11,000 (2300–36000)
Hand	790 (280–1800)	890 (390–2100)	490 (210–1600)	720 (340–17000)	500 (180–13000)	920 (220–5400)	790 (130–1600)	870 (160–32000)
Forearm	280 (90–1100)	240 (100–850)	250 (60–780)	350 (170–910)	230 (100–810)	330 (130–880)	260 (53–520)	230 (130–1500)
Shank	160 (37–320)	120 (53–670)	69 (44–510)	140 (18–360)	100 (36–170)	110 (40–300)	160 (50–290)	69 (29–1410)
PAH dermal exposure levels (ng/kg/d)								
Face	90 (19–290)	270 (90–580)	133 (81–322)	95 (25–176)	92 (45–180)	110 (21–152)	92 (22–328)	172 (31–318)
Hand	4.8 (1.8–12)	8.2 (4.1–15)	7.1 (2.5–15)	3.8 (0.81–112)	4.1 (0.88–17)	2.7 (1.1–9.5)	3.7 (1.8–8.1)	4.2 (0.39–10)
forearm	4.4 (1.3–20)	6.5 (3.2–14)	8.3 (4.1–23)	4.2 (0.6–26)	5.2 (1.2–13)	4.4 (0.85–13)	3.6 (1.9–18)	3.7 (1.9–18)
others	9.5 (2.5–23)	16 (3.5–30	20 (3.0–37)	7.3 (2.9–27)	7.9 (1.7–20)	6.5 (1.8–27)	9.8 (4.8–17)	5.4 (2.5–103)
Total PAH	120 (26–320)	300 (120–620)	178 (111–370)	114 (34–195)	113 (51–215)	119 (30–185)	110 (36–344)	203 (37–330)
Total BaP_{eq}	1.0 (0.18–3.3)	2.8 (1.1–9.5)	5.0 (0.98–20)	2.2 (0.63–29)	1.4 (0.31–4.0)	0.80 (0.093–4.4)	0.67 (0.15–6.6)	0.98 (0.16–37)
Hand-mouth exposure levels (ng/kg/d)								
Total PAH	23 (9.5–51)	26 (11–67)	1.8 (0.92–4.2)	1.1 (0.38–3.7)	2.1 (0.50–13)	0.96 (0.34–2.8)	2.1 (0.34–3.9)	2.0 (0.41–7.8)
Total BaP_{eq}	0.84 (0.23–3.3)	1.2 (0.27–5.1)	0.11 (0.0089–0.75)	0.068 (0.0051–0.90)	0.040 (0.0070–3.5)	0.055 (0.0020–0.36)	0.091 (0.0018–1.0)	0.098 (0.0064–0.40)

varied between 50 and 118% for the spiked blank and between 58 and 112% for the spiked matrices. The average recoveries were 39 ± 13% for Nap-d8, 64 ± 7.9% for Acp-d10, 82 ± 11% for Phe-d10, 84 ± 10% for Chr-d12, and 79 ± 13% for Per-d12. The standard deviations were between 1.4 and 6.4%. The data of nap was not used considering of the low recovery of Nap-d8. The limit of detection of PAHs was 0.08–6.6ng/wipe.

2.4. Data analysis

2.4.1. Calculating PAH concentrations through dermal exposure

We assumed that the PAH levels in the clothing-covered skin areas were similar to those of the shank. The penetration coefficient model (Liu et al., 2017b) was adopted in this study to determine the dermal absorption dose (DAD) using the following equation:

$$\mathrm{DAD} = \frac{\left(\frac{C_{face}\times A_{face}+C_{hand}\times A_{hand}+C_{arm}\times A_{arm}+C_{others}\times A_{others}}{l_m}\right)\times k_{p-l}\times ED}{Bodyweight}$$

where, C_{face}, C_{hand}, C_{arm}, and C_{others} are the area-based concentrations ($\mathrm{ng/m^2}$) of PAHs on the face, hand, arm, and other clothing-covered skin surfaces, respectively. A_{face}, A_{hand}, A_{arm}, and A_{others} are the skin surface area ($\mathrm{m^2}$) of the face, hand, arm, and other clothing-covered skin, respectively. The skin surface was calculated using the formula for people of China proposed by Yu et al. (2003). L_{m} is the thickness of skin lipid, which was assumed to be 1.3 μm for adults and 0.88 μm for children (Nazzaro-Porro et al., 1979). $K_{\mathrm{p\text{-}l}}$ is the permeability coefficient of chemicals (μm/h) from skin lipids into dermal capillaries. $K_{\mathrm{p\text{-}l}}$ is calculated using the method proposed by Weschler and Nazaroff (2012) (Table S1 in Support Information). ED (h/d) is the exposure duration, which is 24 h.

The hand-to-mouth contact exposure dose was calculated using the following equation proposed by Stapleton et al. (2008):

$$\mathrm{DAD_{oral}} = \frac{C_{hand}\times A_{hand}\times TE\times SAC\times EF}{Bodyweight}$$

where, C_{hand} and A_{hand} are the concentrations of PAH in the hand skin and the surface area of the hand, respectively. TE is the transfer efficiency (70%, mass fraction of the chemical transferred per contact), SAC is the percentage of hand area per contact (10%), and EF is the frequency of contact (24/day for adult and 216/day for children) (Stapleton et al., 2008).

2.4.2. Skin cancer risk assessment

The skin cancer risk caused by dermal exposure to PAHs was calculated using the following equation (USEPA, 2009):

$$\mathrm{Risk} = \mathrm{DAD} \times \mathrm{CSF}$$

where, DAD is the Daily Intake Dose of PAHs and CSF is the Cancer Slope Factor for PAHs. BaP equivalent concentration ($\mathrm{BaP_{eq}}$) rather than total PAH concentration was used to calculate the risk. BaP is the most carcinogenic PAH compound. The $\mathrm{BaP_{eq}}$ was calculated by multiplying the PAH concentration with its toxic equivalent factor (TEF) (Table S1). The values of 37.47 mg/kg/day and 23.5 mg/kg/day were proposed as two CSF values for BaP dermal contact exposure based on the incidence of skin tumors in mice (Schmahl et al., 1977) and the gastrointestinal absorption factor of 0.31 (RAIS, 2022), respectively. The EPA recommended a CSF value of 12.0 mg/kg/day for ingestion exposure (Zartarian et al., 2005). However, these studies do not provide recommended value of CSF for BaP dermal contact exposure. A CSF value 37.47 mg/kg/day, recommended for low-dose exposures such as environmental exposures, was used in our study to estimate carcinogenic risk for dermal exposure (Hussain et al., 1998).

3. Results and discussions

3.1. Variation in PAH concentrations in different skin locations with respect to age and gender

The statistical description of PAH concentrations in four skin locations among individuals from different age groups of a population are provided in Table 1. The PAH concentrations in the skin wipe samples varied from 29 to 27000 $\mathrm{ng/m^2}$. Regardless of gender and age-group, PHA concentrations in the four skin locations exhibited the following order: face > hand > forearm > shank (Fig. 1, ANOVA, $p < 0.0001$), indicating higher concentrations of PAH on bare skin than on clothing-covered skin. This was expected given the garments' ability to protect against airborne particles, which reduces particle deposition and dust adhesion on the skin (Licina et al., 2019; Saini et al., 2016). The PAH concentrations from the forearm were higher than those from the shank which could be attributed to more air-exposure chance for the forearm than the shank in the winter.

Sjöström et al. (2019) reported the PAH concentrations of the wrists and collarbone area of firefighters. The concentrations of 32 PAHs were between 20,000 and 160,000 $\mathrm{ng/m^2}$ and between 20,000 and 46,000 $\mathrm{ng/m^2}$ for the wrist and collarbone areas, respectively. Stec et al. (2018) reported PAH levels of the back and front of the neck, jaw, and hands pre- and post-training exercise for firefighters. The PAH concentrations were up to 550 $\mathrm{mg/m^2}$ and followed in the order of hand > neck > jaw. As expected, skin PAH levels were higher in these firefighters than in the general population in the present study. However, it is hard to compare the skin site distribution of PAHs among different groups due to different sample collection sites.

The levels of halogenated flame retardants, polychlorinated biphenyls (Liu et al., 2017a; Cao et al., 2019), and phthalates (Gong et al., 2016) in similar skin sites were reported in previous studies, which make it possible to compare the skin site distribution between PAH and these man-made chemicals. These chemicals were also found in higher concentrations on bare skin than on clothing covered skin. However,

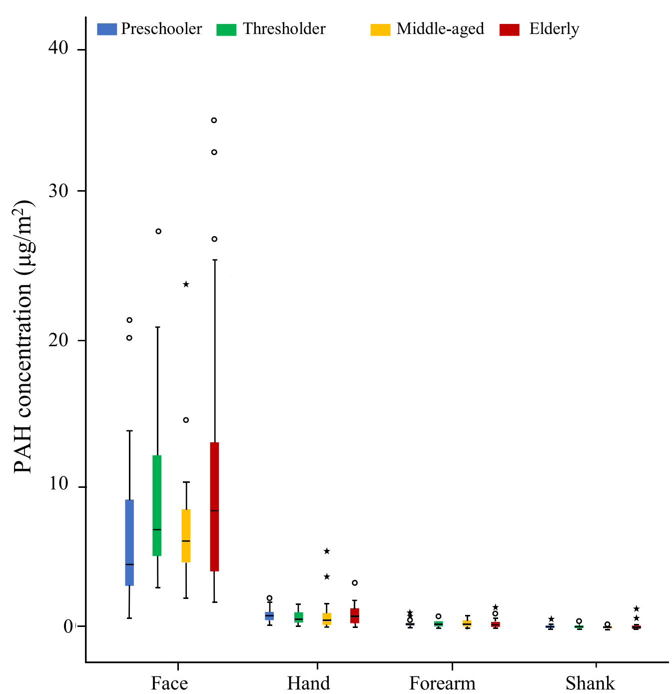

Fig. 1. PAHs concentrations in different skin sites form different age group samples. (The lower and upper ends of the box are the 25th and 75th percentiles of the data, respectively; the median is shown as the middle line, circle and asterisk represent outliers.)

previous studies have reported higher concentrations of the above-mentioned chemicals in samples from the hands than those from the face, in contrast with what was observed in the present study. The PAH concentrations from the face wipes in the present study were one to two orders of magnitude higher than those from the hand wipes (Table 1, Fig. 1). Halogenated flame retardants and phthalates are man-made chemicals commonly used as flame retardants and/or plasticizers in consumer products. The mass of these chemicals can reach up to 30% of the weight of products. The frequent hand contact with indoor surfaces or products would increase the levels of these chemicals in the skin of the hands. However, this is not the case for PAHs, since PAHs are not a component of any consumer products.

The PAH concentrations collected from the faces of the elderly group were significantly higher than those of the preschooler and middle-aged groups ($p < 0.05$), and those of the thresholder group with no statistical significance ($p > 0.05$). In other skin locations, there was no significant difference among the different age groups ($p > 0.05$) (Fig. 1). The higher PAH concentrations for the samples from the face in the elderly group could be attributed to the low frequency of face washing by elderly people in the nursing home. Gender based differences in halogenated organic pollutant concentrations on skin were reported by Cao et al. (2019). For relatively low volatile halogenated organic pollutants such as decabromodiphenyl ether and decabromodiphenyl ethane, males exhibited higher concentrations than females, whereas females showed higher concentrations of relatively highly volatile chemicals such as polychlorinated biphenyl (PCB) and pentabromodiphenyl ether (penta-BDE) mixtures. In the present study, females had significantly higher PAH concentrations in the face and hand samples, in comparison to those of the males, whereas no significant differences in PAH concentrations were observed in samples from the forearm and shank were observed between the two genders (Fig. 2). This result was consistent with the findings of Cao et al. (2019). The difference in usage of personal care products could be a reason for the observed gender-based differences in PAH on the bare skin. Twenty percent of the male volunteers used skincare products, in comparison to the 53% of female who did. Morrison et al. (2015) reported that the use of personal care products can increase the partitioning of gaseous-phase semi volatile organic compounds (SVOCs) and deposition of airborne particles onto the human skin surface, which may have resulted in higher PAH concentrations found for females.

3.2. Composition of PAHs in different skin locations with respect to age and gender

To gain insight into the variation of PAH composition among the different age groups, PAHs were sorted into three categories: the low molecular weight (LMW : two- and three-ringed PAHs), middle molecular weight (MMW : four-ringed PAHs), and high molecular weight (HMW : five-, and six-ringed PAHs). The proportions of the three categories are shown in Fig. 3. No significant differences in the PAH composition were found among four skin locations for a given age group. Thus, all data for the four locations were combined to perform analyses for the different age groups. The location of PAH composition in ternary exhibited age-specific variation, although large variation existed within a given age group as well (Fig. 3). The preschooler group had significantly lower abundance of the LMW (28 ± 13%) but higher abundance of the MMW (48 ± 20%) than those of other age groups (48 ± 20%–55 ± 16% for the LMW and 24 ± 14%–34 ± 19% MMW, $p < 0.001$). The abundance of the HMW (25 ± 17%) was also significantly higher in the preschooler group than that in the middle-aged group (14 ± 10%) and elderly group (18 ± 14%), with no significant difference with that of the thresholder group (21 ± 15%). The thresholder group exhibited significantly lower abundance of the MMW (24 ± 14%) than that of the other groups ($p < 0.001$) and the middle-aged group exhibited significantly lower abundance of the HMW (14 ± 10%) than those of the preschooler and the thresholder groups ($p < 0.001$).

The residential and workplaces of the age groups were different; therefore, the PAH exposure scenario would be different for the different age groups. Thus, it was reasonable to observe significant difference in PAH composition among different age groups. Unfortunately, the PAHs in the surrounding environment such as air and dust for different person was not measured in the present study, making it difficult to validate the above statement.

No gender-based differences in PAH composition were found for all age groups except for the preschooler group. The PAH composition in girls exhibited significant difference from those of boys in samples from the hand, forearm, and shank (Fig. 4). The percentage of HMW PAH in the girls (32 ± 18% from the hand, 37 ± 18% from the forearm, and 41 ± 21% from the shank) was significantly higher than that in the boys (15 ± 7.4% from the hand, 12 ± 5.3% from the forearm, and 14 ± 6.9% from the shank). The surrounding environment for the boys and girls was the same during the daily activities. Such difference between girls and boys was unexpected, and an explanation of this is beyond the scope of the present study.

3.3. Dermal exposure levels and hand-mouth exposure levels

The dermal exposure levels of total PAHs and BaP_{eq} among the eight subgroups are summarized in Table 1. Total dermal PAH and total BaP_{eq} exposure levels varied from 26 to 620 ng/kg/d and from 0.093 to 37 ng/kg/d, respectively. The dermal exposure levels were significantly higher in females than in males (ANOVA, $p < 0.01$) in all the age groups except

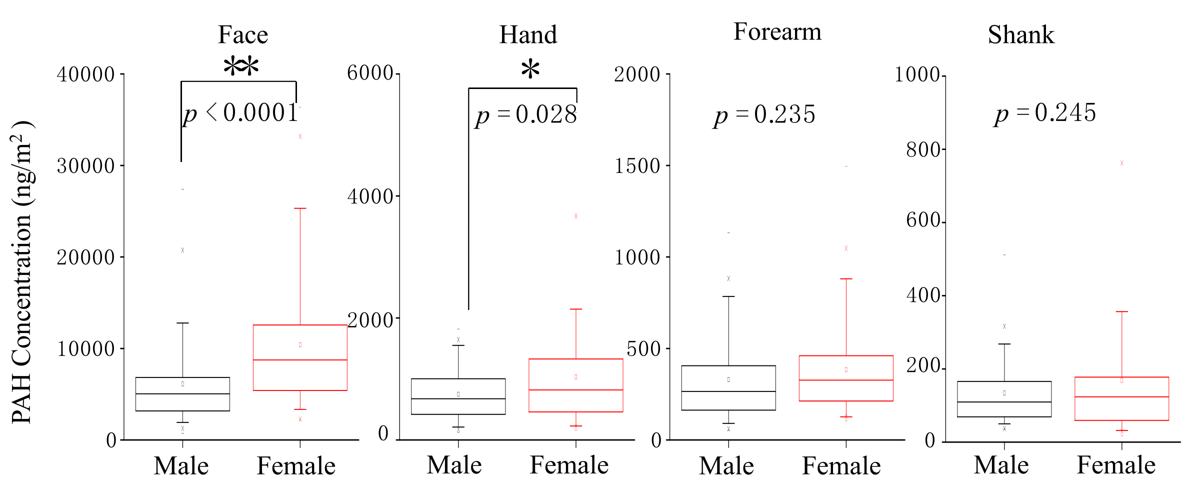

Fig. 2. PAHs concentrations in different skin sites between male and female.

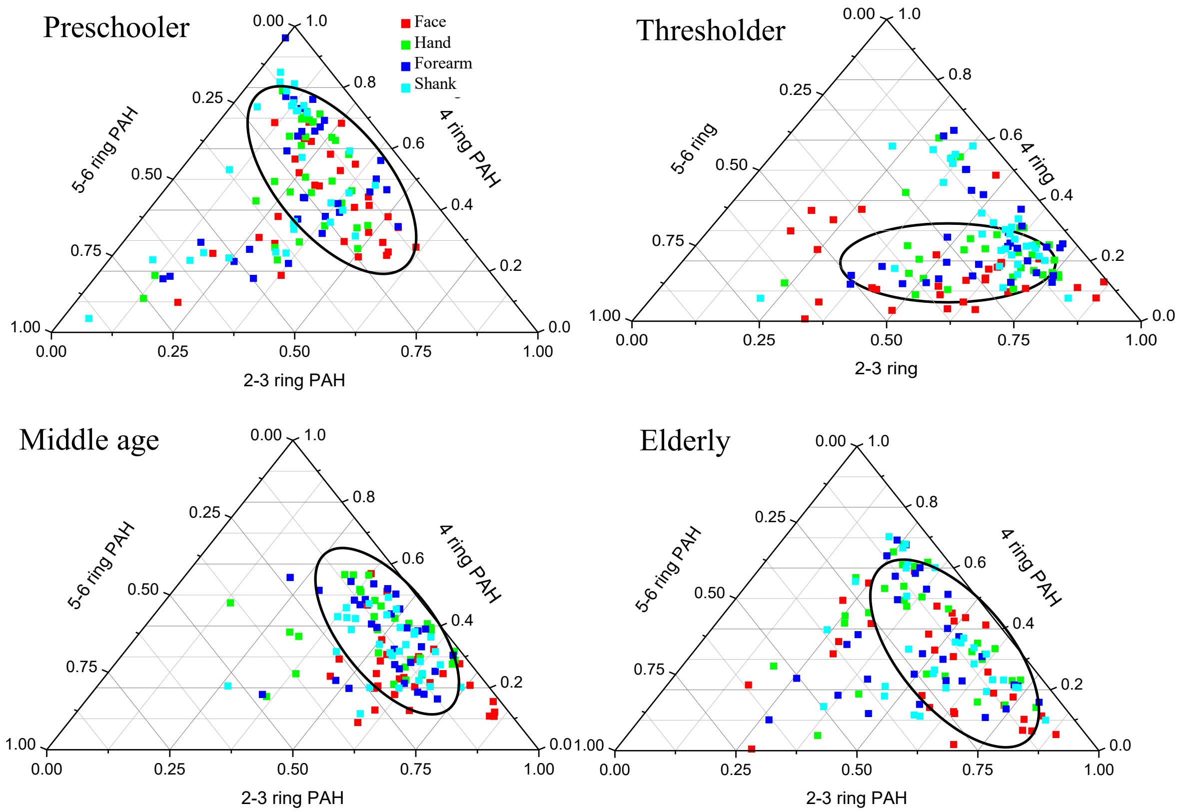

Fig. 3. PAH composition in different age-groups.

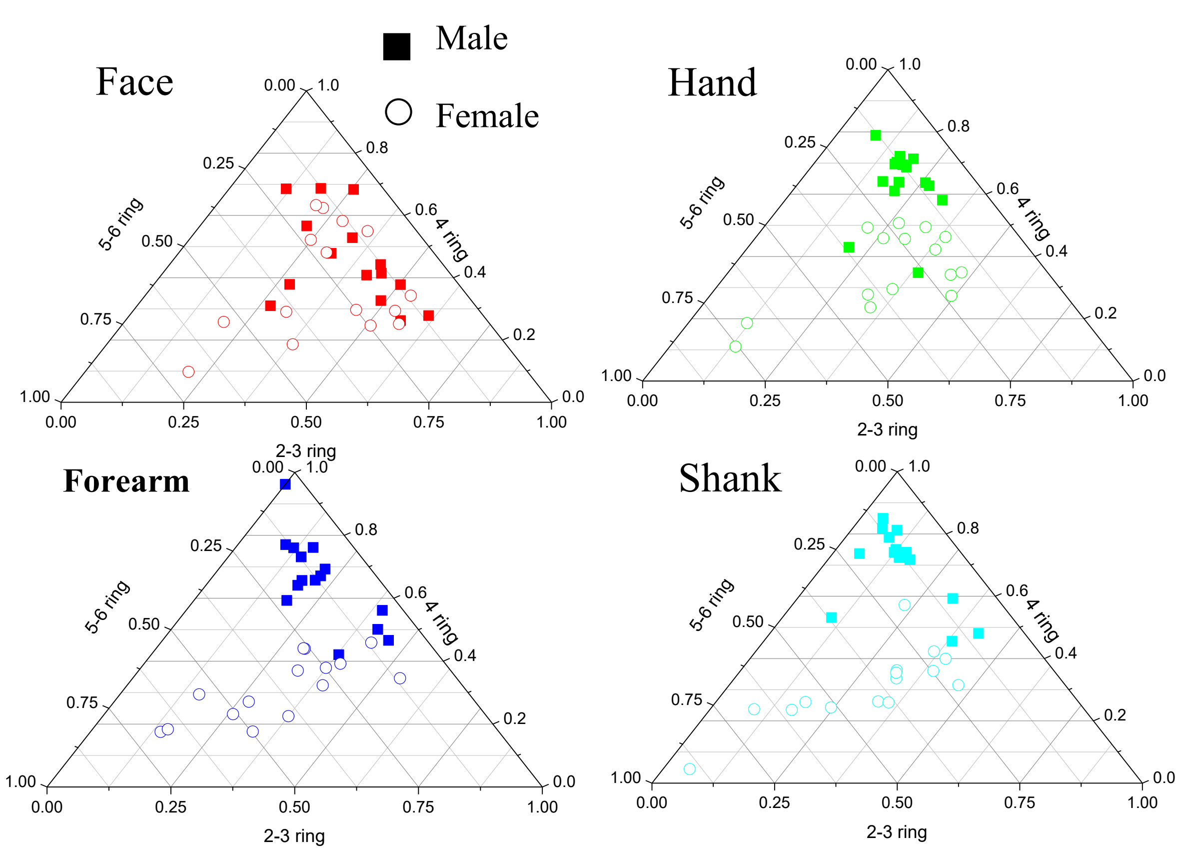

Fig. 4. PAH composition of different skin sites in preschooler groups.

for the middle-aged group ($p = 0.59$). Thus, the differences among age groups were analyzed based on gender. The thresholders exhibited the highest average dermal PAH exposure level (190 ng/kg/d), which was significantly higher than those for middle-aged and elderly groups for the males ($p < 0.05$), which could be attributed to the high levels of BaP in the skank samples of the thresholders. The dermal PAH exposure levels were significantly higher in the preschooler group than in the other three groups ($p < 0.001$) for the females.

The PAH absorption from the skin of the head was found to be the main contributor to the total PAH dermal exposure, accounting for an average of 80–89% for the eight subgroups. As the head PAH concentrations were higher in females than in males, the dermal PAH exposure levels were found to be higher in females than in males. The higher contribution from uncovered surface areas in comparison to surfaces covered by clothing were reported for PCBs and PBDEs in a study by Cao et al. (2019), which was inconsistent with the finding of the present study on PAHs. The difference in skin site distribution between PAH and PCBs/PBDEs was the main cause for this observation.

Hand-mouth exposure levels varied from 0.34 to 67 ng/kg/d for total PAHs and from 0.0018 to 5.1 ng/kg/d for total BaP_{eq}, which was one order of magnitude lower than the dermal exposure levels. The hand-mouth exposure levels in the preschooler group were significantly higher than in other groups. This was expected since the frequency of hand-mouth contact was higher in the preschoolers than in other groups. No significant differences were found between males and females in terms of the hand-mouth exposure levels.

Several studies have reported inhalation PAH exposure levels in the general population in other cities in China and other countries. Xia et al. (2013) used passive air samplers to collect gas and particulate phase PAHs between 2009 and 2010 in Taiyuan, China, and the average BaP_{eq} of 15 PAHs were 9.8 and 6.6 ng/kg/d for children and adults, respectively. Yu et al. (2015) modeled the average BaP_{eq} of 15 PAHs for the population of Beijing, China, and the different age groups had an average BaP_{eq} of 8–18 ng/kg/d. The average BaP_{eq} of 16 PAHs was 6.1 ng/kg/d for children who came from a school located in close of an industrial and heavy traffic area in Delhi, India (Jyethi et al., 2014). The average BaP_{eq} of 19 PAHs was between 0.037 and 24 ng/kg/d during a heating period for the residents in Bariksir, Turkey (Gungormus et al., 2014). Ma and Harrad (2015) estimated the indoor air BaP_{eq} for adults in different regions of the world. The BaP_{eq} in Asia, North America, and other regions of the world were 7.3, 0.029, and 0.27 ng/kg/d, respectively. The inhalation BaP_{eq} exposure levels (0.029–24 ng/kg/d) in populations from different countries were in a similar range to the dermal BaP_{eq} exposure levels observed in the current study (0.093–37 ng/kg/d).

Yu et al. (2015) measured PAH levels in seven food classification samples (fruits, vegetables, grains, fish, meat, eggs, and milk) from Beijing, China, and estimated the PAH exposure dose for different age groups with levels ranging from 250 to 570 ng/kg/d. Meanwhile, Yu et al. (2012) determined concentrations of 15 PAH in 18 food types in Shanghai, China, and estimated the exposure dose of 12 ng/kg/d for general population. The dietary exposure levels of 16 PAHs ranged from 40 to 250 ng/kg/d in different age groups in Catalonia, Spain (Martorell et al., 2010), and from 71 to 240 ng/kg/d for male youth in the Netherlands (De Vos et al., 1990). Based on the food diary questionnaire and weekly analysis of repeated dietary samples in Naples, Italy, the average dietary exposure level of eight PAHs in 30 children was 78 ng/kg/d (Cirillo et al., 2010), and the average exposure dose of PAH in Estonia between 2001 and 2005 was 6.4 ng/kg/d (Reinik et al., 2007). In Korea, the average dietary exposure dose for 16 PAHs measured in 26 seafood samples was 15 ng/kg/d (Moon et al., 2010). Although the dietary PAH exposure levels varied widely due to different food types, living habits, and different PAH types, the dietary PAH exposure doses of populations reported in different countries (6.4–570 ng/kg/d) was equivalent to the dermal PAH exposure levels observed in this study (26–620 ng/kg/d). The above comparisons among inhalation, dietary, and dermal exposure indicated that the dermal exposure for PAH were equally important as those of inhalation exposure and dietary exposure.

3.4. Exposure risk assessment

The dermal risk values were estimated as 3.5×10^{-6} to 1.4×10^{-3} with a mean of 5.6×10^{-5}. Under most regulatory schemes, risk vales $\leq 10^{-6}$ were considered non-significant or largely negligible, risk values between 10^{-6} and 10^{-4} indicated a low cumulative cancer risk, and risk values $> 10^{-4}$ meant a high potential health risk (Qi et al., 2019). In the present study, 29% (35 out of 120 samples) of the probabilities of BaP_{eq}-based total dermal risk are large than 10^{-4}, indicating a high potential health risk. For the thresholder group, 57% of the exposure to carcinogenic PAH was much higher than 10^{-4}, followed by the preschooler group (37%), and the adult and elder groups (10%), indicating higher potential health risk in adolescents than in middle-aged and elderly people.

The inhalation and dermal exposure to PAHs and subsequent risks for adults, children, and infants in Taiwan, China were reported by Chen and Liao (2006). The geometric mean risk for inhalation and dermal exposure routes were 1.04×10^{-4} and 3.85×10^{-5}, respectively. The risks for dermal exposure in adults are close to the values estimated in this study (4.3×10^{-5}). Tsai et al. (2001) performed a health risk evaluation of palletizing and packaging workers exposed to PAHs via both routes of inhalation and dermal contact in a carbon black manufacturing industry. Bap_{eq}-based lifetime skin cancer risk estimated for palletizing and packaging workers was 1.13×10^{-3} and 1.56×10^{-3}, respectively, one order of magnitude higher than the risk found in this research. Chen et al. (2008) performed a study on inhalation and dermal PAH exposure and related health risks of workers in a fastener manufacturing industry. The average risk of skin cancer owing to skin exposure was 9.7×10^{-3}, which was also higher than what was found in this study. Gungormus et al. (2014) reported inhalation and dermal risks of PAHs for adults during a heating period in the city of Balikesir, Turkey. The population risks associated with dermal exposure were lower (6.58×10^{-9} to 2.57×10^{-6}) than those of the present study.

4. Conclusion

In the present study, a refined assessment of exposure and health risk of PAHs for the general population via the dermal exposure pathway was conducted. The result showed that the PAH dermal exposure and the hand-mouth contact exposure varied with respect to age and gender due to different living environments, hygienic habits, and behavioral patterns among different age groups. The dermal PAH exposure levels were in the same range as those reported for dietary exposure and inhalation exposure, indicating the importance of assessment of the dermal absorption pathway. Absorption from the skin of the head accounted for more than 80% of the total dermal PAH exposure, implying the importance of facial cleaning in daily life to reduce the PAH dermal exposure. Approximately one third of the population exhibited dermal risk higher than the acceptable level (10^{-4}), indicating that PAH exposure from the environment represents a substantial threat for human health. The results of the present study also highlight the importance of refined assessment of exposure and health risk of organic pollutants for different populations and age groups.

Author's contributions

Jian Guo: Investigation, Data Curation, Writing - Original Draft Preparation, Xiao-Jun Luo; Conceptualization, Formal analysis, Visualization, Writing - Review & Editing, supervision, Yan Yang; Investigation, Writing - Review & Editing. Yinzhi Lv; Investigation, Writing - Review & Editing, Yanhong Zeng, Project administration, Method. Bi-Xian Mai; Resources, supervision.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envpol.2022.119802.

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