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ORIGINAL PAPER

PAHs in indoor dust samples in Shanghai's universities: levels, sources and human exposure

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Abstract Given the significant amount of time people spend indoors, the occurrence of polycyclic aromatic hydrocarbons (PAHs) in indoor dust and their potential risks are of great concern. In the present study, ten dust samples from lecture theatres and twelve samples from dining halls were collected from university campuses in Shanghai to investigate the PAH levels, possible sources and human exposure. The total concentrations of 18 PAHs ranged from 9.84 to 21.44 μg/g for dust samples from lecture theatres, and 9.63–44.13 μg/g for samples from dining halls. Total PAH concentrations in indoor dust samples showed a better correlation to black carbon compared to total organic carbon contents. PAHs in dining halls samples showed a similar distribution pattern with that of commercial kitchen air, which indicated that cooking activities could contribute most of the PAHs found in dining halls. Principal component analysis revealed both petrogenic and pyrogenic sources. The potential health risk for PAHs was assessed in terms of BaP equivalent carcinogenic power and estimated daily intake (EDI). Relatively high EDI values compared to other studies suggested that PAHs posed a potential threat to human health in indoor environments at Shanghai's universities.

Keywords Indoor dust · PAHs · Source · Human exposure

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Introduction

People spend a majority of their time indoors, and the quality of this environment will affect people's heath enormously (Maetens et al. 2004). As a complex environmental matrix, indoor dust may have a negative impact on human health through dietary and non-dietary ingestion, dermal contact and inhalation. Furthermore, people may be exposed to contaminants, such as heavy metals (Leung et al. 2008), PAHs (Lorenzi et al. 2011), phthalates (Guo and Kannan 2011), PCBs and PBDEs (Harrad et al. 2010) that adhere to such dust particles. Among the hazardous pollutants, PAHs in dusts have gained great attention due to their carcinogenicity and mutagenicity. PAHs in indoor dust have several potential sources, including domestic burning, power generation, cooking, tobacco smoking and parquet floor glue

(Lohmann et al. 2000; He et al. 2005; Riechelmann et al. 2007). With low vapour pressure and low aqueous solubility, PAHs mainly tend to adsorb to particles, such as indoor dust, rather than existing in the gaseous phase (MacKay 2011; Naspinki et al. 2008). Due to the fact that the indoor environment is relatively protected from environmental degradation, PAHs associated with indoor dust can persist and accumulate (Paustenbach et al. 1997), and therefore indoor dust tends to become a sink for PAHs (Butte and Heizow 2002). It was observed that PAH concentrations in indoor dust were substantially higher than in outdoor dust and soil (Chuang et al. 1999).

Shanghai is the largest industrial city in China under rapid development. Due to a significant increase of automobile and construction activities, the amount of total suspended particles (TSP) in Shanghai air is at a very high level. The city is also under the stress of airborne contaminants, such as PAHs. University campuses in China are typical centres of public activities with dense populations of students, university staff and nearby residents, of which lecture theatres and dining halls are particularly relevant in terms of the density of people. In addition, compared to the lecture theatres, dining halls are more seriously affected by indoor pollution activities such as cooking. Furthermore, universities in Shanghai are spread in different districts with contrasting surroundings. In the present study, thirteen universities in Shanghai were selected as the sampling sites. The indoor dust samples from lecture theatres and dining halls in those universities were investigated for PAH concentrations. Through sampling and analysis, the objectives of this study were to evaluate the contamination levels and sources of PAHs in indoor dusts in Shanghai universities, and to assess the risks they pose to human health.

Materials and methods

Dust collection

Indoor dust samples were collected from lecture theatres and dining halls in thirteen universities in Shanghai (Fig. 1). Indoor dust samples were obtained by brushing gently from the surface of fans and air-conditioning filters at each university campus. Samples were taken from seven sites across central Shanghai, two sites in the New Pudong area, and one site each in the Jiading, Baoshan districts, and two sites in New Pudong Area. The dust samples were kept in brown glass bottles (pre-combusted at 450 °C, 4 h), and then immediately transported to the laboratory where they were frozen prior to extraction and analysis.

PAHs determination

Freeze-dried dust samples were extracted by a pressurized solvent extractor (ASE 300; Dionex) in duplicate. Approximately 0.1-g dust samples were extracted with a mixture of dichloromethane (DCM) and acetone (1:1, v/v) at 100 °C and 10 MPa in a static extraction mode for 5 min. Each sample was extracted twice and the extracts were combined. A mixture of naphthalene-d_8, anthracene-d_{10}, phenanthrene-d_{10}, chrysene-d_{10} and perylene-d_{12} was added to each extraction solvent as the internal standards. The cleanup procedure followed the EPA method 3630C. The extracts were then concentrated by rotary evaporation to 2 mL with solvent exchanged to hexane. Concentrated extracts were further cleaned up using a sodium sulphate anhydrous/silica column from which the PAHs were eluted with a mixture of n-hexane and dichloromethane (70 ml, 7:3, v/v). The collected elution was reduced to 1 mL by rotary evaporation prior to GC–MS analysis.

Eighteen PAHs including naphthalene [Nap], 2-methylnaphthalene [2-MNap], 1-methylnaphthalene [1-MNap], acenaphthylene [Any], acenaphthene [Ace], fluorene [Fln], phenanthrene [Phe], anthracene [Ant], fluoranthene [Fth], pyrene [Py], benz(a)anthracene [BaA], Chrysene [Chr], benzo(b)fluoranthene-benzo(k) fluoranthene [B(b/k)F], benzo(a)pyrene [BaP], perylene [Per], indeno(1,2,3-cd)pyrene [InP], dibenz(a,h) anthracene [DahA], benzo(ghi)perylene [BghiP] were determined by GC–MS. The GC used was an Agilent model 7,890 equipped with a mass selective detector (5975C). The column used was a 30-m-long HP-5MS with 0.25-mm i.d. helium was used as the carrier gas at a flow rate of 1 mL/min. Sample extracts (1 μL) were injected in a splitless mode. All results were expressed on a dry weight basis.

Quantification of PAHs was performed via the internal standards with relative response factors determined by calibration with 18 PAHs mix. To determine the recoveries of the PAH compounds

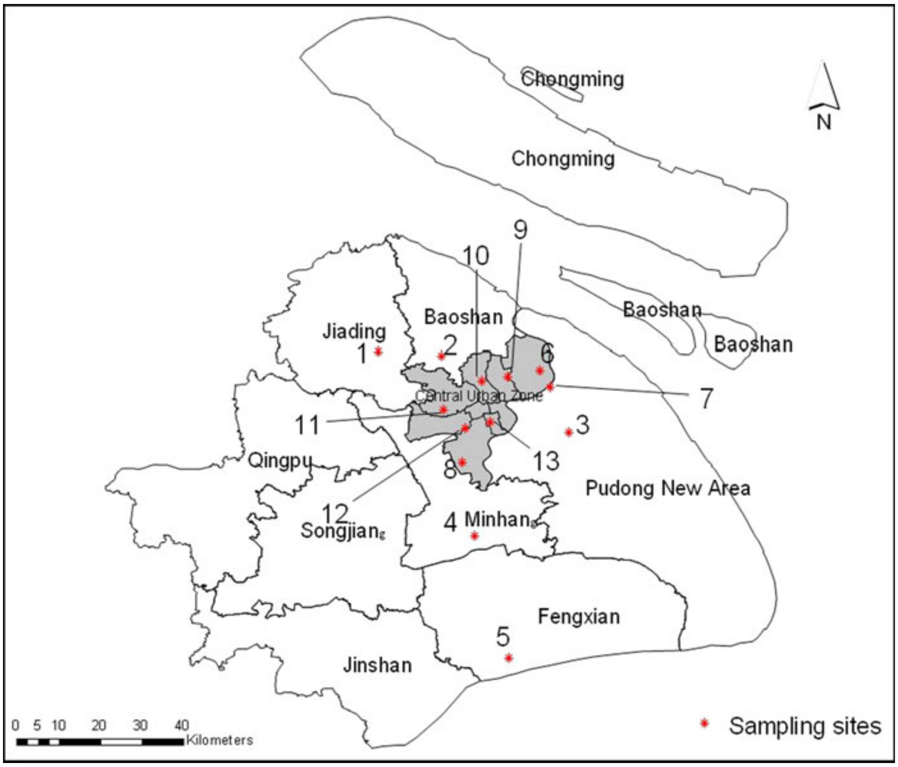

Fig. 1 Sampling sites in lecture theatres and dining halls in Shanghai's universities. *1*-Shanghai University (Jiading Campus, sd-jd), *2*-Shanghai University (Baoshan Campus, sd-bs), *3*-Shanghai University of Traditional Chinese Medicine (Pudong Campus, shzyy-pd), *4*-East China Normal University (Minhang Campus, hs-mh), *5*-East China University of Science and Technology (Fengxian Campus, hl-fx), *6*-Shanghai University of Science and Technology (Yangpu Campus, sl-yp), *7*-Shanghai Maritime University (Pudong Campus, shhs-pd), *8*-East China University of Science and Technology (Xuhui Campus, hl-xh), *9*-Tongji University (Yangpu Campus, tj-yp), *10*-Shanghai University (Zhabei Campus, sd-zb), *11*-East China Normal University (Putuo Campus, hs-pt), *12*-Shanghai Jiaotong University (Xuhu Campus, sj-xh) *13*-Shanghai Jiaotong University School of Medicine (Luwan Campus, jdyxy-lw)

identified, 18 PAH surrogate standard at a known concentration was added to the dust sample prior to extraction. Comparison with the concentration added initially allowed the determination of percentage recovery of the PAH standard. PAHs recovery for samples ranged from 79 to 102 %, and average relative standard deviations of duplicate measurements were below 10 %. The blank tests did not show significant levels of any of the PAHs.

Organic carbon and SEM analysis

Total organic carbon (TOC) and black carbon (BC) were analysed by an elemental analyser (Elementar Vario EL3, Germany). The BC content in each sample was determined according to the direct high temperature oxidization method (CTO-375) (Gustafsson et al. 1997). A Hitachi S4700 scanning electron microscope (SEM) was used to obtain dust sizes and morphologies. The samples were air dried before mounting on carbon stubs and coating with gold.

Results and discussion

Sample characterization

The size and morphology of dust samples from lecture theatres and dining halls were characterized by SEM (Fig. 2). Dust particle size was generally in the range of 10–50 μm with a variety of morphological types. These particles were loosely aggregated from very small particles (1–2 μm or less). There were more rigid particles with irregular structure in samples from the lecture theatres (Fig. 2b) than from dining halls. These could be mineral particles primarily derived

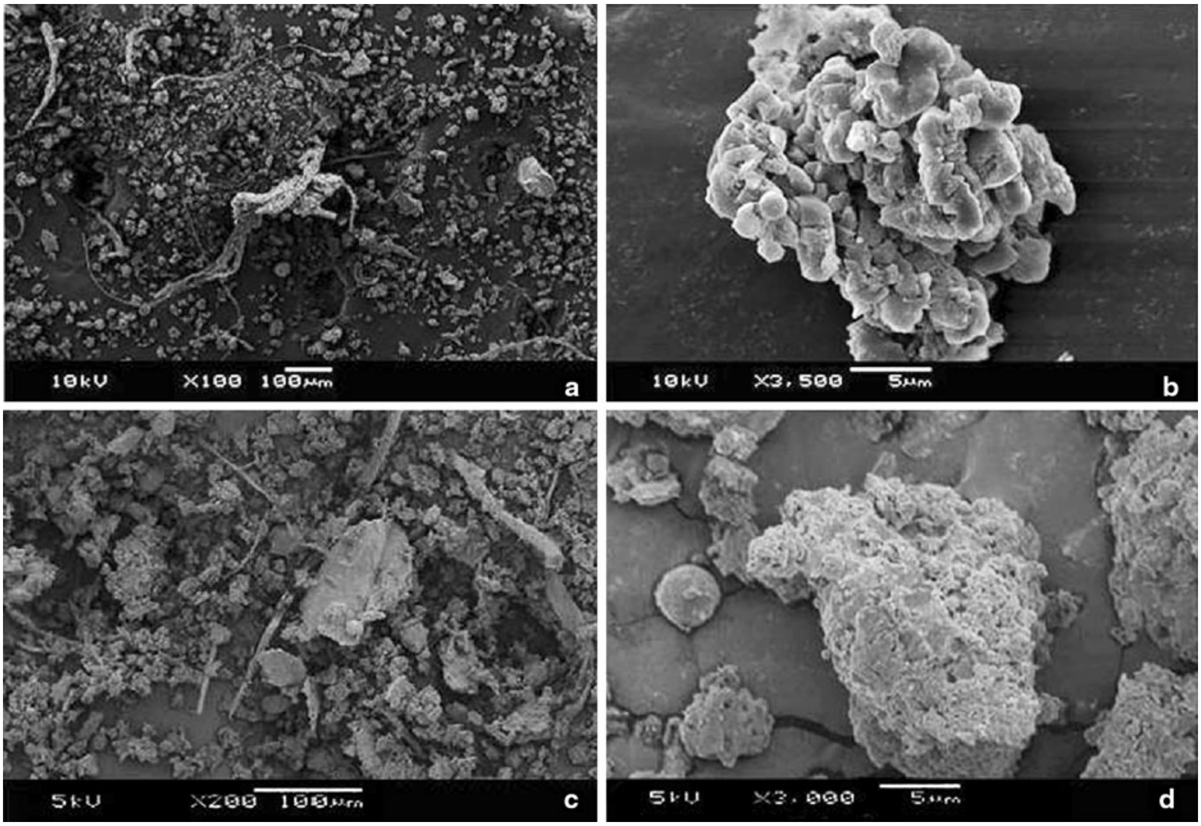

Fig. 2 Scanning electron microphotographs of dust samples from lecture theatres (**a**, **b**) and dining halls (**c**, **d**)

from soil dusts and other anthropogenic activities such as construction (Hu et al. 2009). Particles with a globular structure were visible in most of samples, particularly in samples from dining halls (Fig. 2d). The spherical particles with diameters of about 1–2 μm were most likely to be coal soot and chimney soot (Fernandes and Brooks 2003). TOC and BC contents in dust samples were analysed. TOC contents ranged between 8.9 and 14.9 % in samples from lecture theatres and between 11.9 and 33 % in samples from dining halls. According to CTO375 method, higher BC contents were found in samples from dining halls with a mean value of 3.4 %, than for samples from lecture theatres with a mean value of 1.2 %, confirming important contributions from cooking.

PAH distribution pattern in indoor dust

Table 1 summarized PAH concentrations in indoor dust samples from lecture theatres and dining halls in Shanghai's universities. The total PAH concentrations in dust samples from 10 lecture theatres were from 9.84 to 21.44 μg/g with a mean concentration of 12.47 μg/g, among which 2MNap, Phe and Fl were the dominant PAH components. The total PAH concentrations in dust samples from 12 dining halls ranged from 9.69 to 44.13 μg/g with a mean concentration of 18.94 μg/g, among which 2MNap, Phe, Fl, Py, Chry, B[b/k]F and B[ghi]P were more abundant. High molecular weight (HMW) PAHs (4 + 5 + 6-ring PAHs) accounted for about 30–58 % of total PAH concentrations in dust samples from lecture theatres, and about 41–82 % of total PAH concentrations in dusts samples from dining halls.

As shown in Fig. 3, among 10 dust samples from lecture theatres, dust samples from East China Normal University located in urban area (Putuo district, hs-pt) showed the highest PAH concentration of 21.44 μg/g; while the dust sample from East China University of Science and Technology campus located in a coastal area showed the lowest PAHs concentration (Fengxian district, hl-fx) when compared to the others. Among

Table 1 PAH concentrations in indoor dust samples from lecture theatres and dining halls in Shanghai's universities (μg/g)

	Lecture theatres					Dining halls				
	Mean	Median	Minimum	Maximum	SD[a]	Mean	Median	Minimum	Maximum	SD
Nap	1.60	1.39	1.07	2.27	0.15	1.07	0.88	0.71	1.80	0.05
2MNap	2.38	2.38	1.96	2.85	0.35	1.71	1.78	1.15	2.11	0.13
1MNap	0.84	0.85	0.64	1.02	0.12	0.65	0.70	0.47	0.80	0.05
Acy	0.17	0.14	0.10	0.33	0.03	0.18	0.17	0.09	0.34	0.01
Ace	0.06	0.06	0.05	0.08	0.01	0.08	0.06	0.04	0.18	0.00
Fluo	0.23	0.21	0.17	0.42	0.02	0.26	0.20	0.15	0.47	0.02
Phe	1.47	1.40	0.98	2.61	0.14	2.15	1.90	1.17	4.24	0.14
An	0.10	0.10	0.06	0.19	0.01	0.13	0.10	0.06	0.38	0.01
Fl	1.20	1.11	0.62	2.29	0.15	2.98	2.32	1.18	9.11	0.23
Py	0.89	0.84	0.48	1.56	0.07	2.25	1.42	0.54	10.38	0.23
BaA	0.31	0.28	0.16	0.68	0.03	0.37	0.22	0.06	1.39	0.03
Chry	0.71	0.60	0.27	1.85	0.07	2.15	1.64	0.90	5.98	0.11
B[b/k]F	0.83	0.68	0.46	2.43	0.07	1.77	1.00	0.65	6.76	0.09
BaP	0.47	0.45	0.27	0.78	0.04	0.36	0.23	0.05	1.32	0.02
InP	0.51	0.44	0.31	1.31	0.03	1.04	0.47	0.24	4.11	0.06
DahA	0.10	0.10	0.07	0.23	0.06	0.18	0.09	0.04	0.69	0.01
BghiP	0.59	0.50	0.36	1.43	0.03	1.61	0.67	0.35	6.37	0.11
∑PAHs	12.47	11.66	9.84	21.44	0.94	18.94	12.64	9.69	44.13	1.13

[a] Standard deviation

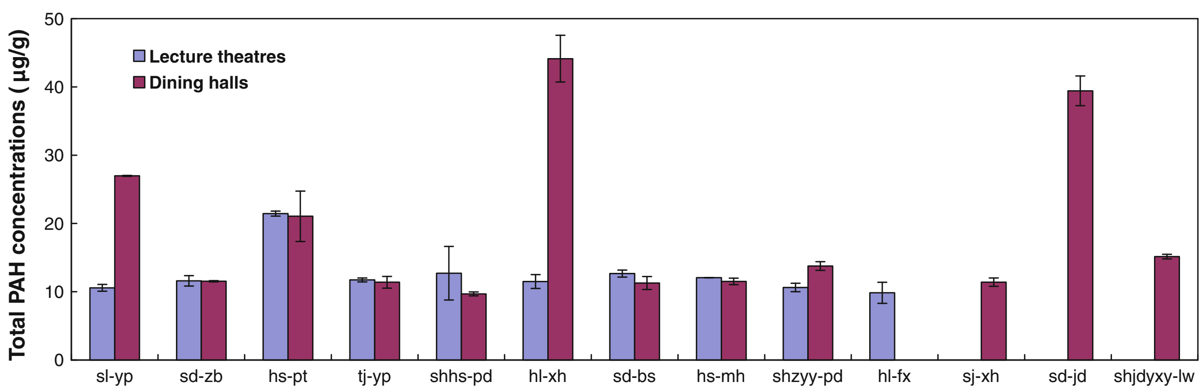

Fig. 3 Total PAH concentrations in indoor dust samples from lecture theatres and dining halls in Shanghai's universities

12 dust samples from university dining halls, the samples from East China University of Science and Technology campus located in inner city (Xuhui district, hl-xh) were the most seriously polluted with a PAH concentration up to 44.13 μg/g. Those were followed by the samples from Shanghai University at Shanghai suburb (Jiading district, sd-jd) with a concentration of 39.43 μg/g. It is noticeable that HMW PAHs contributed about 81 % of total PAHs in these dust samples with elevated PAH concentration. In addition, dust samples from dinning halls generally showed relatively low 2-ring PAHs and high HMW PAHs contribution of total PAHs when compared to the corresponding samples from lecture theatres (Fig. 4). The HMW PAHs are primarily from the incomplete combustion of fossil fuels and were hardly to be evaporated and degraded (Zakaria et al. 2002). In addition, PAH concentrations in samples

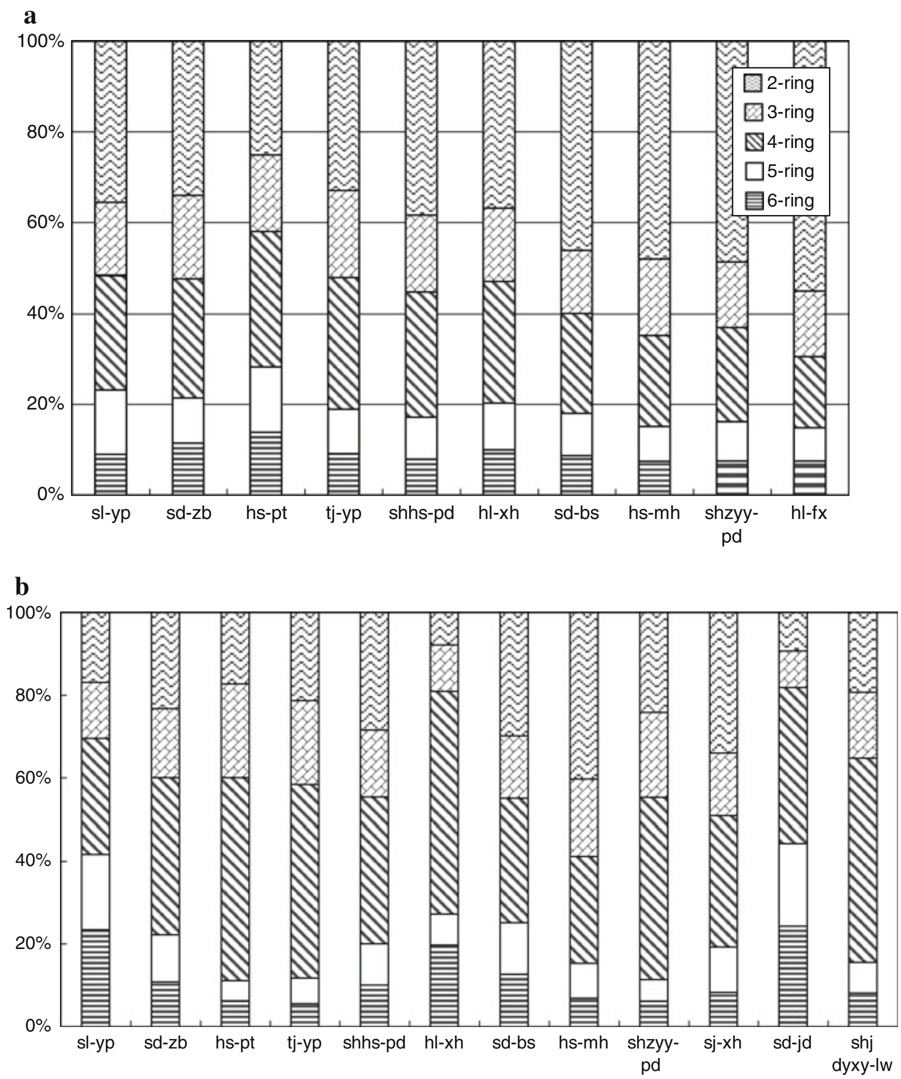

Fig. 4 PAHs distribution pattern in dust samples from lecture theatres (**a**) and dining halls (**b**) in Shanghai's universities

collected from the dinning halls were remarkably higher than those from lecture theatres only at sly-yp and hl-xh sites (Fig. 3). Compared to dining halls in other universities, the environment in these two dinning halls was poorly ventilated. Therefore, PAHs generated by intensive cooking activities tended to accumulated in the closed environment in these two dinning halls.

Polycyclic aromatic hydrocarbons in kitchen environment originate primarily from oil fumes and cooking practices (Zhu and Wang 2003). As shown in Fig. 5, PAH distribution pattern in dust samples from dining halls is quite similar to that in commercial kitchen air except for relatively high contribution from 4-ring PAHs. Compared to cooking air and oil fumes, more 4- and 5-ring PAHs were detected in dust samples from dining halls. Possible reason for the abundance of 4- and 5-ring PAHs in samples might include: (1) endogenous PAHs originating from cooking or oil fumes are less prone to evaporate and more persistent than 2- and 3-ring PAHs. Consequently they tend to accumulate in dining hall samples with relatively high concentrations; (2) exogenous 4- and 5-ring PAHs tend to adsorb onto dust particles with higher adsorption coefficients than those of 2–3 ring PAHs.

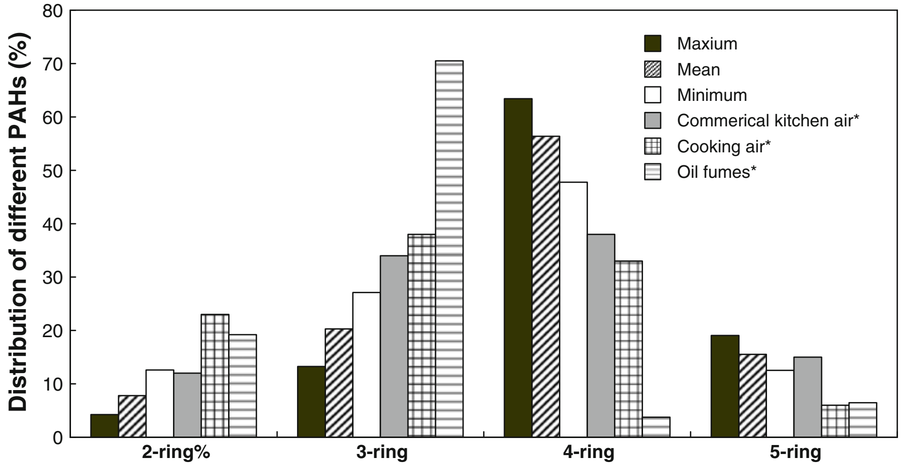

Fig. 5 Distribution of 2–5 ring PAHs in dust samples (Maximum, Mean and Minimum) from dinning halls compared to other studies (*data from Zhu and Wang 2003)

Table 2 Diagnostic PAH components from different sources

Source	Diagnostic PAHs components	Reference
Raw coal, crude oil	Nap, 2MNap, 1MNap	Utvik et al. (1999), Zakaria et al. (2002), Wang et al. (2006), Yang et al. (2008)
Coal combustion, raw coal and charcoal	Phe, Ant, Fl, Py	Harrison et al. (1996), Masclet et al. (1987), Mastral et al. (1996), Jonker and Koelmans (2002)
Oil soot, wood soot	InP, B[b/k]F, BaP, BghiP	Li and Kamens (1993), Jonker and Koelmans (2002)
Traffic fuel emission	BghiP, Phe, Fl, Py	Harrison et al. (1996), Jonker and Koelmans (2002)
Coke ovens	Fluo, Ace	Matt et al. (1999)

PAHs vs. TOC/BC

Total PAH concentrations in all indoor dust samples showed a better correlation to BC ($r = 0.66$, $p < 0.001$) compared to TOC contents ($r = 0.52$, $p < 0.05$). Furthermore, HMW PAHs showed a strong correlation to BC contents ($r = 0.67$, $p < 0.001$); while low molecular weight PAHs (2 + 3-ring PAHs) had no significant correlation to either TOC or BC contents. To get a better understanding, correlation analysis was done between BC and each group of PAHs in dust samples from lecture theatres and dining halls, respectively. For samples from dining halls, BC contents showed significant positive correlation only to 3-ring ($r = 0.78$, $p < 0.005$) and 4-ring PAHs ($r = 0.709$, $p < 0.01$). By analysing different types of BC-like materials, it was found that 4-ring PAHs dominated PAHs investigated in oil soot and coal soot, while 3-ring PAHs were the most abundant group in coal and charcoal samples (Jonker and Koelmans 2002). Therefore, these 3- and 4-ring PAHs were most likely produced by cooking activities. Whereas for samples from lecture theatres, none of the PAH group showed significant correlation to BC contents, indicating more diverse sources of PAHs.

Principal component analysis (PCA)

Principal component analysis has been proven to be an effective method to study complex characteristic sources (Baumard et al. 1998). In this way, a semi-quantitative description was conducted for the relative contribution of major sources. Various possible sources were marked by specific PAH compounds (Table 2). PCA results of 18 PAHs were listed in Table S1, the supplementary materials. For samples from lecture theatres, two principal components (PC) were extracted. The first factor was responsible for 71.4 % of the total variance. This factor was predominately weighted by Fluo, Phe, BaA, Chry, InP, InP and DahA indicating oil combustion, traffic fuel emission and coal combustion sources. The factor 2 (16.4 % of variance) had high loading values of Nap, 2MNap and 1MNap, indicating raw coal and crude oil source.

As for samples from dining halls, three principal components were extracted with the contribution of 57.7, 20.4 and 11.9 %, respectively. High molecular weight compounds of B[b/k]F, InP and DahA showed a better correlation than other compounds in the first principal component, revealing oil and wood combustion sources. Fl and Py were of high loading in the second principal component, indicating coal combustion source. Of the third principle component, 2MNap and 1MNap showed a high loading, implying raw coal, crude oil or related products sources.

Human risk assessment

Universities are a public place for students, university staff and local residents nearby. Indoor dusts contaminated by PAHs pose a potential risk to people regularly visiting those places. Among PAHs, BaP is of primary concern because of its high carcinogenicity. In addition to BaP, high molecular mass PAHs such as BaA, B[b]F, B[k]F, InP and DahA are also of carcinogenic potential. One approach to estimate the health risk exposure to these carcinogenic PAHs is to calculate BaP equivalent carcinogenic power (BaPE) based on PAH carcinogenic potency relative to BaP (Yassaa et al. 2001), as shown in Eq. 1:

$$\mathrm{BaPE} = 0.06\ \mathrm{BaA} + 0.07\ \mathrm{B[b+k]F} + \mathrm{BaP} + 0.6\ \mathrm{DahA} + 0.08\ \mathrm{InP} \quad (1)$$

Calculated BaPE values ranged from 0.37 to 1.11 μg/g for samples from lecture theatres with a mean value of 0.65 μg/g. Values ranged from 0.17 to 2.41 μg/g for dusts collected from dinning halls with a mean value of 0.7 μg/g. The highest BaPE values were found in samples from dining halls with 2.41 μg/g at sd-jd site and 2.25 μg/g at sl-yp site, indicating relatively high carcinogenic risks at these sites.

Hand-to-mouth activities result in the oral ingestion of dust, which tends to be the dominant intake pathway for chemicals found on and in dust (Maetens et al. 2004). Hereby, using Eq. 2, we calculated the estimated daily intake (EDI, ng/kg-bw/day) of PAHs via dust ingestion for people in the study area:

$$\mathrm{EDI} = \frac{C_{\mathrm{dust}} f_1}{M_1} \quad (2)$$

where C_{dust} is the average concentration of each PAH in dust samples (μg/g, dry weight); f_1 is the recommended dust ingestion value per day which is 0.05 g/day for adults (USEPA 1997); M_1 is the average body weight (kg), which is suggested to be 63 kg in China (Guo and Kannan 2011).

People in dining halls are exposed to higher risk of PAHs via ingestion with generally higher average EDI of PAHs than in lecture theatres in universities (Table 3). Furthermore, EDI values in Shanghai's universities were generally 2–3 times higher than those of different indoor dusts collected in Hong Kong and Berlin (Kang et al. 2010; Fromme et al. 2004). In a previous review study, Oomen et al. (2008) calculated EDI values based on the highest geometric means of eight PAH concentrations in house dust samples. In the present study, with the exception of Ace, Fluo and An, almost all the EDI values for dust samples exceeded corresponding values in Oomen's report, where a body weight of 70 kg was used. In addition, EDI values calculated based on average BaPE values were 0.52 and 0.56 for samples from lecture theatres and dining halls, respectively, which were also higher than those from other studies (Table 3). Therefore, human exposure to PAHs by indoor dust ingestion in Shanghai's universities was at a relatively high level, with implications of a potential significant threat to the people concerned.

Conclusions

Elevated PAH concentrations were found in indoor dust samples from lecture theatres and dining halls in thirteen universities in Shanghai. Dust samples from dinning halls were relatively more polluted by PAHs with higher mean, median and maximum PAH concentrations than dust samples from lecture theatres. The total PAH concentrations were from 9.84 to 21.44 μg/g in samples from lecture theatres, and from 9.69 to 44.13 μg/g for samples from dining halls. PAH distribution patterns in dust samples from

Table 3 Estimated daily intake (EDI, ng/kg body weight/day) of PAHs via dust ingestion in indoor dusts in university campuses in Shanghai and other regions

	Lecture theatres, Shanghai	Dining halls, Shanghai	Hong Kong	Berlin	Oomen's research
Nap	1.27	0.81	0.1	0.16	0.2
2MNap	1.89	1.36	NA	NA	NA
1MNap	0.67	0.52	NA	NA	NA
Acy	0.13	0.15	0.01	0.02	0.04
Ace	0.05	0.06	0.02	0.04	0.1
Fluo	0.18	0.21	0.02	0.07	0.4
Phe	1.17	1.77	0.61	0.76	0.3
An	0.08	0.11	0.02	0.06	0.1
Fl	0.95	2.48	0.68	0.76	0.1
Py	0.7	1.88	0.55	0.53	0.3
BaA	0.24	0.3	0.17	0.23	0.2
Chry	0.56	1.79	0.43	0.44	0.3
B[b/k]F	0.66	1.48	0.78	0.72	0.4
BaP	0.37	0.28	0.3	0.21	0.2
InP	0.4	0.87	0.46	0.26	0.2
DahA	0.08	0.15	0.05	0.04	0.1
BghiP	0.47	1.36	0.71	0.28	0.2
$\sum$PAHs	9.9	15.58	4.91	4.59	3.14
BaPE	0.52	0.56	0.43	0.32	0.32
Sources	This study	This study	Kang et al. (2010)*	Fromme et al. (2004)*	Oomen et al. (2008)

* Indicates results calculated from literature data

dinning halls were quite similar to those of commercial kitchen air, revealing the contribution of cooking and oil fumes. Meanwhile, higher contents of 4- and 5-ring PAHs indicated that these contaminants were of both endogenous and exogenous sources. Total PAH concentrations in all samples showed a better correlation to BC compared to TOC contents. In addition, HMW PAH concentrations correlated well to BC contents. A semi-quantitative PCA concluded that PAHs were primarily derived from traffic emission, coal combustion and diesel combustion for dust samples from lecture theatres, whereas coal combustion and petroleum-related products accounted for most PAHs in dust samples from dining halls. As a key exposure pathway, PAHs ingested by people via dust were estimated by *EDI* values. Relatively high daily intake values of PAHs were found for all samples, both from lecture theatres and dining halls, indicating a relatively high threat to human health from dust samples containing PAHs in those university environments.

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