CONCENTRATIONS AND SOURCES OF POLYCYCLIC AROMATIC HYDROCARBONS (PAHs) IN INDOOR DUST IN CHINA

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Introduction

Indoor environment is main space for human daily life, and also a place where human expose to different hazardous substances. Indoor dust, as a special medium, is regarded as a significant source of persistent toxic substances (PTSs) exposure to humans. PTSs in indoor environment can enter human bodies through non-dietary ingestion, including indoor air inhalation, dust ingestion and dust dermal absorption [1], and the route of exposure has a higher influence on children than adults [2]. Polycyclic aromatic hydrocarbons (PAHs) are an important PTS, PAHs, as one of the most widespread organic PTS. are formed by incomplete combustion of fossil fuels, carbon-containing fuels such as wood, coal, and diesel[3-4]. PAH residue levels in Chinese indoor and outdoor air are relatively higher than those in developed country, since coal is the largest source of energy for the generation of electricity in China. In China, up to now, research on PAHs in indoor environment is mainly concentrated in some special polluted areas, such as e-waste sites, but little in common indoor environment. In this research, indoor dust samples were collected from Chinese 23 provinces of China and from different types of indoor environment to investigate the residual levels, spatial distribution and source apportionment.

Materials and methods

1.1 Sample collection:

From January to March, 2010, 81 samples of indoor dust were collected in 23 cities of China, including 45 family homes and 36 non-family site (including public places, laboratories and offices). Each dust sample waited 0.5-1.0 g, and was taken in house but not on floor or close to window using brush rinsed with acetone in a strict procedure. One blank sample for every 10 real samples was preformed to check the contamination during sampling.

1.2 The pretreatment and instrument analysis:

Indoor dust samples were extracted with the method of ultrasonic extraction. The detail procedures are as followed, measure 0.2 g dust sample and put it into the 10 mL glass centrifugal tube. Then added the Naphthalene-D8, Fluorene-D10, Pyrene-D10, Perylene-D12 as spiking surrogate, and extract 20min with water bath ultrasonic extraction in mixture of hexane and acetone(v:v=1:1). After cleaned up using Silica Gel Column, the extracted liquid were condensed to 0.8 mL by highly purified Nitrogen, then add Phenanthrene-D10 as Internal standard , adjust the volume to 1 mL for the following measurement. 16 kind of PAHs were measured with GC/MS(Agilent 6890GC-5975B, USA) in electron ionizationmode (GC-ECNIMS) using HP5-MS capillary column (30 m _ 0.25 mm i.d., 0.25 _m film thickness, J&W Scientific), following temperature program: 80°C for 3 min, 10°C/min to 300°C, and held for 5 min. Injection of 2 uL was performed in splitless mode at an injector temperature of 265°C. The temperatures were 150°C, 230°C, and 250°C for the quadrupole, ion source, and interface, respectively. Further details can be found elsewhere [5].

1.3 QA/QC

Recoveries of PAHs in dust samples were 70.5 ± 7.1% for naphthalene-D8, 69.2 ± 8.1% for fluorene-D10, 103.3 ± 14.6% for pyrene-D10, and 106.4 ± 17.8% for perylene-D12.

The concentrations of most PAHs in blank samples were detected equal or below 1% of PAHs in real samples except naphthalene (26.1%), acenaphthene (5.82%), fluorene(3.34%), and anthracene(7.71%). The data finally reported without correction.

Results and discussion

2.1 PAH residual levels and profiles

Total concentrations of 16 PAHs (in ug/g dry weight) in indoor dust ranged from 1.00 to 470 with a mean value of 30.9 (Table 1), These values were similar to those measured for urban sites worldwide (Table 2). Benzo[b]fluoranthene (6.23 ug/g) was the dominant congener followed by fluoranthene (4.26 ug/g) and phenanthrene (4.19 ug/g). High-molecular weight PAHs (4 ring) are predominant PAHs in indoor dust, 68% of the total PAHs in family homes, and 84.6% of the total PAHs in non-family homes. Four ring PAHs were the main PAH homologue in family homes, while 5 ring PAHs in non-family homes. In addition, indeno[1,2,3-cd]pyrene and benzo[g,h,j]perylene, as the traffic source indicators, were much higher in non-family dust than the family dust, which was probably because of higher influence from outdoor environment through the air exchange for non-family sites than family sites. As 2-ring and 3-ring PAHs are relatively volatile resulted in less abundant in settled dust and generally less toxic and more susceptible to degradation while larger molecular (4–6 ring) PAHs are more toxic and persistent in environment, thus, in comparison to air exposure source, settled dust is an more important exposure source of toxic PAHs through non-dietary ingestion or dermal contact to human health.

Table 1 Levels of PAHs in Indoor dust in China (ug/g d.w.)

PAHs	Min	Max	Mean	Detection rate（%）
Naphthalene	0.224	4.84	0.849	100.0
Acenaphthylene	0.013	1.43	0.173	100.0
Acenaphthene	0.014	1.01	0.093	100.0
Fluorene	0.054	3.6	0.464	100.0
Phenanthrene	0.196	38.7	4.19	100.0
Anthracene	BDL	2.89	0.418	99.0
Fluoranthene	0.064	44.9	4.26	100.0
Pyrene	0.047	32.2	2.89	100.0
benzo[a]anthracene	0.009	48.8	2.37	100.0
Chrysene	0.029	20.3	1.79	100.0
benzo[b]fluoranthene	0.027	122	6.23	100.0
benzo[k]fluoranthene	0.021	15.4	0.763	100.0
benzo[a]pyrene	0.014	41.3	1.71	100.0
indeno[1,2,3-cd]pyrene	0.014	52.3	1.97	100.0
dibenzo[a,h]anthracene	0.008	14.5	0.510	100.0
benzo[g,h,j]perylene	0.018	57.1	2.14	100.0
Total amount	1.01	466	30.9	—

BDL: Below detect limit

The results showed a larger variation of the PAHs distribution in workspace compared to family and public environment (Fig. 1). Levels of PAHs (in ug/g d.w.) ranged from 4.16 to 47.4 in office dust, from 1.01 to 69.5 in home, and from 4.00 to 9.70 in public. Relatively higher mean concentration in office (23.4) than home (15.5) and public (7.2). Three outliers PAH concentrations (222-466) in the dust occurred in the cities in northeastern China, the samples collected at workplace, the most likely explanation for these outliers is these room use coal as fuel or close to chimney. There is no significant difference between urban and rural homes (t-test, p=0.38).

Table 2. Comparison of PAHs levels in different regions of the world (μg/g d.w.)

Sampling site	PAHs Concentration	Sampling site	PAHs compound	Year
Palermo,Italy [6]	0.036-34.5	Home+Office	16	2006
	16.7	Home	16	1991
Kuwait [7]	2.4	Home	16	1992
	1.6	Home	16	1996
Shanghai, China[8]	8.22-42.04	Home+Office+Others	16	2004-2005
Ottwa, Canada[9]	1.5-325	Home	13	2002-2003
This study, China	0.5-54	Home	13	2009
This study, China	1.0-69.6	Home	16	2009
This study, China	1.0-470	Home+Office+Others	16	2009

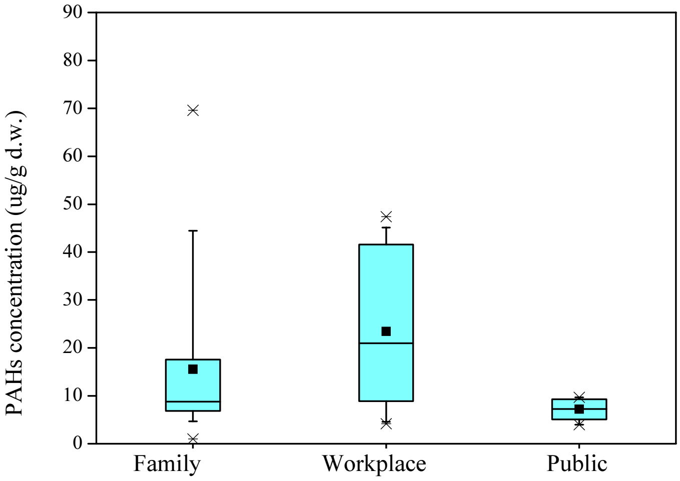

Fig.1 Levels of PAHs in different kind of sampling sites

2.2 Spatial Distribution of PAHs

Figure 2 shows that the higher PAHs level in indoor dust was found in the northern areas, and lower in the south, possibly due to the fact that the biomass/coal combustion in rural family and coal combustion in urban family for heating in winter in the northern China.

Similar spatial pattern also found in the trend of PAHs in dust along latitude. Significant correlation ($r=0.57$; $p=0.0003 < 0.05$) between PAHs residual level in dust and latitude was found. Biomass and coal combustion for heating depending on temperature (latitude) is also a major factor.

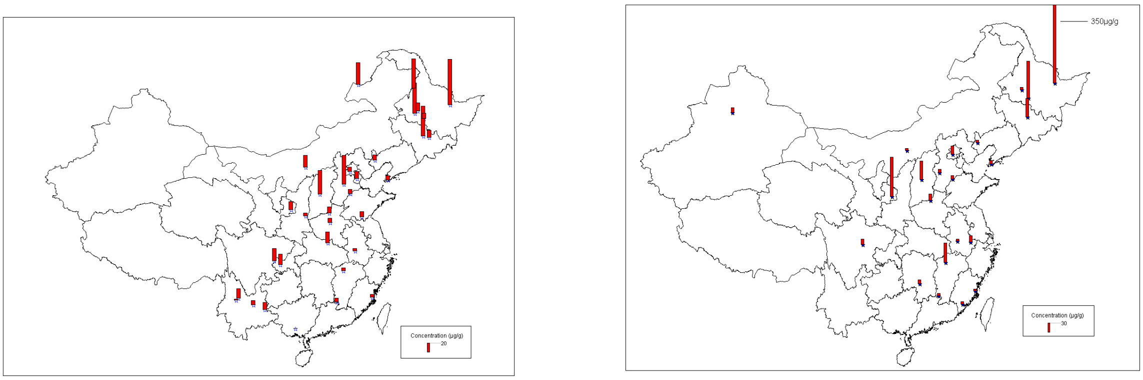

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Fig. 2 Distribution of PAHs in family homes (left) and non-family (right).

2.3 Source apportionment

The source apportionment analysis using diagnostic ratios (Table 3) and principal component analysis (PCA) indicated that different sources of PAHs depend on types of indoor environment. The ratio of Ant/ (Phe+Ant) and BaA/ (BaA+Chr) was used for qualitative analysis of PAHs indoor dust source. Correlation cofficients (0.87-0.95) between BaA and Chr, Ant and Phe indicated these compounds come from the similar pollution source, transport, parthways, and fate. The diagnostic ratios and PCA analysis indicated that PAHs in both urban and rural family dust were mainly from pyrogenic and petrogenic sources from outside (transportation) and cooking/heating from inside.

Table 3 Comparison of characteristic ratio of PAHs in different types of indoor dust

	BaP/BghiP(mean)	BaA/Chr(mean)	Flu/(Flu+Pyr)(mean)	Reference
Vehicle emissions	0.3-0.78	0.63	—	[10-13]
Gasoline combustion	0.3-0.4	0.28-1.2	0.4	[10-13]
Diesel combustion	0.46-0.81	0.17-0.36	0.6-0.7	[10-13]
Kerosene combustion	—	0.37	0.5	[10-13]
Wood burning	—	0.93	0.74	[10-13]
Crude oil combustion	—	0.49	0.44	[10-13]
Coke oven	5.1	0.7	—	[10-13]
Incinerator	0.14-0.6	—	—	[10-13]
Rural family	0.50-0.92(0.7)	0.27-1.61(0.74)	0.55-0.66(0.6)	This study
Urban family	0.48-1.55(0.79)	0.20-3.59(1.19)	0.54-0.73(0.62)	This study
Non-family	0.30-1.43(0.73)	0.28-2.40(1.03)	0.43-0.7(0.6)	This study

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