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Concentrations and sources of polycyclic aromatic hydrocarbons in indoor dust in China

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ABSTRACT

Indoor dust samples were collected across China in the winter of 2010 from 45 private domiciles and 36 public buildings. 16 polycyclic aromatic hydrocarbons (PAHs) were determined by GC–MS. Total concentrations of PAHs ranged from 1.00 μg/g to 470 μg/g with a mean value of 30.9 μg/g. High-molecular weight (HMW) PAHs (4 to 6 rings) are the predominant PAHs found in indoor dust, accounting for 68% of the total PAH concentration in private domiciles, and 84.6% in public buildings. Traffic conditions and cooking methods were the two key factors controlling PAH levels, especially for coal combustion and vehicular traffic emission sources. A significant positive correlation was observed between PAH concentrations in indoor dust and based on location (latitude and longitude). The latitudinal distribution indicated a higher usage of coal for heating in Northern China than in Southern China. The longitudinal distribution indicated that the usage of oil and mineral fuels as well as economic development and population density increased from West China to East China. In addition, diagnostic ratios and principal component analysis (PCA) were used to explore source apportion, as indicated in both the pyrogenic and petrogenic sources of PAHs in indoor dust in China. Furthermore, the BaP equivalent was applied to assess the carcinogenic risk of PAHs, which also indicated that traffic emissions and coal combustion were the two major contributions to carcinogenic risk of PAHs in indoor dust in China.

1. Introduction

People spend the majority of their time (approximately 80%) in an indoor environment (Mitra and Ray, 1995); thus, human exposure to indoor contaminates is of great concern, especially for children and the elderly (Mendell and Heath, 2005). Indoor air and dust are the major two matrixes for contaminates. Contaminates in indoor environment can enter human body via inhalation, ingestion with hand-to-mouth contact, or direct absorption through the skin (Whitehead et al., 2011). Compared with indoor air, indoor dust is more suitable for estimating human exposure to contaminates because indoor dust is a repository of contaminates over a long period of exposure (Butte and Heinzow, 2002). Polycyclic aromatic hydrocarbons (PAHs) are one of the major types of contaminates in indoor dust due to their ubiquitous occurrence and toxicity (Kang et al., 2010; Langer et al., 2010).

PAHs were found to be mainly originated from emissions of coal combustion, coke production, transport oil combustion (traffic sources), and biomass burning (straw and firewood burning) in China (Xu et al., 2006). Almost 22% of the global atmospheric emission inventory of PAHs originated from China in 2004 (Zhang and Tao, 2009). PAH pollution in the environment is continuously increasing due to the rapid social–economic development of China. High levels of PAHs were reported in the water, sediment, soil and air in China (Wan et al., 2006; Wang et al., 2013; S. Zhang et al., 2008; Zhang et al., 2004). For indoor dust, the highest PAH floor surface loadings were observed in China compared to other regions of the world (Naspinski et al., 2008). This is expected because coal is the largest source of energy for the generation of electricity in China. Another important source of PAHs was traffic emissions, especially in cities (i.e., Beijing and Shanghai) (Liu et al., 2007; Tang et al., 2005; Wang et al., 2011).

The objectives of this study were as follows: (1) to determine the concentrations and profiles of PAHs in indoor dust collected on the national scale in China; (2) to establish the national scale distribution of PAH concentration; (3) to examine the relationship between the characteristics of indoor environments and PAH concentrations in

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indoor dust; and (4) to distinguish the sources of PAHs in indoor dust of China.

2. Materials and methods

2.1. Sample collection

From January to March 2010, 81 samples of indoor dust were collected in 23 cities around China, including 45 family homes and 36 public buildings (including public places schools, laboratories and offices). Each dust sample weighed 0.5–1.0 g and was taken in the building using a brush rinsed with acetone under strict procedures, avoiding the floor surface and windows. During sampling, characteristics such as house type, floor types, number of electronic devices, cooking conditions and whether the inhabitants were smokers were also recorded.

2.2. Treatment and analysis

Indoor dust samples were extracted using ultrasonic extraction. The detailed procedures are as follows: a 0.2 g dust sample was placed into a 10 mL glass centrifuge tube. Naphthalene-D8, fluorene-D10, pyrene-D10, and perylene-D12 were added as spiking surrogates, and a water bath was used for extraction for 20 min ultrasonic extraction in a mixture of hexane and acetone (1:1, v/v). After the sample was prepared using a silica gel column, the extracted liquid was condensed to 0.8 mL by highly purified nitrogen, and then, phenanthrene-D10 was added as an internal standard. The volume was adjusted to 1 mL for the following measurement. 16 EPA PAHs [naphthalene (Nap), acenaphthylene (Acy), acenaphthene (Ace), fluorene (Flo), phenanthrene (Phe), anthracene (Ant), fluoranthene (Flu), pyrene (Pyr), benzo(a)-anthracene (BaA), chrysene (Chr), benzo(b)-fluoranthene (BbF), benzo(k)fluoranthene (BkF), benzo(a)pyrene (BaP), dibenz(a,h)anthracene (DahA), indeno(1,2,3-cd)pyrene (IcdP), benzo(g,h,i)perylene (BghiP)] were analyzed using an Agilent 6890 N GC coupled with an Agilent 5975B mass spectrometer in electron ionization mode using a HP5-MS capillary column (30 m × 0.25 mm i.d., 0.25 μm film thickness, J&W Scientific), and the following temperature program: 80 °C for 3 min, 10 °C/min to 300 °C, and held for 5 min. Next, 2 μL was injected in the 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 (Ma et al., 2010).

2.3. QA/QC

Contamination arising from sampling, transport, and treatment was evaluated by the analysis of field and procedural blanks. One blank sample for every 10 real samples was analyzed to check for contamination during sampling. The concentrations of most PAHs in the blank samples were measured at equal or below 1% of PAHs in real samples except for naphthalene (26.1%), acenaphthene (5.82%), fluorine (3.34%), and anthracene (7.71%). The final data is reported without correction. For surrogates, 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. Instrumental calibration curves were obtained with an injection of 5 levels of standards, and the regression coefficients (R) of all calibration curves were ≥0.99.

3. Results and discussion

3.1. PAH residual levels, profiles and influencing factors

3.1.1. Concentrations of PAHs and profiles

Overall, 16 PAHs were detected in all dust samples, except for anthracene, with a detection rate of 99%. The concentration of PAHs is summarized in Table 1. Total indoor dust concentrations of 16 PAHs (Σ_{16}PAHs) (in μg/g) extend over 2–3 orders of magnitude, ranged from 1.00 to 470 with a mean value of 30.9. benzo(b)fluoranthene (6.23 μg/g) was the dominant congener followed by fluoranthene (4.26 μg/g) and phenanthrene (4.19 μg/g). In terms of compositional pattern, high-molecular weight (HMW) PAHs (4 to 6 rings) are the predominant PAHs in indoor dust, accounting for 68% of the total PAHs in private homes, and 84.6% of the total PAHs in public indoor spaces. Furthermore, 4-ring PAHs are the main PAH homologues in private homes, while 5-ring PAHs dominated in public indoor dust. In addition, 2- and 3-ring PAHs are relatively more volatile, which resulted in less abundance in settled dust and are more abundant in the air. However, low-molecular weight (LMW) PAHs are generally less toxic and are more susceptible to degradation, while HMW PAHs are more toxic and persistent in environment. Compared with air exposure sources, settled dust is a more important exposure source of toxic PAHs through non-dietary ingestion or dermal contact. It can be concluded that our results indicated the ubiquitous pollution of HMW PAHs and the high risk of exposure to PAHs for humans from indoor dust.

The comparison of PAH with other areas is shown in Table 2, the measurements resulting from this study were much higher than the median PAH levels in Berlin in Germany (0.3 μg/g) and Southern

Table 1
Concentrations of PAHs in indoor dust in China (μg/g).

PAHs	Min	Max	Mean	Median	SD[a]	Percentage (%)
Naphthalene	0.224	4.84	0.849	0.599	0.759	5.85
Acenaphthylene	0.013	1.43	0.173	0.055	0.327	1.19
Acenaphthene	0.014	1.01	0.093	0.052	0.130	0.64
Fluorene	0.054	3.6	0.464	0.200	0.627	3.19
Phenanthrene	0.196	38.7	4.19	1.67	6.97	29.1
Anthracene	BDL[b]	2.89	0.418	0.196	0.636	2.95
Fluoranthene	0.064	44.9	4.26	1.77	8.20	29.8
Pyrene	0.047	32.2	2.89	1.18	6.15	20.3
Benzo(a)anthracene	0.009	48.8	2.37	0.569	6.70	16.7
Chrysene	0.029	20.3	1.79	0.717	3.65	12.6
Benzo(b)fluoranthene	0.027	122	6.23	1.52	19.1	44.0
Benzo(k)Fluoranthene	0.021	15.4	0.763	0.248	2.22	5.39
Benzo(a)pyrene	0.014	41.3	1.71	0.310	6.66	12.1
Indeno(1,2,3-cd)pyrene	0.014	52.3	1.97	0.356	8.00	13.9
Dibenzo(a,h)anthracene	0.008	14.5	0.510	0.090	2.14	3.61
Benzo(g,h,j)perylene	0.018	57.1	2.14	0.398	8.60	15.1
Carcinogenic PAHs (ΣPAH_7)	0.120	288	15.6	4.01	46.7	40.5
ΣPAH_{16}	1.01	466	30.9	10.3	74.9	–

[a] Standard deviation.

[b] BDL: below detect limit.