Source, Characterization of Indoor Dust PAHs and the Health Risk on Chinese Children*

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Summary: Polycyclic aromatic hydrocarbons (PAHs) in indoor dust are one of the common exposure sources for children worldwide. The aim of this study is to explore PAHs pollution status in indoor dust and estimate health risk on Chinese children with big data. Weighted average concentration was used to analyze source and characterization of PAHs in indoor dust based on peer-reviewed literature. According to specific inclusion criteria, 17 studies were included finally to analyze weighted average concentration. The national average concentration of $\sum_{16}$PAHs was approximately 25.696 μg/g. The highest concentration of $\sum_{16}$PAHs was in Shanxi (2111.667 μg/g), and the lowest was in Hong Kong (1.505 μg/g). The concentrations in Shanxi and Guangdong were higher than national level and the over standard rate was 18.18%. The concentrations of individual PAHs varied greatly across the country, and Flu in Shanxi was the highest (189.400 μg/g). The sources of PAHs varied in different regions and combustion processes played a leading role. PAHs exposure through ingestion and dermal contact was more carcinogenic than inhalation. The incremental lifetime cancer risk model indicated that children lived in Shanxi were found in the highest health risk coupled with the highest BaPE concentration (54.074 μg/g). Although PAHs concentrations of indoor dust showed a downward trend from 2005 to 2018, indoor environmental sanitation should be improved with multidisciplinary efforts. Health standard should be possibly established to minimize children exposure to PAHs in indoor dust in China.
Key words: indoor dust; polycyclic aromatic hydrocarbons; source; children; China; health risk

Most people spend more than 90% of their time living indoors usually[1, 2], especially infants, the elderly and patients with chronic diseases. Therefore, people may be exposed to indoor pollutants for a long time which may be higher than those of outdoors[3]. Indoor dust is considered to be an exposure medium and a global indicator of residential pollution[1, 4]. Infants and young children are in highest risk since their hand-to-mouth habit[5]. A large number of epidemiological data show that indoor dust exposure is related to human health problems, including cardiovascular diseases, respiratory diseases and eye diseases[6, 7]. In addition, there is evidence that the harmful health effects may depend on the pollutants in indoor dust[8]. Polycyclic aromatic hydrocarbons (PAHs) have been detected as the main toxic components in indoor dust[9, 10].

PAHs are a class of persistent organic pollutants (POPs) generally existing in environment, consisted of two or more benzene rings. Tobacco smoking, cooking, kerosene burning, and wood burning are common sources of PAHs in indoor environment[11–14]. Sixteen PAHs have been given priority to control by the US Environmental Protection Agency (EPA) due to their carcinogenicity and mutagenicity[15]. Seven PAHs were classified as possible carcinogens or class 2B human carcinogens[16]. Concentration-dependent relationships between organic pollutants, such as PAHs, in indoor dust and human health issues have been established[17, 18].

The current researches mainly focus on the sources and pollution of PAHs outdoors and there are

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few studies on indoor PAHs pollution and risk analysis. The objectives of this study are: (1) to determine the concentrations and profiles of PAHs in indoor dust in China; (2) to distinguish the sources of PAHs in indoor dust in China; (3) to assess health risk on Chinese children via inhalation, dust ingestion and dermal contact using the incremental lifetime cancer risk (ILCR) standard model established by US EPA[19]; (4) to enrich the research of indoor PAHs in China and to provide basic information for indoor environment and health management of residents.

1 MATERIALS AND METHODS

1.1 Data Sources

Papers were identified through searching PubMed, Science Direct, Web of Science, China National Knowledge Infrastructure and WanFang online electronic database, using relevant terms for PAHs and indoor dust. The Boolean operators "AND" and "OR" were used to combine topic areas, such as [(indoor) OR (house) OR (residential) OR (residence)] AND [(China) OR (Chinese)] AND (dust) AND (PAHs). A total of 2533 articles were retrieved from January 1975 to December 2020. The flow chart of qualified literature screening process is shown in fig.1. According to the relevant requirements of this study, articles including raw numeric values of PAHs in indoor dust in China were retained, and review articles were excluded.

1.2 Inclusion Criteria and Exclusion Criteria

Inclusion criteria: (1) Research on PAHs concentrations in indoor dust in China; (2) raw numeric of PAHs concentrations in indoor dust can be extracted; (3) the unit of PAHs concentrations can be converted into μg/g.

Exclusion criteria: (1) Research on indoor dust concentration based on simulated model; (2) review articles; (3) research on air detection of airborne dust; (4) research on forced intervention measures during the experiment.

1.3 Data Standardization

The types and sample sizes of PAHs involved in published studies are different. Direct calculation of the average concentration will lead to deviation. In order to make the results more accurate, the weighted average concentration is used in this study. The specific formula is shown in (1):

$$C_t = \frac{\sum\limits_{n=1} C_n \times N_n}{\sum\limits_{n=1} N_n} \tag{1}$$

C_n represents the raw concentration of each sample in the original literature; N_n represents the sample size in the original literature; C_t represents the weighted average concentration of sample.

1.4 Source Analysis of PAHs

The diagnostic ratio was used to analyze the

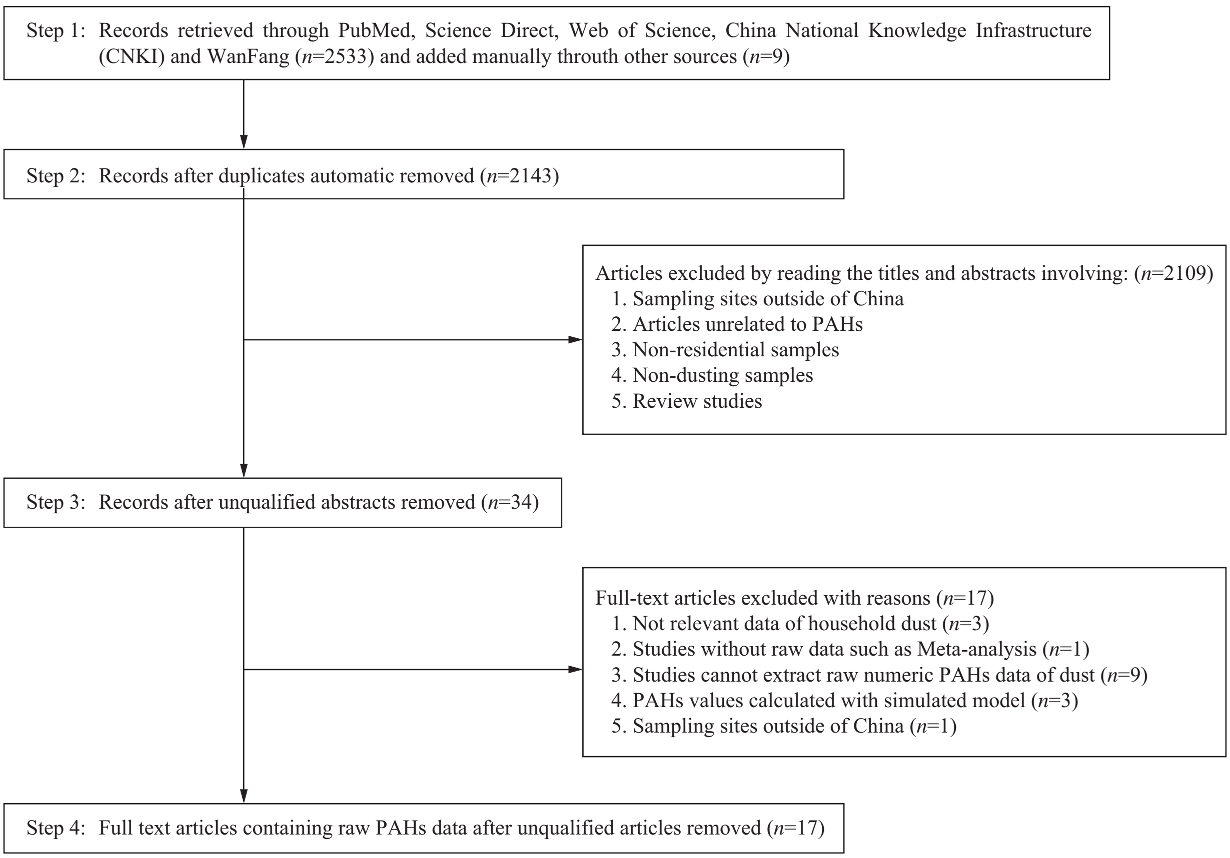

Fig. 1 Flow chart of qualified literature screening

source of PAHs[20, 21]. Different diagnostic ratios indicate specific emission sources[22], such as LMW/HMW (low molecular weight, 2–3 rings PAHs; high molecular weight, 4–6 rings PAHs), Ant/(Ant+Phe), Flu/(Flu+Pyr), BaA/(BaA+Chr), IcdP/(IcdP+BghiP) and ∑COMB/∑PAHs (∑COMB, including Flu, Pyr, BaA, Chr, BbF, BkF, BaP, IcdP, and BghiP).

1.5 PAHs Health Risk Assessment

ILCRs model was used to evaluate the latent cancer risk of exposure to indoor dust[23]. Humans expose to indoor dust PAHs via dermal contact, ingestion and inhalation. The ILCR (no unit) is calculated on the basis of three contact routes[23] and the following standard models from the US EPA are used:

$$ILCR_{Ingestion}=\frac{TEQ_{BaP}\times ED\times EF\times IR_{Ingestion}\times CSF_{Ingestion}\times\left(\frac{BW}{70}\right)^{\frac{1}{3}}}{BW\times AT\times 10^{6}} \quad (2)$$

$$ILCR_{Inhalation}=\frac{TEQ_{BaP}\times ED\times EF\times IR_{Inhalation}\times CSF_{Inhalation}\times\left(\frac{BW}{70}\right)^{\frac{1}{3}}}{BW\times AT\times PEF} \quad (3)$$

$$ILCR_{Dermal}=\frac{TEQ_{BaP}\times ED\times EF\times AF\times SA\times ABS\times CSF_{Dermal}\times\left(\frac{BW}{70}\right)^{\frac{1}{3}}}{BW\times AT\times 10^{6}} \quad (4)$$

The parameters in the child model for the risk assessment are shown in table S1. TEQ_{BaP} represents the summation of PAH concentrations calculated according to the toxic equivalent factor (*TEF*) and the toxic equivalent of BaP[24].

TEQ_{BaP} is calculated by formula (5):

$$TEQ_{BaP}=\sum_{i=1}^{n}(C_i\times TEF_i) \quad (5)$$

Where TEQ_{BaP} is total toxic equivalent concentration of BaP (μg/g); C_i is the mass concentration of the initial PAHs (μg/g); TEF_i is its toxic equivalency factor[24] (Nap, Acy, Ace, Fl, Phe, Flu and Pyr are 0.001. Ant, Chr and BghiP are 0.01. BaA, BbF, BkF and IcdP are 0.1. BaP is 1. DahA is 5)[22].

BaP is the first and strongest discovered environmental carcinogenic PAHs, and other HWM PAHs (BaA, BbF, BkF, IcdP and DahA) also have carcinogenic potential. The BaP equivalent (BaPE) is a useful index to assess the potential toxicity of PAHs in sediments, which can be calculated according to the following equation (6)[25, 26].

$$BaPE=BaA\times 0.06+(BbF+BkF)\times 0.07+BaP+DahA\times 0.60+IcdP\times 0.08 \quad (6)$$

1.6 Statistical Analysis

Excel2010 was used for data entry, data sorting and chart drawing, and SPSS21.0 was used for statistical analysis. Non-parametric Kruskal-Wallis *H* test was used for analyzing the difference in the level of risk among the three exposure routes. Spearman correlation analysis was used for analyzing the relationship between BaPE and $ILCR_{Ingestion}$, $ILCR_{Inhalation}$, $ILCR_{Dermal}$.

2 RESULTS

2.1 General Characteristics of Articles Included

A total of 17 articles[5, 8, 27–41] from 2007 to 2020 were included in this study through screening of qualified literature. Some critical information including provinces in China was collected. Samples were collected from 2005 to 2018 (2005, 2007, 2008, 2011, 2012, 2013, 2014 and 2018).

In 1976, the US EPA identified 16 kinds of PAHs as the priority control objects[15]. These are naphthalene (Nap, 2 rings), acenaphthene (Ace, 3 rings), acenaphthylene (Acy, 3 rings), fluorene (Fl, 3 rings), phenanthrene (Phe, 3 rings), anthracene (Ant, 3 rings), fluoranthene (Flu, 4 rings), pyrene (Pyr, 4 rings), chrysene (Chr, 4 rings), benz[a]anthracene (BaA, 4 rings), benzo[b]fluoranthene (BbF, 5 rings), benzo[k]fluoranthene (BkF, 5 rings), benzo[a]pyrene (BaP, 5 rings), dibenz[a, h]anthracene (DahA, 5 rings), benzo[g, h, i]perylene (BghiP, 6 rings) and indeno[1, 2, 3-cd]pyrene (IcdP, 6 rings). A total of 16 different kinds of PAHs were presented in previous literature. One study examined 18 kinds of PAHs, the other 6 studies examined 15 kinds of PAHs (table 1). The highest detection rate was 99.88% of Flu, Pyr and Chr. The lowest detection rate was 97.30% of Ant.

2.2 Characteristics of PAHs Concentrations in 11 Provinces

The concentration of different PAHs varied widely across the country (fig. 2). The concentration of Flu in Shanxi province was the highest (189.400 μg/g) and the concentrations of Ace, Acy and Fl were relatively

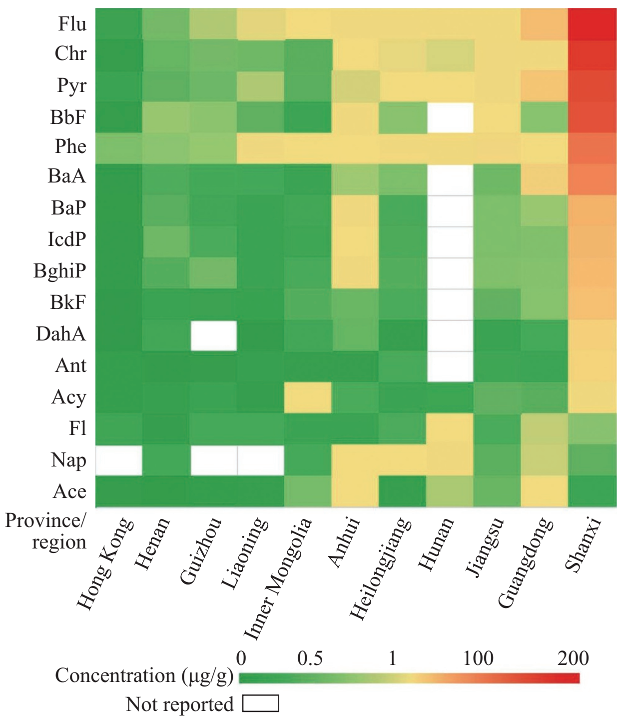

Fig. 2 Heat map of indoor dust concentration of 16 PAHs in 11 provinces/region

Table 1 Summary of PAHs (μg/g) concentrations in indoor dust and literature information in peer-reviewed studies

Provinces	Sam-year	Sam-size	Nap	Ace	Acy	Fl	Phe	Ant	Flu	Pyr	Chr	BaA	BbF	BkF	BaP	DahA	BghiP	IcdP	BeP	BEA	BjF	∑PAHs	Reference
Shanxi	2005–2005	1	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	–	–	–	ND	Zhou *et al*. 2007
Shanxi	2005–2005	1	1.200	0.500	8.000	1.800	240.000	20.000	530.000	355.000	410.000	195.000	325.000	60.000	85.000	25.000	70.000	80.000	–	–	–	5760.000	Zhou *et al*. 2007
Shanxi	2005–2005	1	0.020	0.010	0.200	0.100	10.000	0.900	16.000	14.000	24.000	9.000	20.000	3.000	5.000	1.300	4.000	4.000	–	–	–	255.000	Zhou *et al*. 2007
Shanxi	2005–2005	1	0.030	0.020	0.500	0.030	10.300	1.400	22.200	18.500	27.000	13.000	23.000	1.900	5.400	1.200	3.000	3.500	–	–	–	320.000	Zhou *et al*. 2007
Guangdong	2007–2008	6	–	0.047	0.068	0.289	0.737	0.061	0.139	0.158	0.092	0.055	0.137	0.037	0.034	0.007	0.050	0.055	–	–	–	2.000	Pan *et al*. 2010
Hong Kong	2007–2008	9	–	0.043	0.054	0.204	0.591	0.053	0.118	0.129	0.039	0.028	0.038	0.015	0.017	0.009	0.033	0.026	–	–	–	1.505	Pan *et al*. 2010
Jiangsu	–	61	0.424	–	0.573	0.425	5.353	0.241	4.301	3.035	2.481	0.657	1.437	0.482	0.688	0.280	0.641	0.570	–	–	–	21.096	Cui *et al*. 2012
Guangdong	2011–2012	50	0.140	0.090	0.090	0.260	0.730	0.040	1.130	1.360	0.830	0.660	0.440	0.440	0.310	0.280	0.650	0.800	–	–	–	8.270	Wang *et al*. 2013
Guangdong	2011–2012	20	0.400	0.110	0.190	0.710	5.620	0.190	6.230	7.030	4.370	1.760	1.845	1.845	2.300	0.440	1.330	0.820	–	–	–	35.200	Wang *et al*. 2013
Jiangsu	2008–2008	215	0.400	0.500	–	0.300	3.700	0.200	3.300	2.200	2.000	0.500	1.100	0.400	0.600	0.200	0.600	0.600	–	–	–	16.400	Wang *et al*. 2014
Jiangsu	2011–2011	203	0.400	0.500	–	0.300	3.700	0.200	3.300	2.200	2.000	0.500	1.100	0.400	0.600	0.200	0.600	0.600	–	–	–	16.400	Wang *et al*. 2014
Guizhou	2012–2012	23	ND	0.080	0.160	0.380	0.580	0.050	1.140	0.650	0.720	0.350	1.070	0.160	0.290	–	0.580	0.350	0.200	0.470	0.460	7.690	Yang *et al*. 2015
Guizhou	2012–2012	23	ND	0.050	0.130	0.270	0.730	0.040	0.880	0.540	0.600	0.290	0.800	0.110	0.250	–	0.540	0.310	0.160	0.400	0.280	6.380	Yang *et al*. 2015
Guizhou	2012–2012	27	ND	0.040	0.120	0.200	0.770	0.060	0.420	0.340	0.330	0.160	0.190	0.070	0.170	–	0.480	0.290	0.110	0.240	0.280	4.270	Yang *et al*. 2015
Henan	2012–2012	1	0.589	0.120	0.173	0.153	1.647	0.072	1.211	1.153	0.593	0.363	1.122	0.104	0.070	0.203	0.951	0.857	–	–	–	9.380	Yang *et al*. 2015
Henan	2012–2012	1	0.236	ND	0.019	0.013	0.130	ND	0.184	0.132	0.148	0.104	0.105	0.020	0.119	0.040	0.183	0.162	–	–	–	1.594	Yang *et al*. 2015
Henan	2012–2012	1	0.081	ND	0.025	0.021	0.168	ND	0.203	0.129	0.167	0.099	0.144	0.016	0.030	0.039	0.188	0.160	–	–	–	1.471	Yang *et al*. 2015
Henan	2012–2012	1	0.101	0.006	0.037	0.039	0.568	ND	0.836	0.666	0.747	0.482	0.571	0.108	0.268	0.154	1.006	0.901	–	–	–	6.489	Yang *et al*. 2015
Henan	2012–2012	1	0.061	0.005	0.024	ND	0.328	0.011	0.867	0.756	1.253	0.164	0.796	ND	0.019	0.070	0.274	0.113	–	–	–	4.741	Yang *et al*. 2015
Henan	2012–2012	1	0.243	0.029	0.106	0.220	2.154	0.018	1.094	0.643	1.018	0.255	1.155	0.035	0.343	0.056	0.221	0.085	–	–	–	7.675	Yang *et al*. 2015
Henan	2012–2012	1	0.185	0.011	0.075	0.064	0.744	ND	0.633	0.497	0.836	0.867	2.388	ND	0.850	0.115	0.493	1.047	–	–	–	8.803	Yang *et al*. 2015
Henan	2012–2012	1	0.161	0.006	0.053	0.037	0.368	ND	0.355	0.269	0.606	0.777	0.939	0.009	0.917	0.150	0.228	0.785	–	–	–	5.662	Yang *et al*. 2015
Henan	2012–2012	1	0.308	0.003	0.193	0.367	5.551	ND	1.861	0.825	0.278	0.231	0.275	0.026	0.235	0.039	0.146	0.225	–	–	–	10.563	Yang *et al*. 2015
Henan	2012–2012	1	0.339	0.010	0.073	0.042	0.431	ND	0.424	0.696	0.436	0.388	1.006	0.042	0.683	0.184	0.385	0.435	–	–	–	5.574	Yang *et al*. 2015
Henan	2012–2012	1	0.226	0.016	0.092	0.071	0.633	ND	0.540	0.377	1.049	0.928	1.693	0.007	0.018	0.008	0.732	0.042	–	–	–	6.431	Yang *et al*. 2015
Henan	2012–2012	1	2.057	0.013	0.057	0.091	1.003	ND	0.898	0.773	1.148	1.617	2.129	0.765	1.742	0.107	1.467	1.959	–	–	–	15.826	Yang *et al*. 2015
Henan	2012–2012	1	0.210	0.010	0.033	0.076	0.825	ND	0.707	0.470	0.603	0.278	1.388	ND	0.019	0.092	0.396	0.594	–	–	–	5.698	Yang *et al*. 2015
Henan	2012–2012	1	0.112	0.007	0.041	0.043	0.630	ND	0.927	0.738	0.828	0.534	0.633	0.120	0.297	0.171	1.115	0.999	–	–	–	7.194	Yang *et al*. 2015
Henan	2012–2012	1	0.151	0.014	0.076	0.062	0.678	ND	0.606	0.446	0.895	0.550	1.375	0.123	0.591	0.225	0.383	1.022	–	–	–	7.198	Yang *et al*. 2015
Henan	2012–2012	1	0.802	0.009	0.065	0.055	0.728	ND	0.734	0.556	0.761	0.439	1.776	ND	0.453	0.150	0.464	0.795	–	–	–	7.786	Yang *et al*. 2015
Henan	2012–2012	1	0.088	0.007	0.043	0.046	0.979	ND	1.080	0.603	0.596	0.122	1.239	ND	0.037	0.060	0.356	0.505	–	–	–	5.763	Yang *et al*. 2015

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Provinces	Sam-year	Sam-size	Nap	Ace	Acy	Fl	Phe	Ant	Flu	Pyr	Chr	BaA	BbF	BkF	BaP	DahA	BghiP	IcdP	BeP	BEA	BjF	∑PAHs	Reference
Henan	2012–2012	1	0.315	0.023	0.141	0.118	1.326	ND	1.378	0.881	1.576	1.511	3.284	1.532	3.387	1.588	2.242	2.511	–	–	–	21.814	Yang *et al.* 2015
Henan	2012–2012	1	0.227	0.010	0.138	0.067	0.875	ND	1.095	0.795	1.220	1.208	2.023	0.864	2.146	0.103	1.424	3.087	–	–	–	15.284	Yang *et al.* 2015
Henan	2012–2012	1	0.346	0.017	0.150	0.132	0.983	ND	0.741	0.579	1.110	0.373	2.515	0.061	1.699	0.012	0.391	4.477	–	–	–	13.586	Yang *et al.* 2015
Hunan	–	15	2.856	0.763	0.170	1.044	2.793	ND	3.230	1.464	0.921	ND	ND	ND	ND	ND	ND	ND	–	–	–	14.049	Kang *et al.* 2015
Anhui	2014–2014	13	1.820	1.720	0.310	0.140	1.220	0.070	2.130	0.910	1.250	0.730	3.320	0.480	2.000	0.460	2.570	1.430	–	–	–	20.700	Li *et al.* 2016
Jiangsu	–	16	0.140	0.030	0.043	0.010	1.300	0.060	1.210	0.720	0.980	0.360	0.920	0.920	0.280	0.150	1.020	0.720	–	–	–	7.940	Xiang *et al.* 2016
I-Mongolia	2014–2014	46	0.275	0.668	1.356	0.121	1.246	0.063	1.346	0.291	0.340	0.113	0.096	0.422	0.239	0.278	0.334	0.215	–	–	–	7.404	Zhou *et al.* 2017
Henan	2014–2014	22	0.185	–	0.106	0.033	0.309	0.022	0.274	0.233	0.157	0.138	0.119	0.050	0.081	0.269	0.105	0.023	–	–	–	2.100	Cao *et al.* 2017
Jiangsu	2013–2013	7	0.045	0.009	0.005	0.022	0.066	0.270	0.251	0.325	0.216	0.134	0.263	0.249	0.195	0.024	0.117	0.102	–	–	–	2.294	Zhang *et al.* 2019
Jiangsu	2013–2013	1	0.004	ND	ND	ND	ND	0.018	0.025	0.016	0.018	0.009	0.018	0.018	0.011	ND	0.008	ND	–	–	–	0.145	Zhang *et al.* 2019
Heilongjiang	2013–2014	25	1.260	0.069	0.152	0.329	3.130	0.300	2.190	1.840	0.976	0.592	0.637	0.316	0.301	0.069	0.361	0.325	–	–	–	12.800	Li *et al.* 2019
LN&I-Mongolia	2018–2018	26	–	0.075	0.066	0.280	2.010	0.095	0.955	0.780	0.507	0.254	0.420	0.104	0.147	0.020	0.145	0.147	–	–	–	6.000	Yang *et al.* 2020
Guangdong	–	21	3.060	4.410	1.300	2.520	0.886	0.536	107.000	78.400	8.700	40.600	0.089	0.089	0.338	0.162	0.045	0.121	–	–	–	248.256	Luo *et al.* 2020

Sam-year means sampling year. Sam-size means sampling size. ND means under the detection limit. LN&I-Mongolia represents Liaoning and Inner Mongolia.

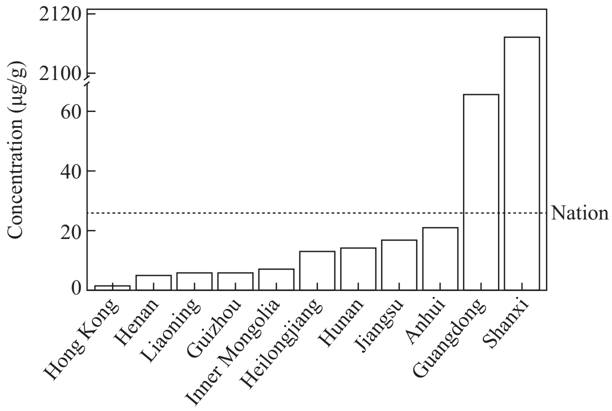

Fig. 3 Comparison of $\sum_{16}$PAHs concentrations in 11 provinces/region with national concentrations

low. Fig. 3 shows the analysis of the weighted average concentration of indoor dust $\sum_{16}$PAHs. The highest residential $\sum_{16}$PAHs concentration was in Shanxi (2111.667 μg/g), and the lowest was in Hong Kong (1.505 μg/g). The national average concentration of $\sum_{16}$PAHs was 25.696 μg/g in this study. Only the concentration in Shanxi and Guangdong was higher than the national level, and over standard rate was 18.18%. The composition profiling of PAHs was different in 11 provinces/region (fig. 4). There were mainly 4 rings PAHs (83.13% and 59.22%) in Guangdong and Shanxi, respectively, 3 rings PAHs (63.53%) in Hong Kong, 3 and 4 rings PAHs (77.97%, 67.21% and 60.92%) in Liaoning, Jiangsu and Inner Mongolia, respectively.

2.3 Temporal Differences of Sample-weighted Mean Concentration of Indoor Dust PAHs

PAHs concentration in Shanxi province from one study was significantly higher than that in other regions. Therefore, the data of Shanxi province (2005) were removed in data analysis to avoid information errors. The weighted average concentrations of different rings and different kinds of PAHs were calculated and the trend of their variation over time was plotted (figs. 5 and 6). Four rings PAHs were the highest in each time period, especially in 2005–2010 (1.876 μg/g in fig. 5A) and rings showed a downward trend with time (fig. 5B). $\sum_{16}$PAHs were also the highest (15.442 μg/g) in 2005–2010 but the lowest in 2016–2020 (6.000 μg/g). In 2005–2010, the concentration of Phe was the highest (3.501 μg/g), DahA was the lowest in 2016–2020 (0.021 μg/g) in fig. 6. Nap, Acy, BaA, BkF, DahA and BghiP were higher in 2011–2015 than in 2005–2010. Overall, the concentrations of PAHs showed a downward trend.

2.4 Source Analysis of PAHs

HMW PAHs are mainly originated from high-temperature combustion process and LMW PAHs are chiefly originated from low or moderate temperature combustion[42]. Table S2 shows the principle of PAHs source classification. In general, combustion induces the production of relatively higher concentrations of

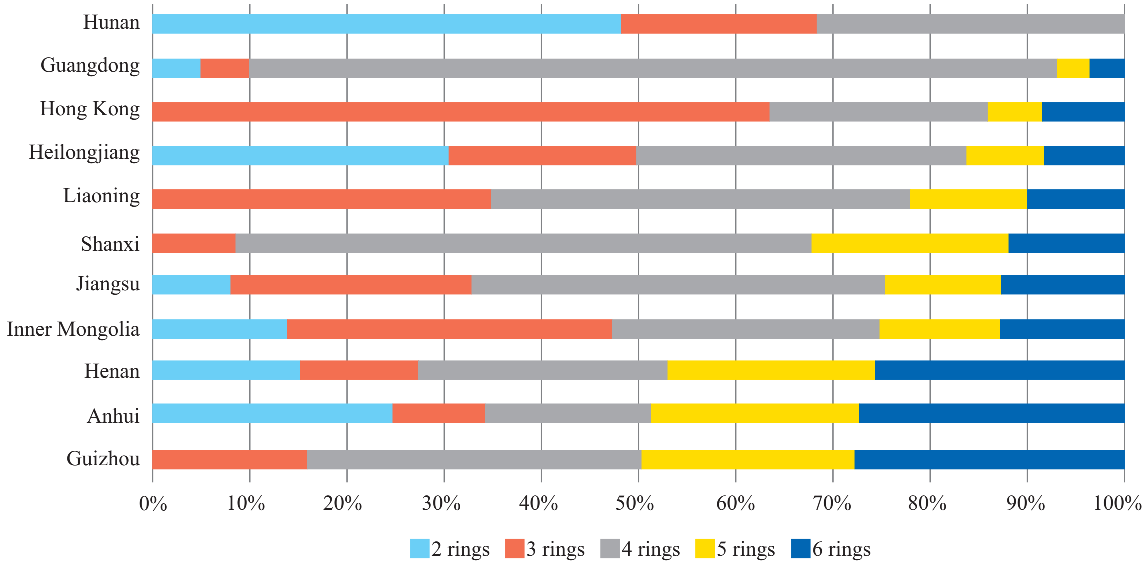

Fig. 4 PAHs composition profiling characteristics of indoor dust in 11 provinces/region

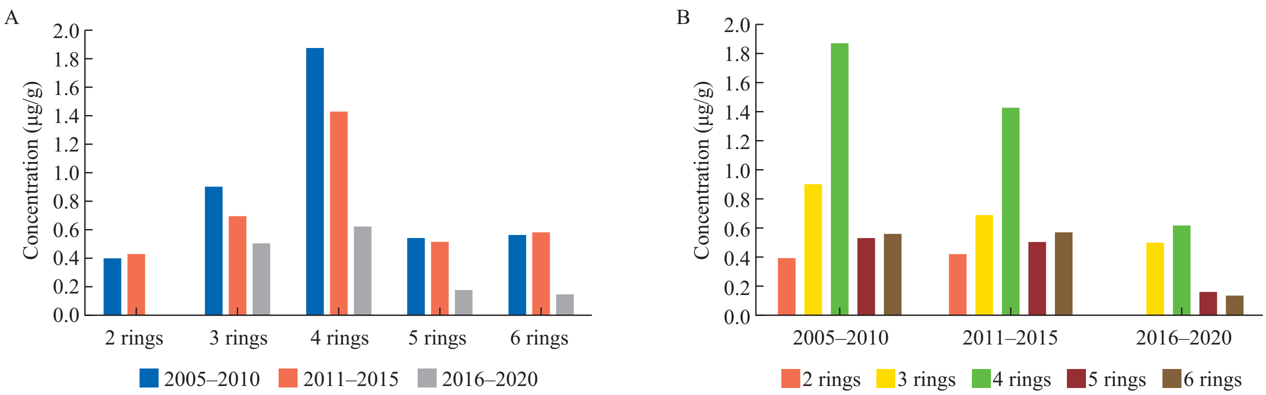

Fig. 5 Temporal differences of sample-weighted mean concentration of PAHs with different rings
The data in Shanxi province are not included.

∑COMB; therefore, a large proportion of ∑COMB is a characteristic of PAHs that originated from combustion processes[43].

Table S3 and fig. 7 show diagnostic ratios of PAHs in indoor dust in 11 provinces/region. The ratios of the ∑COMB/∑PAHs ranged from 0.294 to 0.919, which inferred that the PAHs in samples mainly originated from combustion processes. In Henan (fig. 7B, 7E and 7F), Shanxi (fig. 7B), Anhui (fig. 7F), Heilongjiang (fig. 7F) and Guangdong (fig. 7F), PAHs may originate from combustion of coal, grasses, and wood. In Hong Kong (fig. 7B), Inner Mongolia (fig. 7E), Guizhou (fig. 7E), PAHs may originate from petroleum combustion.

2.5 Health Risk Assessment of PAHs Exposure

Human exposure to PAHs may occur via ingestion, inhalation, and dermal contact[44]. The most sensitive subpopulation is young children because of their hand-to-mouth activity, whereby contaminated dust can be readily ingested[45]. The ILCR formula with default parameters was used to identify the different exposure pathways for children to evaluate the health risk. Table 2 presents the calculated ILCR values. Previous studies indicated that an ILCR value of 10^{-6} generally represented an acceptable level, an ILCR value from 10^{-6} to 10^{-4} indicated a potential human carcinogenic risk, and an ILCR value higher than 10^{-4} indicated a serious carcinogenic risk[46, 47]. The calculated results indicated that PAHs exposure posed a potential human carcinogenic risk in Shanxi, Guangdong and Anhui. The ILCR value was the highest in Shanxi province and the lowest in Hunan province. And the cancer risk levels via ingestion, dermal contact and inhalation were 5.301×10^{-8} *vs.* 2.572×10^{-4}, 6.608×10^{-8} *vs.* 3.207×10^{-4}, and 1.028×10^{-12} *vs.* 4.988×10^{-9}, respectively. Since the level of risk among the different exposure pathways was non-normal distribution, the non-parametric Kruskal-Wallis *H* test was performed to compare the difference. The results indicated that the health risk level was different among different exposure routes. All these results indicated that ingestion and dermal contact exposure was more carcinogenic than inhalation[22].

The concentration of BaPE ranged from 0.030 μg/g to 54.074 μg/g. Spearman correlation analysis revealed there was a positive correlation between BaPE

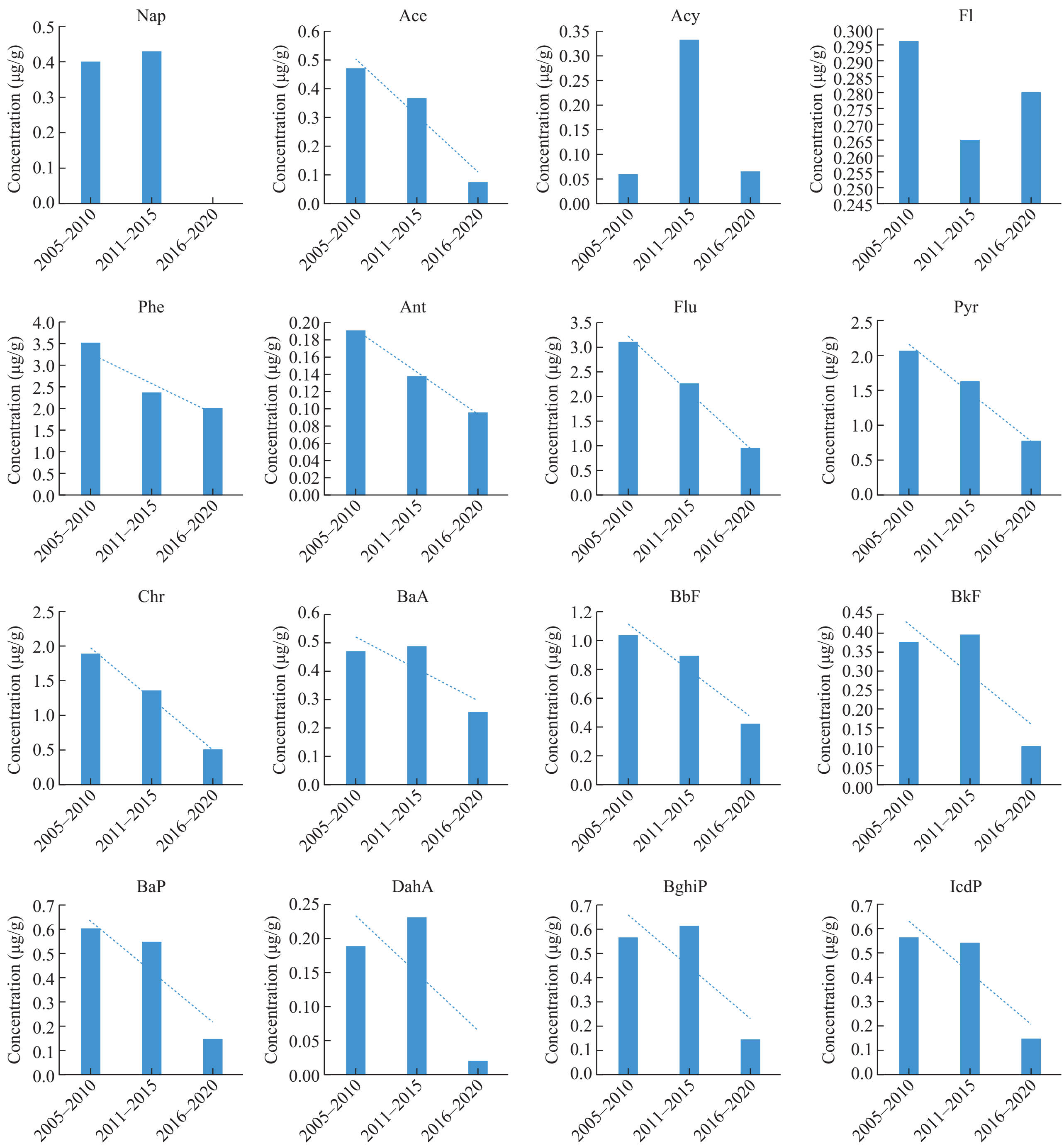

Fig. 6 Temporal differences of sample-weighted mean concentration of 16 kinds of PAHs
The data in Shanxi province are not included.

Table 2 Carcinogenic risk values of PAHs (μg/g) in indoor dust from different exposure routes

Provinces/region	TEQ_{BaP} (μg/g)	$ILCR_{Ingestion}$	$ILCR_{Inhalation}$	$ILCR_{Dermal}$
Hunan	0.022	5.301×10^{-8}	1.028×10^{-12}	6.608×10^{-8}
Hong Kong	0.075	1.849×10^{-7}	3.584×10^{-12}	2.304×10^{-7}
Liaoning	0.354	8.706×10^{-7}	1.688×10^{-11}	1.085×10^{-6}
Guizhou	0.381	9.388×10^{-7}	1.820×10^{-11}	1.170×10^{-6}
Heilongjiang	0.857	2.111×10^{-6}	4.093×10^{-11}	2.631×10^{-6}
Inner Mongolia	1.481	3.647×10^{-6}	7.071×10^{-11}	4.546×10^{-6}
Jiangsu	1.522	3.746×10^{-6}	7.264×10^{-11}	4.670×10^{-6}
Henan	1.681	4.139×10^{-6}	8.025×10^{-11}	5.159×10^{-6}
Guangdong	3.288	8.096×10^{-6}	1.570×10^{-10}	1.009×10^{-5}
Anhui	4.943	1.217×10^{-5}	2.360×10^{-10}	1.517×10^{-5}
Shanxi	104.490	2.572×10^{-4}	4.988×10^{-9}	3.207×10^{-4}

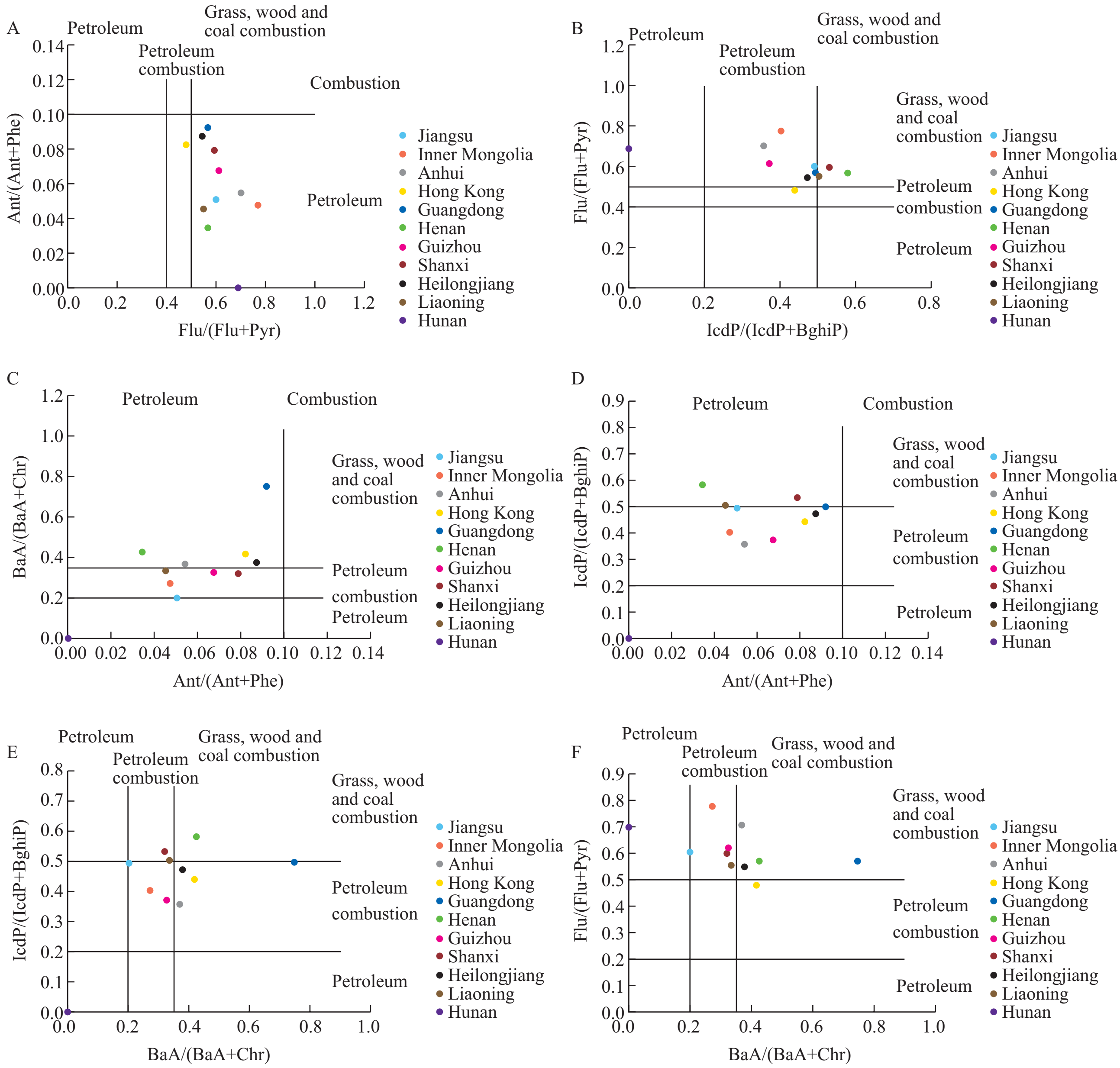

Fig. 7 PAHs source analysis based on the diagnostic ratios in 11 provinces/region

and $ILCR_{Ingestion}$, $ILCR_{Inhalation}$, $ILCR_{Dermal}$. Therefore, there was the highest health risk level in Shanxi where BaPE concentration was also at the highest level (fig. 8). There were low BaPE concentration and low health risk level in Hunan.

3 DISCUSSION

This article reviewed the literature on PAHs contamination in indoor dust in 11 provinces across China. PAHs concentrations varied greatly across the country, and they were the highest in Shanxi and the lowest in Hong Kong. The concentrations in Shanxi and Guangdong were higher than national level in this study. The source of PAH varies in different regions, but almost originates from combustion processes. PAHs exposure through ingestion and dermal contact were more carcinogenic than inhalation. Shanxi had highest BaPE concentration and health risk level.

In China, few papers evaluated the PAHs contamination in dust, especially in indoor dust[48]. While in the other countries, a large number of papers were related to indoor dust PAHs, little is known about the impact of indoor pollution sources on personal PAHs exposure[49]. In recent years, the number of published articles related to indoor dust of PAHs increased exponentially. However, in this paper, 17 included studies were published from 2007 to 2020, and there were no data records before 2007, indicating that indoor dust PAHs draw more attention just in recent years.

Sixteen PAHs have been prioritized for control by

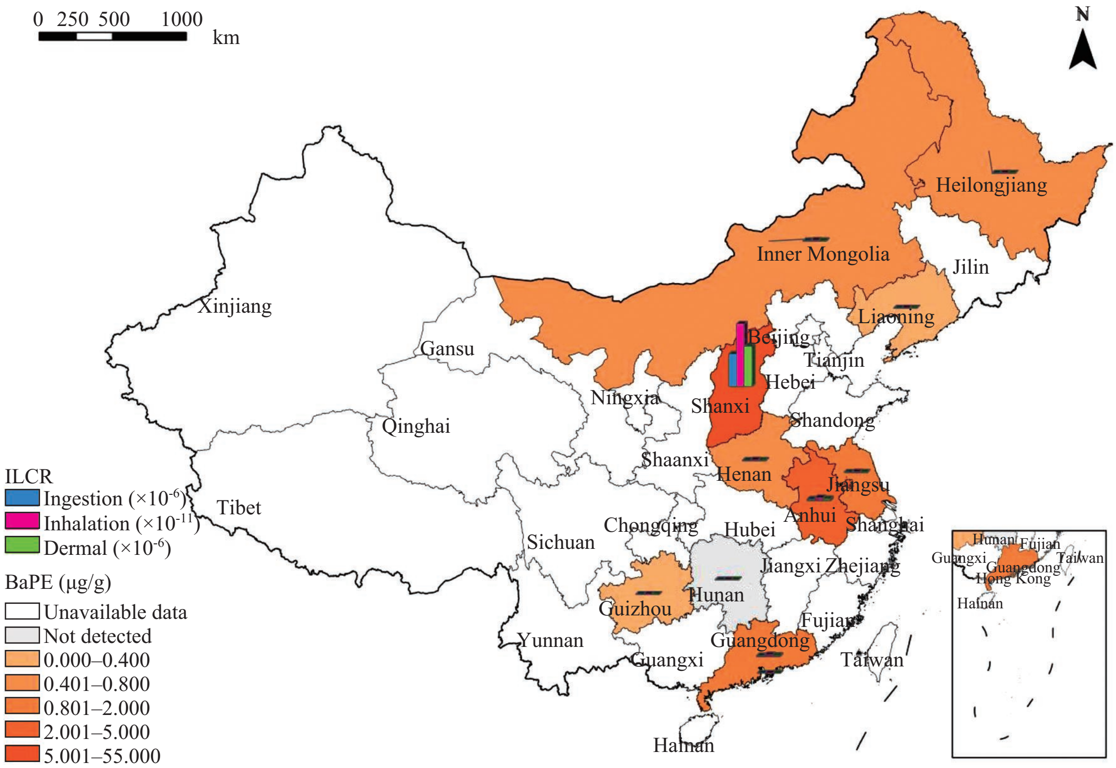

Fig. 8 Geographical distribution of BaPE exposure and ILCR values of three exposure pathways in 11 provinces/region

the US EPA because of their carcinogenic and mutagenic properties[15]. These 16 PAHs were detected and analyzed in many studies. The concentrations of Ace, Acy and Fl were relatively low in 11 provinces/region and Flu in Shanxi was the highest. The concentration of PAHs varied greatly across the country, with the lowest average concentration in Hong Kong and the highest in Shanxi. Shanxi is the leading province in coal production in China, and coal is commonly used indoors for cooking and heating in this region[27]. The major products of thermal decomposition of coal include PAHs as well as related nitrogen- and sulfur-containing PAHs[50, 51]. The national average concentration of $\sum_{16}$PAHs was approximately 25.696 μg/g in this study. It was higher than those of settled house dust (SHD) samples collected from homes in urban areas of San Diego County, CA, USA (0.163–4.390 μg/g; median, 0.990 μg/g)[14] but was lower than those in Ohio, USA (11.1–513 μg/g; median, 47.4 μg/g)[9], and Texas, USA (1.12–341 μg/g; median, 28.8 μg/g) [10]. The PAHs concentrations of indoor dust tended to decrease with time approximately. It may be because people's awareness of environmental protection has been raised, and the national governance of indoor and outdoor environment has been done. However, due to the lack of consecutive surveillance data of PAHs, the actual changing trend of PAHs can't be observed.

PAHs diagnostic ratios have been used to identify different sources that contribute PAHs to environmental samples[52]. PAHs in most samples mainly originate from combustion processes. Our results indicated that petroleum combustion was the origin of PAHs in Hong Kong, Inner Mongolia, and Guizhou. PAHs may originate from combustion of coal, grasses, and wood in Henan, Anhui, Shanxi, Guangdong and Heilongjiang. The PAHs source of indoor dust in other provinces may originate from complex sources. Vehicle restriction and switching clean energy can effectively reduce indoor PAHs pollution levels. However, this method is not an exact method of distinguishing sources and only provides a qualitative estimation[53]. If we want to identify the sources of PAHs in indoor dust more clearly, we need more data to analyze or use other analytical methods such as Principal Component Analysis. Gustafson *et al*[54] concluded that the concentration of PAHs of households using wood burning for heating in winter in Sweden was significantly higher. Ansari *et al*[55] found that indoor PAHs mainly originated from cooking fuel in rural India such as wood. Abbasi *et al*[56] showed that indoor PAHs may come from petroleum in developing and oil-rich countries such as Iran. So the source of PAHs in indoor dust is related to the local industrial structure and the lifestyle of residents.

BaP is the first environmental carcinogen to be discovered with highly carcinogenic PAHs. The BaPE value is often used as an index to evaluate the toxicity of PAHs. BaPE concentration was the highest in Shanxi which is the leading province in coal production in China and provides as much as one-third of China's coal[27]. High temperature combustion process (such as vehicular exhaust, mining processing activities)[35] and living habits (such as cooking, heating, smoking)[57] may make contributions to the high BaPE concentration.

At present, there is not a perfect standard for health risk assessment of PAHs in indoor dust in China. The results of exposure assessment of PAHs in house dust to children is quantitative and therefore is much more comparable than health risk of different endpoints. The ILCR model is widely used now[23]. The ILCR is taken as an ensign to identify the age-specific potential cancer risks in the study of human exposure to environmental PAH pollution sources[54, 58]. Results indicate that dermal contact and ingestion exposure are more carcinogenic than inhalation[22]. Dermal contact is the most dominant exposure route, which is easily to be ignored[34]. Due to behavioral characteristics such as hand-to-mouth activities and food handling, non-dietary inhalation of contaminants is potentially the main route of exposure for children. Dermal contact and ingestion exposures are also important route for children and are associated with behaviors such as crawling on the floor and contact with dirt and grass[59]. In addition, early development of organ, nervous, and immune systems may enhance the carcinogens sensitivity in children[56]. However, the results calculated based on ILCRs model still have certain limitations. Exposure by ingestion, inhalation, or dermal contact is according to the size of the dust particles. Li *et al*[39] found that inhalation was the main exposure route for LWM PAHs, and ingestion was the major route for HWM PAHs. Most current research is unaware of this problem. So, the accuracy of the assessment results will be affected to some extent[37]. Therefore, relevant standards and models for health risk assessment of PAHs in indoor dust need to be improved to be more suitable for the actual conditions of indoor environment in China, and provide basic information for residents' protection.

There are several limitations in this paper. Although we searched literature of nearly 45 years, raw numeric concentrations of PAHs in indoor dust in other provinces were lack to estimate national concentration of PAHs in indoor dust. In addition, different instruments and methods were chosen to analyze dust samples in the 17 studies, resulting in different detection limits of PAHs. PAHs were detected with different methods including gas chromatography-mass spectrometry[28, 33, 35, 37–39], high performance liquid chromatography[27, 29, 30, 34], gas chromatograph equipped with a flame ionization detector[31]. Moreover, the sampling requirements or sampling sites were not consistent in these papers. The influence of these confounding factors existed and would bring some bias on the results in this study to some extent.

In conclusion, this study analyzed public literature in the last 45 years to explore PAHs pollution characterization in indoor dust nationwide. PAHs are widely found in indoor dust. There are differences in PAHs concentrations (they were the highest in Shanxi province) and sources in indoor dust at different provinces. Although there is a downward trend of PAHs over time and the indoor environment condition has been greatly improved, its negative impact can't be ignored. Especially children as sensitive subpopulation, efforts should be conducted to prevent and control their dermal contact and ingestion exposures. There are rare articles similar to this study. Therefore, in our opinion, the study on PAHs in indoor dust from Chinese households should be further strengthened.

Conflict of Interest Statement

The authors declare that they have no conflict of interest.

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