Global NEST Journal, Vol 19, No 3, pp 540-546

Concentrations and potential risk assessment of polycyclic aromatic hydrocarbons (PAHs) from indoor dust of Bushehr, Iran

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Received: 11/06/2017, Accepted: 02/10/2017, Available online: 24/10/2017
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Abstract

Indoor dust samples were collected in the summer of 2017 from 10 residential buildings, 7 commercial buildings and 7 official buildings of Bushehr. Sixteen polycyclic aromatic hydrocarbons (PAHs) were analyzed by GC–MS. The sum concentration of the 16 PAHs in dust spanned between 87.8 to 14619 $mgkg^{-1}$d.w. with a mean of 2211.6 $mgkg^{-1}$d.w. The mean concentration of PAHs for the dust samples from the commercial buildings was higher than from residential buildings and, from residential building was higher than from official buildings. The cancer risk through dermal and ingestion route ranged from 10^{-6} to 10^{-5} across all the dust samples, while the average of cancer risk through inhalation pathway was 10^{-9} to 10^{-12}, about 10^4 to 10^6 times lower, when compared with other two pathways (ingestion and dermal contact). Also, the level of cancer risk for children was significantly higher than the corresponding risk for adults. The results of this work would be suitable to enhance the quality of living environment and human health in Bushehr province.

Keywords: Polycyclic aromatic hydrocarbons, Indoor dust, Risk assessment, Bushehr

1. Introduction

Since people (especially kids, housewives and old people) spend approximately 85% of their lifetime indoors (Amorello *et al.*, 2015; Rudel *et al.*, 2003), indoor air quality has a prominent role in their health (Maertens *et al.*, 2008). Moreover, healthy housing environment is one of the key indexes to evaluate social stability and development (Li *et al.*, 2009; Tao *et al.*, 2016); however, rapid industrialization and urbanization ha caused deterioration of human settlement environments in cities through increasing pollutant emissions (Kang *et al.*, 2011; Whitehead *et al.*, 2011; Xu and Shu, 2012). In particular, contaminants accumulate in the indoor environment via multiple routes, after which they can enter human body through inhalation, ingestion, or direct skin absorption, where they are hazardous to human health and inevitably pose health risks (Song and Xu, 2011; Whitehead *et al.*, 2011). Therefore, indoor environments should be controlled for efficient risk management and human health protection (Mannino and Orecchio, 2008).

Indoor dust is one of the major matrixes that can be utilized as a useful indicator for indoor air contaminants transport and deposition(Xu and Shu, 2014). Indoor dust also acts as a reservoir for pollutants and can provide useful information for estimating human exposure to contaminants(Butte and Heinzow, 2002). The presence of various environmental contaminants such as heavy metals, flame retardants, plasticizers, asbestos, smoke residues, pesticides, phthalates, and PCBs has been surveyed in indoor dust (Lioy *et al.*, 2002; Mirzaei *et al.*, 2015; Rasmussen *et al.*, 2001; Rudel *et al.*, 2003). Among these contaminants, polycyclic aromatic hydrocarbons (PAHs) are one of the major human health concerns, owing to their ubiquitous occurrence and high carcinogenic and mutagenic potential (Kameda *et al.*, 2005). In indoor environments, elevated levels of PAHs have been found to be mainly produced by endogenous sources such as domestic heating, cooking, wood-burning, tobacco smoke, decorative candle and incense burning (Castro *et al.*, 2011). These semi volatile compounds accumulate on dusts, and the dust that is settled indoors can be a sink for these pollutants (Roberts *et al.*, 2009). Although the concentration and sources of outdoor PAHs have been widely studied, dust-bounded PAHs in indoor environments (such as residential, commercial and official buildings) are still a challenging issue. Moreover, indoor air quality in these environments significantly influences the human welfare and work performance (Fanger, 1998; Wolkoff, 2013). For example, it has been shown that some of the most common symptoms in offices caused by polluted indoor air are headache, irritation, tiredness, and dry or itchy eyes (Wolkoff, 2013), with these symptoms causing 2% reduction in efficiency and subsequently economic loss (Buchanan *et al.*, 2008).

This paper was aims to investigate the concentration of PAHs in indoor dust at buildings, located in Bushehr city center, Iran. Moreover, the potential health risks of PAHs in indoor dust were also evaluated.

2. Materials and methods

2.1. Sample collection

Indoor dust samples used in this study were collected in June 2016 from 24 indoor environments (including 18 residential buildings (URB1 to URB10), seven commercial buildings (UCB11 to UCB17) and seven official buildings (UOB18 to UOB24) of Bushehr city (Table1), according to the standardized protocol (Stuart *et al.*, 2008) (Table1). To control the impact of rainfall, sampling was conducted in dry seasons (summer [July]) and all samples were collected after two consecutive weeks without precipitation. About 20 g of dust samples was taken in each site using a polyethylene brush and a stainless steel scoop (for transferring the dust samples into plastic bags) rinsed with acetone under exact procedures. To avoid cross-contamination, all sampling equipment was cleaned after each sampling by ultrasound, rinsed with deionized water and then air dried. During the sampling, the characteristics of the building such as building type, floor types, number of smokers, number of electronic devices, cooking conditions, ventilation type and distance off road were also recorded. Each sample was transferred into clean plastic bags, sealed and then refrigerated (4 °C) on site. The samples were protected from sunlight with aluminum foils and then rapidly transported to the laboratory. In the laboratory, the dust samples were put at – 20 °C until further analysis (Raeisi *et al.*, 2016).

Table 1. Areas, descriptive profile and ongoing activities of indoor sampling sites

Station	Area type	Environment	Characteristics
URB1	Urban	Bedroom	Away from street, no smokers, natural ventilation 1 windows, cooking with gas
URB2	Urban	Kitchen	Near the street, no smokers, natural ventilation 2 windows cooking with gas
URB3	Urban	Bedroom	Away from street, smokers, natural ventilation no windows cooking with electricity
URB4	Urban	Living room	Heavy traffic, smokers, natural ventilation 2 windows, cooking with gas
URB5	Urban	Kitchen	Near the street, smokers, natural ventilation no windows, cooking with biomass
URB6	Urban	Bedroom	Heavy traffic, smokers, natural ventilation 1 windows, cooking with biomass
URB7	Urban	Bedroom	Moderate traffic, no smokers, natural ventilation 2 windows, cooking with electricity
URB8	Urban	Bedroom	Away from street, no smokers, natural ventilation 2 windows cooking with electricity
URB9	Urban	Living room	Light traffic, smokers, natural ventilation no windows, cooking with gas
URB10	Urban	Living room	Near the street, no smokers, natural ventilation 2 windows cooking with electricity
UCB11	Urban	Clothes shop	Heavy traffic, smokers, natural ventilation 1 windows, no cooking
UCB12	Urban	Market	Light traffic, no smoker, natural ventilation 1 windows, no cooking
UCB13	Urban	Restaurant	Moderate traffic, no smokers, natural ventilation 1 windows, cooking with gas
UCB14	Urban	Clothes shop	Heavy traffic, no smokers, natural ventilation no windows, no cooking
UCB15	Urban	Barbecued	Heavy traffic, smokers, natural ventilation no windows, cooking with coal
UCB16	Urban	Pizza store	Moderate traffic, no smokers, natural ventilation no windows, cooking with gas
UCB17	Urban	Market	Moderate traffic, no smoker, natural ventilation 1 windows, no cooking
UOB18	Urban	Laboratory	Away from street, no smokers, air conditioning, no cooking
UOB19	Urban	Office	Near the street, no smokers, natural ventilation 2 windows, no cooking
UOB20	Urban	Laboratory	Moderate traffic, no smokers, air conditioning, no cooking
UOB21	Urban	Office	Light traffic, smokers, natural ventilation no windows, no cooking
UOB22	Urban	Office	Heavy traffic, no smokers, air conditioning, no cooking
UOB23	Urban	School	Heavy traffic, smokers, natural ventilation no windows, no cooking
UOB24	Urban	Office	light traffic, no smokers, air conditioning, no cooking

2.2. Chemicals

Standards of 16 PAHs (concentration: 100–2000 μg/ml) were purchased from Sigma-Aldrich (USA) including naphthalene (NA), acenaphthene (ACE), acenaphthylene (ACY), phenanthrene (PH), fluorine (FL), anthracene (AN), pyrene (PY), fluoranthene (FLU), benzo[a]anthracene (BaA), chrysene (CH), benzo[b]fluoranthene (BbF), benzo[a]pyrene (BaP), benzo[k]fluoranthene (BkF), indeno[1,2,3-cd] pyrene (IP), and benzobenzo[g,h,i]perylene (BP), dibenzo[a,h]anthracene (DA). Dichloromethane, n-hexane, cyclohexane, acetone and other solvents were of spectral purity level (United States World Company).

2.3. Sample preparation, extraction and analysis procedure

Prior to the extraction, dusts were eliminated from the plastic bags using suitable forceps and placed into a mechanical sieve shaker equipped with a 100‑mesh sieve and were shaken for 7 min. After that, large debris such as shavings, chips, soil hunk, hair and etc. were eliminated by tweezers or a paint brush. The dust samples were then resieved for an additional 5 min with a 100‑mesh sieve to obtain dust particles < 150 µm. The sieved dusts were extracted through ultrasonic extraction procedure. The extract was passed through a 0.45 µm Whatman Teflon, concentrated to 250 mL using gentle stream of nitrogen (N-E VAP 112, USA) at 30 °C, and brought up to 2 mL in dichloromethane. Finally, the extracts were collected in suitable vials for the following measurements. The analysis was performed on an Agilent 7890 Gas Chromatograph-5975C Mass selective Detector (GC-MS) system equipped with a fused silica capillary Hp-5 column (30* 0.25 mm i.d., 0.25 mm film thickness).

Table 2. Factors used in health risk assessment

Factors	Unit	child	Adult
Exposure frequency (EF)	Day $year^{-1}$	180	180
Exposure duration (ED)	Year	6	24
Body weight (BW)	Kg	15	61.5
Dust ingestion rate (IR ingestion)	$mg.day^{-1}$	200	100
Inhalation rate (IR inhalation)	$m^3.day^{-1}$	10	20
Dermal adherence factor (AF)	$mg.cm^{-2}$	0.2	0.07
Dermal exposure area (SA)	cm^2	2800	5700
Particle emission factor (PEF)	$m^3.kg^{-1}$	$1.36*10^9$	$1.36*10^9$
Dermal adsorption fraction (ABS)	Unitless	0.13	0.13
Averaging life span (AT)	Day	70*365=25,550	70*365=25,550

2.4. Risk Assessment of PAHs in indoor Dust

The incremental life time cancer risk (ICLRs) was used to quantitatively assess the potential risk of PAHs exposure indoors according to the U.S. EPA standard models(EPA, 1991). Following mathematical equations were used to the ICLRs for indoor environment residents based on three main pathways including direct ingestion, inhalation and dermal contact.

$$\text{ICLRs Inhalation}=\frac{CS\times\left(CSF_{Inhalation}\times\left(\frac{BW}{70Kg}\right)^{\frac{1}{3}}\right)\times IR_{Inh}\times CF\times ED}{BW\times AT_C\times PEF}$$

$$\text{ICLRs Ingestion}=\frac{CS\times\left(CSF_{Ingestion}\times\left(\frac{BW}{70Kg}\right)^{\frac{1}{3}}\right)\times IR_{Ing}\times CF\times ED}{BW\times AT_C\times 10^6}$$

$$\text{ICLRs dermal}=\frac{CS\times\left(CSF_{Dermal}\times\left(\frac{BW}{70Kg}\right)^{\frac{1}{3}}\right)\times SA\times ABF\times AFd\times CF\times ED}{BW\times AT_C\times 10^6}$$

In these equations, CS stands for the Concentration of PAHs indoors (µg/g), which was calculated according to toxicity equivalent of BaP with the Toxic equivalency factor (TEF) listed in Table 2. CSF_i is carcinogenic slope factor $(mg.kg^{-1}.day^{-1})^{-1}$, which has been 7.3, 25, and 3.85 for ingestion, dermal and inhalation , respectively, based on the cancer-causing ability of BaP (Hu *et al.*, 2007). IR_{Inh} is the particle inhalation rate ($m^3.day^{-1}$), IR_{Ing} is the intake rate of dusts ($m^3.day^{-1}$), ABS is the dermal adsorption fraction (dimension less), AFd is the particle to dermal adherence factor ($mg.cm^{-2}.h^{-1}$), SA is the dermal surface area exposure (cm^2), and EF denotes the exposure frequency (day $year^{-1}$) indoors. Here, we have calculated non-working days and national holidays in a year and then deducted them from the total days of the year (365 days) to calculate exposure frequency for workers in official and commercial buildings. Further, EF for residents in residential buildings was calculated according to the Risk Assessment Guidance of U.S. EPA and related studies, as shown in table 2. ED is the exposure duration (year). Here, we have multiplied the average working years of study subjects by the number of working hours in a day to calculate exposure duration for workers in official and commercial buildings and ED for residents in residential buildings was calculated according to the Risk Assessment Guidance of U.S. EPA and related studies, as shown in table 2. Finally, PEF is the emission factor for particles ($m^3.kg^{-1}$), BW is the Body weight (kg), and AT is the average life span (day).

2.5. Data analysis

Statistical processing of data was done using SPSS ver. 20 (IBM Corp., USA). Before data analysis, the normality of all data was checked with the Shapiro-Wilk test. Descriptive statistics (mean, standard deviation and range) were employed for presentation of PAHs concentrations in indoor dusts.

3. Results and Discussion

3.1. Level of PAHs in indoor dust

The presence of 16 targeted PAHs was tracked in indoor dust samples of various areas and their individual concentrations (on dry weight basis) have been given in Table 3. The sum concentration of the 16 PAHs in dust of Bushehr spanned between 87.8 to14619 mgkg^{-1} d.w. with a mean of 2211.6 mgkg^{-1}d.w. Naphthalene was not detected in any of the samples. With a molecular mass of 128.17 and a vapor pressure of 8.5×10^{-2} mm Hg(Ambrose *et al.*, 1975). Acenaphthylene was generally detected only at low levels in dust samples and this can be related to its more affinity for volatilization. In contrast, as a result of less volatility, benzo[b]fluoranthene has been at in the highest levels in most of dust samples. In general, PAHs with a higher number of rings have lower volatilities (Maertens *et al.*, 2008).

As indicated in Table 3, considerable differences were observed between the levels of PAHs in dust across residential buildings, commercial buildings and official buildings. The levels (in mgkg-1 d.w.) of ΣPAHs ranged from 22.9 to 2883.9 in the dust samples of the residential buildings, from 71 to 13105 for commercial buildings and 26.4 to 3790 mgkg^{-1} d.w. The mean concentration of PAHs for the dust samples of the commercial buildings was higher than for residential buildings. Also it was higher for residential buildings than for official buildings.

Table 3. PAHs residues (mean of three analysis) in mgkg^{-1} d.w. in indoor dust of residential, commercial and official buildings of Bushehr

	Residential buildings										Commercial buildings			
PAHs	URB1	URB2	URB3	URB4	URB5	URB6	URB7	URB8	URB9	URB10	UCB11	UCB12	UCB13	UCB14
NA	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
ACE	1.2	13	0.14	62	12	17	1.1	0.8	30	1.3	21	2.3	921	19
ACY	0.9	78	0.7	54	154	23	3.2	1	23	1.8	54	4.8	1786	23
FL	1.4	121	1.8	38	211	41	6.7	0.4	14	1.2	84	4.3	413	32
PH	3.2	478	2.8	178	615	256	1.8	1.1	67	1.9	318	5.7	1760	167
AN	1.9	51	2.3	65	718	283	4.1	0.9	29	2.1	221	5.4	1434	16
FLU	4.3	378	3.2	145	526	311	6.2	2.1	78	3.4	248	6.7	1423	172
PY	1.9	311	0.9	171	498	234	2.3	1.8	87	2.7	193	3.2	1314	142
BaA	2.7	67	1.2	37	21	21	2.1	1.2	12	2.6	87	5.4	412	12
CH	2.9	76	1.9	93	102	32	1.9	2.1	65	3.2	156	4.3	645	78
BbF	4.1	404	2.1	159	170	19	24	2.2	44	1.8	158	4.5	1534	113
BaP	2.3	7.9	2.1	47	11	111	17	3.2	19	2.2	76	6.5	598	51
BkF	1.8	11	1.1	43	123	143	34	1.1	17	3.1	102	1.3	222	78
IP	1.9	112	1.2	188	211	267	37	2	89	4	67	5.4	67	45
BP	3	137	2.6	264	176	314	41	1.8	91	1.2	156	4.5	234	119
DA	2.3	141	1.5	192	152	271	33	1.2	67	3.2	113	6.7	342	109
ΣPAHs	36.7	2883.9	25.54	1735	3700	2349	215.4	22.9	732	35.7	2054	71	13105	1176

	Commercial buildings			Official buildings						
PAHs	UCB15	UCB16	UCB17	OCB18	OCB19	OCB20	OCB21	OCB22	OCB23	OCB24
NA	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
ACE	254	113	7.8	0.9	1.1	5.6	4.4	32	98	2.1
ACY	298	172	9.8	1.1	0.9	6.7	2.8	28	78	2
FL	387	198	9	0.8	0.3	7.4	3.2	19	89	1.7
PH	1190	412	2.5	1.8	1.3	3.2	0.9	76	314	1
AN	1421	319	1.1	1.9	1	1.8	1.1	33	287	0.7
FLU	1023	451	6.2	3.2	1.7	6.2	5.2	87	309	2.1
PY	978	333	2.3	1.2	1.5	4.3	4.8	67	289	2.2
BaA	78	67	2.1	3.9	1.3	3.2	2.1	19	41	1.2
CH	215	87	1.9	2.7	3.2	2.8	3.3	87	71	2.3
BbF	1212	413	24	3.2	3.4	27	17	43	328	8.9
BaP	879	231	17	4.9	2.9	16	11	26	198	5.6
BkF	718	312	37	1.1	1.9	43	23	21	214	11
IP	1652	545	34	4.5	1.3	47	38	87	425	18
BP	1978	676	33	4.3	2.3	54	42	128	562	12
DA	2345	509	41	3.9	3.2	44	37	87	487	17
ΣPAHs	14619	4838	238.7	39.8	26.4	272.2	194.8	840	3790	87.7

The highest levels of both ΣPAHs (14619 mgkg^{-1} d.w) was found at the UCB15 station. The features of this station include heavy traffic, existence of smokers, natural ventilation, no windows, and cooking with coal. The levels of 5- and 6-ring PAHs in indoor dust collected from buildings with coal combustion was higher than others. For buildings with coal for heating, cooking and etc., the PAH was the highest in dust samples, showing that coal combustion can significantly affect the PAHs generation (Qi *et al.*, 2014). Regarding the effect of smoking on dust, some studies show a higher concentration of PAHs in buildings with smokers than nonsmokers' buildings (Maertens *et al.*, 2004; Ren *et al.*, 2006). However, other studies demonstrate no correlation between smoking and non-smoking dust (Mahler *et al.*, 2010; Whitehead *et al.*, 2011). There is still uncertainty regarding the effect of smoking on the concentration of PAH in dust.

PAHs were classified based on their aromatic rings as follows: PAHs with 3-ring including Phe, Ant; 4-rings including FLT, PYR, BaA, CHR; 5-rings including BbF, BkF, BaP, BeP, DahA; 6-rings including BghiP and IND. The percentage of each group to total PAHs in dust samples during the sampling period has been shown in Fig. 1. As illustrated in this figure, high molecular weight PAHs with 4-6 ring have a higher fraction of about 70% in total PAHs, while low molecular weight PAHs with 2-3 ring participates a lower fraction. The higher fraction of high molecular weight PAHs in $PM_{2.5}$ samples is mainly due to fossil fuel consumption. Similar observations have also been reported by Liu and Cheng (Liu *et al.*, 2007) and D.P. Singh (Singh *et al.*, 2011).

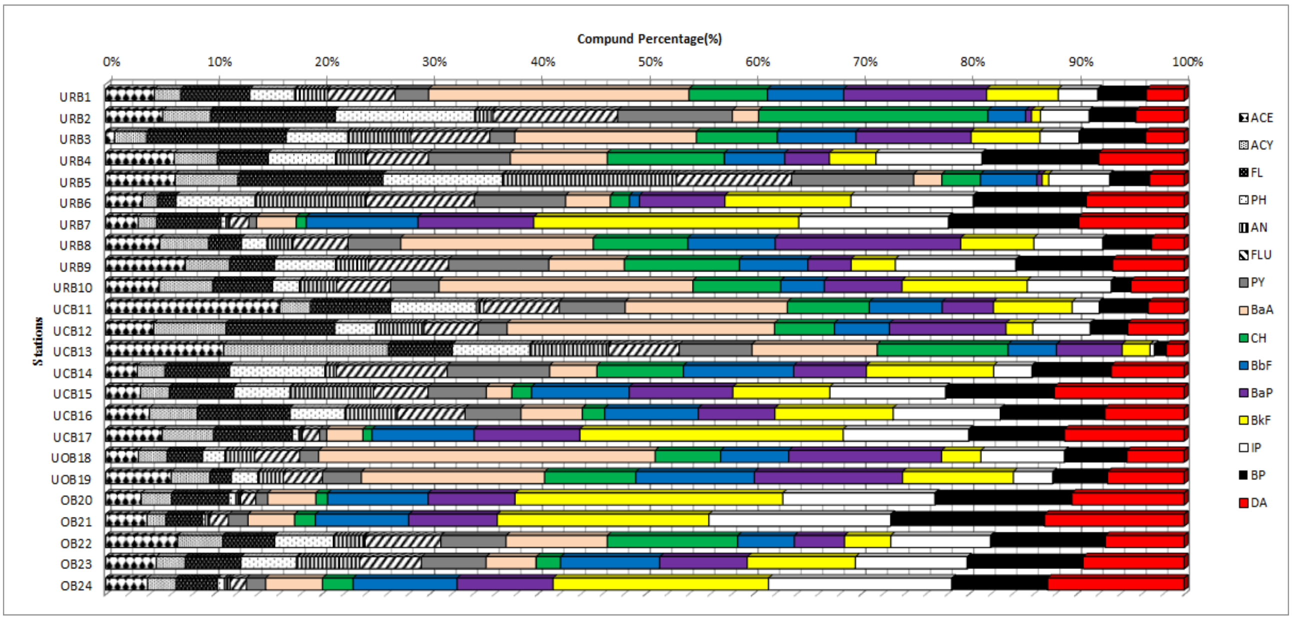

Figure 1. Percentage of PAHs compounds in dust samples from different stations

3.2. Health risk assessment

A probabilistic risk assessment was done based on TEF and CSF, with the findings shown in Table 4. As shown in the table, the cancer risk through dermal and ingestion route ranged from 10^{-6} to 10^{-5} across all the dust samples, while the average of cancer risk through inhalation pathway was 10^{-9} to 10^{-12}, about 10^{4} to 10^{6} times lower in comparison with the other two pathways (ingestion and dermal contact). These findings were consistent with those reported by other studies (Yang *et al.*, 2015). Thus, inhalation of PAHs-bound dust via mouth and nose was not considerable, when compared with dermal and ingestion routes. For children, the cancer risk values through dermal and ingestion pathways were about 10^{-6} to 10^{-5}, demonstrating that both routes (ingestion and dermal contact) contributed considerably to the cancer risk of children. On the other hand, for adults, the cancer risk values through dermal, ingestion and inhalation pathways were about 10^{-6} to 10^{-5}, 10^{-6} to 10^{-5} and 10^{-10} to 10^{-12}, respectively. Therefore, the risk value of cancer for infants was significantly higher ($p<0.01$) than the corresponding risk for adults. This can be related to the more hand-to mouth activity of infants, so contaminated dust can be easily ingested by children(Xia *et al.*, 2010). The other reason is the lower body weight of infants. So, the intake of PAHs compounds (mg per kg of body weight) in children is greater than in adults. Moreover, early development of body systems such as organs, immune and nervous systems can probably increase the sensitivity of children to carcinogens (Kamal *et al.*, 2014).

Table 4. The potential risk of PAHs in indoor dust of each building and three exposure routes

		Residential	**Commercial**	**Official**
CS (µg.kg^{-1})		0.9 ×10^{3}	2.37×10^{3}	8.9 ×10^{2}
Adults	Ingestion	7.87 ×10^{-5}	9.98 ×10^{-5}	1.14 ×10^{-6}
	Inhalation	2.33 ×10^{-10}	6.76 ×10^{-10}	5.43 ×10^{-11}
	Dermal contact	9.65 ×10^{-6}	4.65 ×10^{-5}	2.12 ×10^{-6}
	Cancer	8.83 ×10^{-5}	1.46 ×10^{-4}	3.26 ×10^{-6}
Child	Ingestion	8.76 ×10^{-5}	2.71 ×10^{-4}	8.76 ×10^{-5}
	Inhalation	4.43 ×10^{-10}	7.33 ×10^{-10}	4.43 ×10^{-10}
	Dermal contact	3.77 ×10^{-6}	8.45 ×10^{-5}	3.77 ×10^{-6}
	Cancer	9.13 ×10^{-5}	3.61×10^{-4}	9.13 ×10^{-5}

This research recommends better ventilation techniques, producing of foods in cooking oils with lesser PAHs in the kitchen and use of cooking fuels with lesser PAHs generations. Based on the study results, the study accentuates the urgent need of developing indoor air quality standards in Iran for home, office and occupational settings like kitchens in Bushehr city (Arfaeinia *et al.*, 2015).

Moreover, since the dominant pathway for polluted dust intake are oral ingestion and dermal contact, which together contribute 99% of carcinogenic risk and 90% of non-carcinogenic risk on average, utilize of a respirator or other protective equipments and reducing exposed skin area could greatly reduce the health risk.

4. Conclusion

A total of 24 indoor dust samples (10 residential indoor, 7 commercial and 7 official indoor buildings) collected from Bushehr, south of Iran, were analyzed for 16 PAHs. The total PAHs concentrations in dust ranged from 87.8 to14619 mgkg^{-1}d.w. with a mean of 2211.6 mgkg^{-1}d.w. The concentration of PAHs in indoor dust samples in the study area indicated that commercial buildings were the most polluted area, followed by residential and official buildings. The cancer risk levels due to exposure to indoor dust-bound PAHs in residential, commercial and official buildings of Bushehr province, Iran were 1.46 $\times10^{-4}$, 8.83 $\times10^{-5}$and 3.26 $\times10^{-6}$ for adults and 3.61$\times10^{-4}$, 9.13 $\times10^{-5}$ and 9.13 $\times10^{-5}$for children, respectively. The findings of this research would be useful to provide the information for enhancing the quality of living environment and human health in Bushehr province.

Acknowledgment

This work is supported by a project founded by Bushehr University of Medical Sciences (Grant No:2637).

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