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Original Research Paper

Research on Content, Distribution and Health Risk Assessment of PAHs in Surface Dust in Shenyang City

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ABSTRACT

As the "source" and "collector" of PAHs, surface dust in cities, cause invisible, potential and long-term destructions to ecosystems. PAHs content in the surface dust and distributions of functional areas in Shenyang city was discussed systematically. PAHs composition differences in different functional areas were analysed. It was concluded that parks and residential districts present the least PAHs pollution, while commercial districts and roads suffer the heaviest PAHs pollution. Different functional areas show similar composition of PAHs. Most PAHs are high-ring ones, accompanied with some low-ring PAHs. The content of high-ring PAHs is proportional to the number of rings or molecular weight. PAHs pollution and health risk in Shenyang city were evaluated through various methods. Results demonstrated that PAHs pollution in Shenyang city has reached a high level, which is threatening the local ecological environment and human health significantly.

INTRODUCTION

Surface dust is one of an important pollution sources in cities. It causes invisible, potential and long-term destructions to ecosystems. On one hand, surface dust can enter into human bodies through the respiratory tract and skin (Skeller & Lamprecht 1995), and be digested, absorbed and accumulated in human bodies, threatening human health. On the other hand, surface dust can be carried into water and soil by rain water, and finally enter into human bodies through the food chain (Behnisch et al. 2003). Due to urban transportation, industrial production and urban constructions, surface dust has accumulated abundant polycyclic aromatic hydrocarbons which intensify harms to the human health and environment. Polycyclic aromatic hydrocarbons (PAHs) is a kind of typical persistent organic pollutant, with characteristics of stable chemical structure, difficult degradation, high lipophicity, strong toxicity and easy accumulation in living bodies. It participates in metabolic activities of living organisms and human bodies, thus threatening human survival and reproduction directly. PAHs are good markers of some pollution sources, for example, coronene and benzopyrene are markers of auto-pollution sources, benzofluoranthrene/benzopyrene can be used to distinguish coals for civil use from those for vehicles (Paasivirta et al. 1999, Parkk & Kim 2005). Kong et al. (2010) studied PAHs pollution level in $PM_{2.5}$ and $PM_{2.5\sim10}$ in Shenyang, Anshan, Fushun, Dalian and Jinzhou. Their PCA results showed that PAHs mainly comes from fire coals and vehicle emissions. Zhang & Zhang (2008) analysed sources of PAHs in atmospheric deposition in Lanzhou City, finding heavy dust fall in Lanzhou City at the end of the heating period. Surface dust not only is an important pollutant and a key collector of PAHs, but also can migrate with water and air to cause secondary pollution. Therefore, it will threaten human health significantly.

In this paper, PAHs content in surface dust and distribution of functional areas in Shenyang city are discussed. PAHs composition differences in different functional areas were analysed and finally carried out an ecological risk assessment and health risk assessment.

MATERIALS AND METHODS

Sampling method: Samples were collected according to the following principles for the sake of high representativeness: (1) sampling sites distribute evenly in different functional areas of Shenyang city and could reflect the spatial distribution characteristics of the functional area; (2) sampling sites have satisfying representativeness for experimental analysis; (3) there are adequate sampling sites and the total samples are controlled within financial affordable range.

Samples were collected in Shenyang city in March, 2014. Shenyang city was divided into five functional areas, namely, parks, institutes and colleges districts, residential districts, commercial districts and roads. To avoid sampling error, we collected samples from three parallel sampling sites in each functional area. Equal amount of these three samples were mixed evenly as the sample of a functional area. All samples were stored in freshness protection packages independently.

Test method: 1.00g soil samples were put into a 100mL bottle with blue cover. Later, 20mL acetone, 15mL dichloromethane and 10mL NaCl solution (1.5gNaCl+10mL water) was added into a bottle to extract PAHs in soil. The bottle was vibrated in dark for 16h (30°C, 150r/min). 10g upper organic phase was taken from the bottle and put into a small beaker for air drying. The dried upper organic phase was dissolved into a constant volume by 2mL acetonitrile and then filtered by a piece of 0.22μm organic filter membrane. The filtrate was stored for the following test.

Soil samples were tested by fluorescence detector and ultraviolet detector. Chromatographic conditions were: (1) column temperature: 35°C. (2) Mobile phase: acetonitrile and water were used as mobile phase. First, keep 60% acetonitrile + 40% water for 35min. Secondly, increase acetonitrile from 60% to 100% at a linear velocity and keep it for 10min, and finally, decrease acetonitrile from 100% to 60% at a linear velocity and keep it for 5min. (3) flow rate of mobile phase: 0.5 mL/min. (4) Sample size: 10μL. PAHs were tested under their emission/excitation wavelengths.

RESULTS AND DISCUSSION

PAHs content differences in different functional areas: Accumulation of PAHs in surface dust in cities is significantly influenced by spatial position. Therefore, Shenyang city was divided into five functional areas in this paper: parks, institutes and colleges districts, residential districts, commercial districts and roads. Total PAHs in surface dust in parks, institutes and colleges districts, residential districts, commercial districts and roads were 1,906.207-9,187.359 μg/kg (4,982.66 μg/kg in average), 3,073.964-16,397.45 μg/kg (8,992.12μg/kg in average), 3,499.857-1,108.12 μg/kg (7,702.46μg/kg in average), 5,511.242-12,879.59 μg/kg (8,389.43μg/kg in average) and 4,881.314-17,109.35 μg/kg (11,149.54μg/kg in average) respectively. 16 PAHs regulated by U.S.EPA were detected in all the parks. Zhongshan Park shows the highest total PAHs, followed by Beiling Park, Nanhu Park, Xinghua Park, Bainiao Park and Youth Park successively. PAHs in main streets contain less dibenzanthracene. Roads surrounding the thermal power plant generally have high total PAHs. Total PAHs in institutes and colleges districts is higher than the expectation, especially in surface dust collected from Shenyang University and Northeastern University. Commercial districts present lower total PAHs than the expectation. They have low average level, but high maximum level. Residential districts have generally lower total PAHs. The total PAHs in Shenyang is significantly higher than those in Isfahan metropolis, Iran (Naghmeh Soltani et al. 2015) and Shanghai (Wang et al. 2013). Shenyang shows similar PHAs content with Beijing, but has little higher PHAs content in some functional areas (Zhong et al. 2013).

Contents of different PAHs vary greatly. Except for PAHs collected from some sampling sites, most PAHs samples contain a little small-molecular-weight PAHs. Meanwhile, parks in different administrative districts have different sources of PAHs. This can be proved by most cities. Most of the existing researches concluded higher PAHs content in roads surrounding industrial parks and shopping malls, but lower PAHs content in parks and residential districts (Wang et al. 2012, Du 2014). PAHs content in surface dust varies significantly in different functional areas. Parks have the lowest PAHs content, followed by residential districts. Parks are places of entertainment and have high greening rate, and trees, flowers and grasses can absorb the dust in air. Residential districts are living places of human beings and have less pollution sources. On the contrary, commercial districts suffer the heaviest PAHs pollution compared to rest of the functional areas. Crowded population, heavy traffic, full of high-rise buildings, but low greening rate in commercial districts cause poor airflow and high accumulation of PAHs.

Evaluation of pollution status: Single-factor index (SI) is often used to evaluate pollution level of pollutants. It can present impact of single index effectively. Hankanson (1980) used SI to evaluate degree of contamination of settlings (Zhang 2008, Peng 2011, Cheng 2006). The formula is:

$$C^i_f = C^i / C^i_n \quad ...(1)$$

Where, C^i_f is the contamination factor of component i in total PAHs, C^i is actual concentration of i and C^i_n is the reference value of i.

Standard of Agricultural Soil Remediation (Canada) limits the highest content of Nap, Phe, Py, BaA, BaP, BbF, BkF, DahA and InP in agricultural soil at 0.1μg/g. Hankanson (1980) used C^i_f to represent degree of contamination of a single pollutant. He disclosed the correlation between the numerical range of C^i_f and degree of contamination, which has been widely applied. According to this correlation, $C^i_f <$ 1 means low pollution; $1 < C^i_f \leq 3$ means moderate pollution; $3 \leq C^i_f < 6$ means high pollution; $3 \leq C^i_f < 6$ and means extremely high pollution.

The evaluation revealed that Shenyang city has high contamination factor of PAHs in surface dust. The contamination factors of single PAHs in parks are listed in Table 1. It can be seen from Table 1 that all PAHs except for DahA (0-0.94) pertain to low pollution. Moreover, all pollutants have 1-4 extremely high pollution areas except for BkF which is identified as moderate pollution (1.03-2.48, 1.92 in average).

Health risk assessment of PAHs in surface dust: Health risk assessment mainly covers the estimation of pollutants entered into human bodies and evaluation of relationship between dosage and negative health effect. It is composed of pollutant identification, determination of exposure pathways, calculation of exposure dosage and exposure risk assessment (Health 1994, Khillare et al. 2012, Christopher et al. 2014). According to the migration and the transformation law of surface dust in an urban landmark environment, it is believed that the dust enters into human body mainly through hands, mouth, respiratory system and skin contact (Li & Hu 2010, Yin et al. 2006, Chang et al. 2009, Yan et al. 2012). Considering different weights and dust intake rates of different ages, this paper estimated the daily oral intake of PAHs of five age groups, from the dust in Shenyang city. These five age groups are infants (<1), children (1-3), juveniles (4-10), youths (11-18) and adults (≥19). Since skin absorbs very little PAHs, only oral intake and breathing intake of PAHs were estimated when discussing exposure pathways of human beings to PAHs in surface dust.

Oral intake: In this paper, daily oral intake of PAHs from surface dust in Shenyang was calculated from the following formula:

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

Where, C_{dust} is the mean content of different PAHs in dust (μg/g); f_1 is the dust intake rate (g/day), which is 0.02g/day for infants, 0.1g/day for children and 0.05g/day for juveniles, youths and adults; M_1 is the evaluated weight of different age groups (kg), with values 5kg, 19kg, 29kg, 53kg and 63kg for infants, children, juveniles, youths and adults in Shenyang. BaP has strong carcinogenicity and becomes the primary control component of PAHs. Except for BaP, high-ring PAHs like BaA, BbF, BkF, Ind and DahA also have carcinogenic potential. To reflect toxicity of PAHs comprehensively, this paper evaluated the health risk of PAHs through equivalent mass concentration of benzopyrene (BaPE) suggested by Yassaa.

BaPE = 0.06BaA + 0.07B [b+k] F + BaP + 0.6 DahA + 0.08Ind ...(3)

EDI results of oral intake of PAHs in some functional areas in Shenyang and other regions are given in Table 2.

Table 1: Contamination factors of single PAHs in parks.

Number	Bainiao Park	Beiling Park	Nanhu Park	Zhongshan Park	Xinghua Park	Qingnian Park
Nap	2.52	1.21	3.73	4.20	7.05	0.00
Phen	3.59	12.05	1.15	14.65	0.70	0.00
Pry	4.27	8.62	0.00	11.71	3.54	4.42
BaA	2.09	6.01	8.91	12.24	1.94	2.87
BkF	1.99	2.32	1.69	1.03	2.38	1.56
BaP	1.36	3.62	8.09	5.51	1.70	2.46
DahA	0.66	0.00	0.00	0.00	0.00	0.00

The total EDI range of 16 PAHs in surface dust in parks for different age groups is 0.42-2.82. Children (1-3 yr) show the highest daily intake of PAHs, while adults (≥19 yr) show the lowest daily intake of PAHs. The total EDI range of 16 PAHs in surface dust in commercial districts for different age groups is 0.77-5.08. Children (1-3 yr) show the highest daily intake of PAHs, while adults (≥19 yr) show the lowest daily intake of PAHs. Compared to parks, commercial districts have higher oral exposure risk to PAHs in surface dust viewed from both total PAHs and composition. Oral intake of BaPE in Shenyang is higher than that in Hong Kong and Berlin, indicating the higher oral exposure risk to surface dust in Shenyang. Influence of surface dust to human health deserves high attention.

Breathing intake: In this paper, daily breathing intake of PAHs from surface dust in Shenyang was calculated from the following formula (Guo & Kanan 2001):

$$EDI = \frac{C_{air} f_2}{M_1} \quad ...(4)$$

Where, C_{air} is the mean content of different PAHs in air (μg/g); f_2 is the breathing intake rate of dust (m^3/day), which is 4.5, 7.6, 10.9, 14.0 and 13.3m^3/day for infants, children, juveniles, youths and adults; M_1 is the evaluated weight of different age groups (kg), with values 5kg, 19kg, 29kg, 53kg and 63kg for infants, children, juveniles, youths and adults respectively in Shenyang.

EDI results of breathing intake of PAHs from surface dust in Shenyang are depicted in Table 3. The total EDI range of 16 PAHs in surface dust in parks for different age groups is 1.19-5.07. Infants show the highest breathing intake, followed by children, juveniles, youths and adults successively. The total EDI range of 16 PAHs in surface dust in commercial districts for different age groups is 2.74-11.68. Similarly, infants show the highest breathing intake, followed by children, juveniles, youths and adults successively. The results were consistent with Isfahan metropolis, Iran (Naghmeh et al. 2015). This finding was similar to the human cancer risk resulted from PAHs exposure in urban soils of Beijing, China (Peng et al. 2011), and urban surface

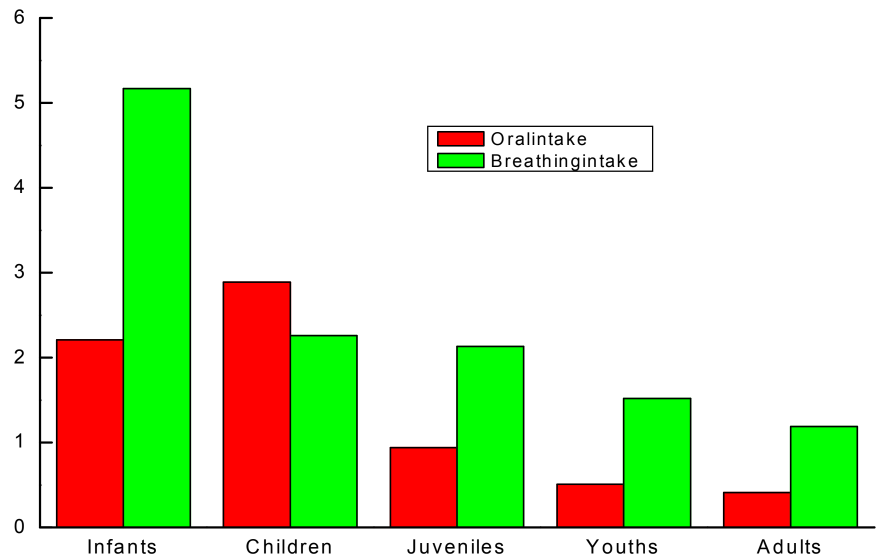

Fig. 1: Daily oral intake and breathing intake of surface dust in parks in Shenyang (EDI).

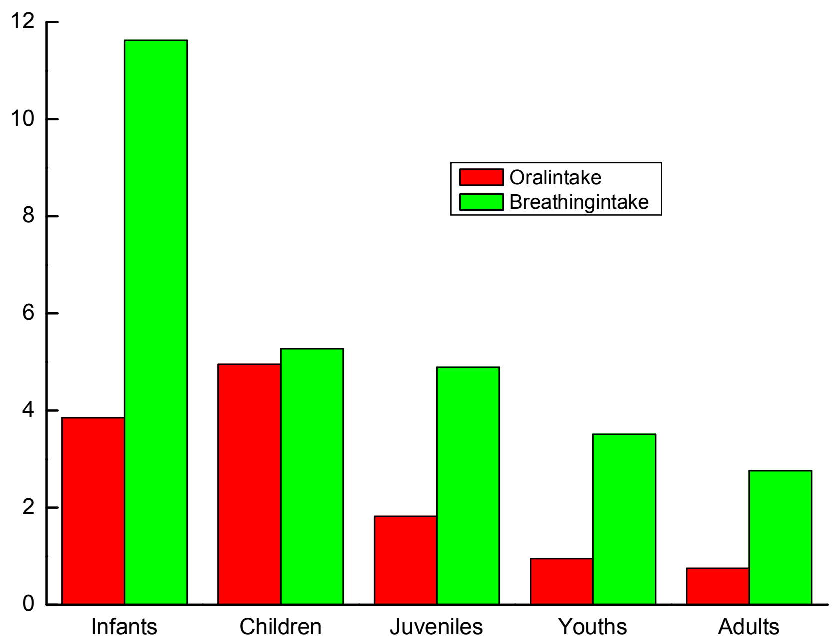

Fig. 2: Daily oral intake and breathing intake of surface dust in Commercial Districts in Shenyang (EDI).

dust of Guangzhou, China (Wang et al. 2011). Compared to parks, commercial districts present higher EDI of 16 PAHs and EDI of single PAHs. This indicates that human beings suffer higher exposure risks to PAHs in commercial districts. Dust in commercial districts threatens human health more seriously than those in other functional areas.

Based on the comparison between breathing intake and oral intake (Figs.1 and 2), all age groups (except for children) present higher EDI of breathing intake than EDI of oral intake in parks. This reflects that PAHs in surface dust enter into human bodies mainly through respiratory system in Shenyang.

CONCLUSIONS

1. In different functional areas in Shenyang, parks show the minimum total PAHs in surface dust followed by

Table 2: EDI results of oral PAHs intake in some functional areas in Shenyang and other regions.

	Infants	Children	Juveniles	Youth	Adults
Parks	2.14	2.82	0.92	0.50	0.42
Commercial Districts	3.86	5.08	1.66	0.91	0.77
Hong Kong (Chung et al. 2007)	2.18	2.86	0.94	0.51	0.43
South Korea (Nam et al. 2003)	1.63	2.14	0.70	0.38	0.32
Bangkok (Wilcke et al. 1999)	66.81	87.91	28.80	15.76	13.26
Tarragona (Nadal et al. 2004)	NA	NA	NA	NA	0.32

Table 3 EDI results of breathing intake of PAHs from surface dust in Shenyang

	Infants（<1）		Children（1～3）		Juveniles（4～10）		Youths（11～18）		Adults（≥19）	
	Parks	Commerce	Parks	Commerce	Parks	Commerce	Parks	Commerce	Parks	Commerce
acy	0.02	0.07	0.01	0.03	0.01	0.03	0.00	0.02	0.00	0.02
ant	0.03	0.05	0.01	0.02	0.01	0.02	0.01	0.01	0.01	0.01
baa	0.52	1.34	0.23	0.60	0.22	0.56	0.15	0.39	0.12	0.31
bap	0.39	0.54	0.17	0.24	0.16	0.23	0.11	0.16	0.09	0.13
bbf	0.42	0.56	0.19	0.25	0.18	0.23	0.12	0.16	0.10	0.13
bghip	0.24	0.54	0.10	0.24	0.10	0.22	0.07	0.16	0.06	0.13
bkf	0.17	0.28	0.08	0.12	0.07	0.12	0.05	0.08	0.04	0.07
chry	0.48	1.03	0.21	0.46	0.20	0.43	0.14	0.30	0.11	0.24
daha	0.02	0.24	0.01	0.11	0.01	0.10	0.01	0.07	0.00	0.06
flt	0.65	2.61	0.29	1.16	0.27	1.09	0.19	0.77	0.15	0.61
flu+ace	0.34	0.54	0.15	0.24	0.14	0.23	0.10	0.16	0.08	0.13
icdp	0.13	0.58	0.06	0.26	0.06	0.24	0.04	0.17	0.03	0.14
nap	0.41	0.46	0.18	0.20	0.17	0.19	0.12	0.13	0.10	0.11
phen	0.67	1.71	0.30	0.76	0.28	0.71	0.20	0.50	0.16	0.40
pry	0.58	1.14	0.26	0.51	0.24	0.48	0.17	0.33	0.14	0.27
$\sum PAHs$	5.07	11.68	2.25	5.19	2.12	4.88	1.49	3.43	1.19	2.74

residential districts, commercial districts, institutes and colleges districts, and roads successively. This reflects that the total PAHs content in surface dust in different functional areas varies from each other.

2. Different functional areas show similar composition of PAHs. Most PAHs are high-ring ones, accompanied with some low-ring PAHs. The content of high-ring PAHs is proportional to number of rings or molecular weight. In low-ring PAHs, naphthalene and anthracene have higher contents. In high-ring PAHs, benzopyrene has higher content.
3. The SI results show that PAHs in surface dust in Shenyang has high contamination factor.
4. According to the health risk assessment, human beings have higher oral exposure risk to PAHs in commercial districts in Shenyang. EDI of breathing intake is higher than EDI of oral intake, indicating that PAHs in surface dust enters into human bodies mainly through the respiratory system in Shenyang.

The results are all calculated under the hypothesis of uniform environmental conditions. However, since commercial districts and main streets are cleaned more frequently, surface dust samples collected from these areas may be fresher than others. This will influence experimental results to a certain extent. However, the maximum and minimum PAHs contents can further disclose pollution law of PAHs.

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