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Polycyclic Aromatic Hydrocarbons in Urban Soils of Zhengzhou City, China: Occurrence, Source and Human Health Evaluation

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Abstract

Increasing contamination of urban soil by persistent organic pollutants is a major environmental issue. The purpose of the present study was to investigate the distribution, source and human health risk of polycyclic aromatic hydrocarbons (PAHs) in different functional areas in Zhengzhou City, China. Total 130 soil samples were collected from surface layer (0–10 cm) in urban road, overpass, residential area and park in the city during January 2019. Concentrations of $\sum PAH_{16}$ in the urban soil ranged from 49.90 to 11,565 μg kg^{-1} and seven carcinogenic PAHs accounted for 69% of the total PAHs. The mean concentrations of PAHs decreased in the following order: urban road > overpass > residential area > park. Analysis based on diagnostic rate demonstrated that PAHs mainly originated from pyrolysis sources including traffic emissions and combustion of coal and biomass. Health risk assessment indicated that PAHs in urban road in the city have potential carcinogenic risks to residents. The present study suggested that the control of urban PAHs pollution in Zhengzhou City should be strengthened.

Keywords Polycyclic aromatic hydrocarbons · Urban soil · Human health assessment

Cities around the world provide an environment for more than half of the earth's population (UN DESA 2010). Urbanization has developed rapidly as people migrate from rural area to urban area in the past few decades. Especially for China, the urbanization rate increased from 17.92% to 59.58% from 1978 to 2018 (China State Statistical Bureau 2019). The development of urbanization is accompanied by industrialization, with the result that energy depletion and contamination emissions are greatly concentrated in urban region. It is inevitable that anthropogenic activities result in continuous and increased emissions of pollutants such as polycyclic aromatic hydrocarbons (PAHs) in urban ecosystems. These persistent and poor mobility pollutants entered urban environmental systems and seriously threatened human health (Man et al. 2013). Therefore, investigating the distribution of PAHs and their negative effects is a critical issue in assessing urban public health.

PAHs are toxic chemicals that can cause chronic diseases with mutagenic and carcinogenic properties (Gope et al. 2018). They are ubiquitous pollutants present in urban air, soil and water and are generally derived from traffic, industrial emissions and coal combustion, etc. Although PAHs were initially emitted into the air in gaseous form, they can eventually be deposited on soil through wet and dry sedimentation (Desalme et al. 2013). Soil is the primary sink of PAHs because the pollutants could adhere to organic matter and minerals and remain persistent in soil for a long time. PAHs generally poison the growth of plants, cause rivers to be contaminated, and eventually threaten human health via ingestion, dermal and inhalation into the body (Keshavarzi et al. 2015).

Usually, anthropogenic factors exhibit the greatest effect on distribution of PAHs in the environment (Jiang et al. 2009). Urban regions are densely populated and have huge energy needs, which resulted in the increase of industrial

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waste, vehicular emission and coal combustion and generated many PAHs pollutants. Generally, concentrations of PAHs in urban regions are much higher than in rural regions. For example, PAHs concentrations in industrial area and urban road area in Delhi, India were 7.4 and 4.5 times of those in agricultural areas, respectively (Singh et al. 2012). For China, the average concentration of PAHs in urban soil was 2802 µg kg^{-1}, which was significantly higher than that in agricultural soil (158 µg kg^{-1}) (Sun et al. 2017; Yu et al. 2019). Therefore, the situation of PAHs in urban soils should be paid more attention.

Zhengzhou is the capital city of Henan Province, China with a population of 10.13 million (China State Statistical Bureau 2019). Zhengzhou City has one of the largest transportation hubs in central China and is an intricate industrial base dominated by electricity, automobiles, and railways. Simultaneously, the urbanization rate in the city increased from 32.4% in 1978 to 73.4% in 2018 (Zhengzhou Statistical Bureau 2019). The local energy structure is dominated by coal combustion, which consumed 30 million tons of coal annually and resulted in large amounts of PAHs emitting into the environment. The concentrations, distribution, and sources of PAHs in the atmospheric particulate matter and water have been explored in Zhengzhou City (Li et al. 2019; Fu et al. 2011). However, limited studies have been investigated the status and potential hazards of PAHs in urban soil in the city. The main purposes of the present study were: (1) to explore the level and spatial distribution of PAHs in the urban soils of Zhengzhou City; (2) to identify possible source of PAHs; and (3) to assess human health risk of PAHs in the urban soils.

Materials and Methods

During January 2019, soil samples were collected from the main district of Zhengzhou City (112° 42′–114° 14′ E, 34° 16′–34° 58′ N), including urban road, overpass, residential area and park in Zhengzhou City as shown in Fig. S1. The position of each sampling site was recorded by GPS. Four surface soil subsamples (0–10 cm) from each site (10 m × 10 m) were collected and mixed as one sample. The soil samples were placed in polyethylene sealing bag, transferred to the laboratory immediately, air-dried and sieved through a 2-mm mesh to remove stones and coarse materials and stored at − 80 °C for analysis of total PAHs. Partial soil samples were passed through a 100 mesh sieve for analysis soil organic matter (SOM).

SOM was measured by the potassium dichromate volumetric method (Lao 1988). For determination of PAHs concentrations in soil, soil samples (5.0 g) were Soxhlet extracted with mixture of acetone and dichloromethane (120 mL, 1:3 v/v) (USEPA 1996; Jia et al. 2018). Methods for sample purification and detection were described in our previous study (Tian et al. 2018). The strict quality control and quality assurance including the procedural blank (all reagents without sample addition), duplicate analysis and certified standard reference materials were used to check the accuracy of PAHs analysis. The spiked recovery rate was 88% (Pyr) to 126% (Nap) for PAHs. The coefficient of variation of concentration between samples duplicate was less than 10%.

The health risk model used in this study is taken from the USEPA model (USEPA 2005). In general, there are three major pathways which humans are exposed to PAHs in the soil, including ingestion, dermal and inhalation. Therefore, according to different exposure pathways, three health risk calculation equations are used in this study (Qi et al. 2019). Due to physiological differences between different genders and ages, the risks were calculated separately for male and female of different age groups.

The toxicity equivalency factor (TEF) is used to quantify the relative toxicity of a chemical compound and compared it to other reference chemicals. For PAHs, the reference chemical benzo(a)pyrene (BaP) is widely accepted (Yang et al. 2017). Thus, BaP is used as a reference compound to estimate the BaP-equivalent concentration (BaP_{eq}) of other PAHs. The toxic equivalent quotient (TEQ) of each sample was calculated as follows:

$$\mathrm{TEQ}_{\mathrm{BaP}} = \mathrm{C_i} \times \mathrm{TEF_i}.$$

$$\sum \mathrm{BaP}_{\mathrm{eq}} = \sum \mathrm{TEQ}_{\mathrm{BaP}}$$

where C_i is the concentrations of PAH i (µg kg^{-1}); TEF_i is the corresponding toxic equivalency factor. The toxicity equivalent coefficients of each PAH are listed in Table S1. The incremental life time cancer risks (ILCR) exposed to PAHs contamination in soils were calculated by relevant parameters. The ILCR equations for ingestion, dermal, and inhalation for urban soils are as following equations:

$$\mathrm{ILCR}_{Ingestion} = \frac{\mathrm{C}_{PAHs} \times \mathrm{CSF}_{Ingestion} \times \sqrt[3]{\left(\frac{\mathrm{BW}}{70}\right)} \times \mathrm{IngR}_{\mathrm{Ingestion}} \times \mathrm{EF} \times \mathrm{ED} \times \mathrm{CF}}{\mathrm{BW} \times AT} \tag{1}$$

$$\mathrm{ILCR}_{Dermal} = \frac{\mathrm{C}_{PAHs} \times \mathrm{CSF}_{Dermal} \times \sqrt[3]{\left(\frac{\mathrm{BW}}{70}\right)} \times \mathrm{SA} \times \mathrm{AF}_{\mathrm{soil}} \times \mathrm{ABS} \times \mathrm{EF} \times \mathrm{ED} \times \mathrm{CF}}{\mathrm{BW} \times AT} \tag{2}$$

$$\mathrm{ILCR}_{Inhalation} = \frac{\mathrm{C}_{PAHs} \times \mathrm{CSF}_{Inhalation} \times \sqrt[3]{\left(\frac{\mathrm{BW}}{70}\right)} \times \mathrm{IngR}_{Inhalation} \times \mathrm{EF} \times \mathrm{ED}}{\mathrm{BW} \times AT \times \mathrm{PEF}} \tag{3}$$

$$\mathrm{ILCR}_{Total} = \mathrm{ILCR}_{Ingestion} + \mathrm{ILCR}_{Dermal} + \mathrm{ILCR}_{Inhalation} \tag{4}$$

For these equations, where C_{PAHs} is the sum of toxic equivalent concentrations of 16 US EPA priority PAHs ($\sum\mathrm{PAH}_{16}$) in soil (mg kg^{-1}); CSF is the carcinogenic slope factor (mg/kg/day); BW is body weight(kg) ; $\mathrm{IngR}_{Ingestion}$ is soil ingestion rate in one day (mg/day); $\mathrm{IngR}_{Inhalation}$ is soil inhalation rate in one day (m^3/day); EF is exposure frequency (days/year); ED is exposure duration (years); CF is the conversion factor of concentrations (kg/mg); AT is averaging life time (days); SA is body exposed dermal area (cm^2); $\mathrm{AF}_{\mathrm{soil}}$ is dermal adherence factor (mg/cm^2); ABS is the dermal absorption faction; PEF is the particle emission factor (m^3/kg). All the parameters used are showed in Table S2.

Statistical analyses, including correlation analysis and one-way ANOVA were implemented in SPSS 25.0 Statistics. In addition, graphical plots of data were produced using Origin 8.0. Spatial distribution map for PAHs concentration was carried out with ArcGIS 10.2.

Results and Discussion

The statistical evaluation (min, max and mean) of $\sum\mathrm{PAH}_{16}$ concentrations in the soil samples collected from four different functional zones of the Zhengzhou City are present in Table 1. Total concentrations of $\sum\mathrm{PAH}_{16}$ ranged from 49.90 to 11,565 μg kg^{-1}, with mean of 1567 μg kg^{-1}. The concentrations of seven carcinogenic PAHs ($\sum\mathrm{PAH}_7$), including BaA, CHr, BbF, BaP, IcdP, and DahA were in the range of 13.80 to 5773 μg kg^{-1}, accounting for 45.74% of $\sum\mathrm{PAH}_{16}$. The sequence of total concentrations of $\sum\mathrm{PAH}_{16}$ in the four urban functional zones was as follows: urban road (1879 μg kg^{-1}) > overpass (817.6 μg kg^{-1}) > residential area (596.5 μg kg^{-1}) > park (448.7 μg kg^{-1}). PAHs concentrations in the soil from the overpass were lower than those from urban road. This may be due to that the overpass is located above the ground and PAHs caused by vehicle emissions are mainly accumulated in road dust. In addition, overpasses were mostly built in Zhengzhou in recent years and trees and grasses were planted under the overpasses to absorb dust and pollutants, resulting in lower PAHs concentrations. $\sum\mathrm{PAH}_{16}$ concentrations in urban soils in this study were higher than those in urban soils of Nanjing (980 μg kg^{-1}) of China (Yang et al. 2017) and Torino (857 μg kg^{-1}) of Italy (Morillo et al. 2007). As an inland city, Zhengzhou is mainly based on the development of transportation and industry. It has urban automobile traffic congestion and has 3.48 million vehicles in 2018 (Zhengzhou Statistical Bureau 2019). In addition, both domestic heating in winter and coal burning in industrial area also are important sources of PAHs in Zhengzhou and the city's energy consumption was up to 15,542,400 tons of standard coal in 2018 (Henan province Bureau of Statistics 2019). In addition, the degradation rate of PAHs decreases with the decrease of soil temperature (Wang et al. 2017). Annual average temperature in Zhengzhou is apparently lower than Nanjing of China and the lower degradation degree of PAHs leads to higher level of PAHs in urban soil of Zhengzhou. The concentrations of $\sum\mathrm{PAH}_{16}$ found in this study were higher than the average concentrations of $\sum\mathrm{PAH}_{16}$ in urban soils in China (1,083 μg kg^{-1}) and the background value of global soil PAHs (328 μg kg^{-1}) (Zhang et al. 2019; Nam et al. 2008), suggesting that PAHs concentrations in urban soils of Zhengzhou are serious.

Spatial distribution of PAHs in the urban soils of Zhengzhou City are shown in Fig. 1. The spatial distribution patterns of PAHs can help understand their current distribution in urban areas and identify sources of the pollutants. The highest concentration of $\sum\mathrm{PAH}_{16}$ was recorded in the central region of the city, while the lower were recorded in the east and north of the city. Heavy traffic is a major cause of high levels of contamination in urban areas (Glaser et al. 2005). The transportation junctions including Zhengzhou railway station, coach central station and Erqi square and commercial district are located in the central areas of the city. Due to the large population and heavy traffic in these areas, a large number of PAHs are generated from vehicle traffic and commercial activities. In contrast, the northeast part of Zhengzhou is mostly residential area, which is far away from transportation and commercial centers. There are fewer urban transportation tasks and fewer pollutants in the northeast of Zhengzhou City.

The various diagnostic ratios can be used to identify sources of PAHs in the environment, including Ant/(Ant + Phe), Fla/(Fla + Pyr), BaA/(BaA + Chr), and IcdP/(IcdP + BghiP). The ratio of Ant/(Ant + Phe) < 0.1 indexes a petroleum source, while > 0.1 indicates sources from combustion. The ratio of Fla/(Fla + Pyr) < 0.4 likely implies a petroleum source, while 0.4–0.5 indexes petroleum combustion and > 0.5 indexes biomass and coal combustion (Dvorská et al. 2011). The ratio of BaA/(BaA + Chr) < 0.2

Table 1 Concentration of PAHs (μg kg^{-1}) in soils of different functional areas in Zhengzhou City of China

Compounds	Urban road (n = 98)		Overpass (n = 10)		Residential area (n = 10)		Park (n = 12)		Total (n = 130)	
	Mean	Range	Mean	Range	Mean	Range	Mean	Range	Mean	Range
Nap	93.51 ± 82.14	ND–369.1	46.35 ± 67.90	ND–216.6	69.26 ± 102.9	ND–296.3	66.60 ± 77.12	ND–275.5	85.54 ± 82.83	ND–369.1
Acy	25.12 ± 23.90	ND–157.6	9.40 ± 10.47	ND–30.73	6.48 ± 10.80	ND–34.54	10.89 ± 10.54	ND–27.41	21.16 ± 22.45	ND–157.6
Ace	8.40 ± 11.28	ND–50.38	7.28 ± 13.52	ND–44.85	1.40 ± 1.87	ND–4.20	2.31 ± 2.20	ND–7.89	7.21 ± 10.72	ND–50.38
Flu	28.95 ± 25.21	ND–147.0	21.82 ± 21.14	ND–58.95	10.39 ± 8.52	ND–21.87	14.48 ± 12.98	ND–38.94	25.64 ± 23.82	ND–147.0
Phe	188.3 ± 185.2	1.68–1202	86.46 ± 64.10	18.65–199.1	72.80 ± 65.83	1.49–196.7	52.42 ± 41.40	1.36–136.0	159.1 ± 171.0	1.36–1202
Ant	33.75 ± 33.45	ND–228.1	10.26 ± 5.76	4.77–19.88	7.82 ± 7.79	0.19–20.27	6.41 ± 5.09	0.66–16.20	27.42 ± 31.21	ND–228.1
Fla	233.1 ± 301.2	9.60–1827	104.4 ± 92.38	17.68–313.5	59.71 ± 72.80	ND–241.1	43.25 ± 26.41	8.10–86.21	192.3 ± 273.0	ND–1826
Pyr	190.3 ± 258.3	2.62–1469	81.37 ± 60.68	16.13–175.7	47.78 ± 64.06	3.68–214.0	38.9 ± 23.36	6.20–76.46	157.0 ± 232.9	2.62–1469
BaA	108.5 ± 136.9	3.15–869.5	43.81 ± 32.70	15.15–113.2	26.01 ± 31.96	2.54–104.8	19.72 ± 11.04	3.26–37.64	89.01 ± 124.3	2.54–869.5
Chr	228.8 ± 244.7	10.66–1292	90.11 ± 70.80	9.61–197.2	62.05 ± 72.63	ND–220.9	42.36 ± 28.98	7.42–92.74	188.1 ± 225.9	ND–1292
BbF	188.9 ± 209.1	3.83–1338	84.61 ± 71.54	6.89–211.6	57.80 ± 65.74	1.28–183.8	33.36 ± 21.55	4.42–73.69	156.4 ± 192.2	1.28–1338
BkF	66.60 ± 84.17	ND–481.2	27.47 ± 24.08	ND–72.96	19.68 ± 23.60	1.24–71.40	17.07 ± 8.47	1.72–24.91	55.04 ± 76.38	ND–481.2
BaP	107.0 ± 144.0	2.86–923.9	40.18 ± 38.03	0.29–120.5	23.91 ± 30.45	0.77–87.37	18.30 ± 12.66	2.35–37.58	87.28 ± 130.4	0.29–923.9
DahA	62.26 ± 90.78	ND–504.0	20.23 ± 19.48	ND–64.76	25.46 ± 31.25	ND–93.98	16.08 ± 12.70	ND–35.61	51.93 ± 81.48	ND–504.0
BghiP	216.4 ± 257.4	8.30–1739	94.70 ± 71.10	8.72–208.0	72.45 ± 81.78	7.33–209.9	39.44 ± 26.23	5.65–83.81	179.6 ± 234.5	5.65–1739
IcdP	99.54 ± 141.4	ND–945.3	49.14 ± 49.72	ND–168.1	33.54 ± 36.83	ND–105.8	31.06 ± 28.44	3.22–111.1	84.26 ± 126.9	ND–945.3
LMW PAHs[a]	378.1 ± 293.1	10.87–1858	181.6 ± 142.6	38.80–433.7	165.2 ± 130.3	18.09–446.4	153.1 ± 107.5	17.47–352.9	326.0 ± 276.7	10.87–1858
HMW PAHs[b]	1501 ± 1816	59.37–9775	636.0 ± 492.1	93.75–1457	428.4 ± 496.3	29.37–1533	295.6 ± 168.3	42.33–586.2	1241 ± 1652	29.37–9775
$\sum PAH_7$[c]	861.6 ± 1025	37.23–5773	355.6 ± 276.0	51.23–791.8	248.5 ± 288.8	13.80–868.0	174.0 ± 98.21	22.38–339.7	712.0 ± 933.9	13.80–5773
$\sum PAH_{16}$	1879 ± 2077	86.94–11,565	817.6 ± 603.8	222.2–1697	596.5 ± 559.9	49.90–1723	448.7 ± 234.9	59.80–900.8	1567 ± 1898	49.90–11,565

ND indicating that it is not detectable

[a] Low molecular weight 2–3 ring PAHs

[b] High molecular weight 4–6 ring PAHs

[c] Sum of seven carcinogenic PAHs including BaA, Chr, BbF, BkF, BaP, IcdP, and DahA

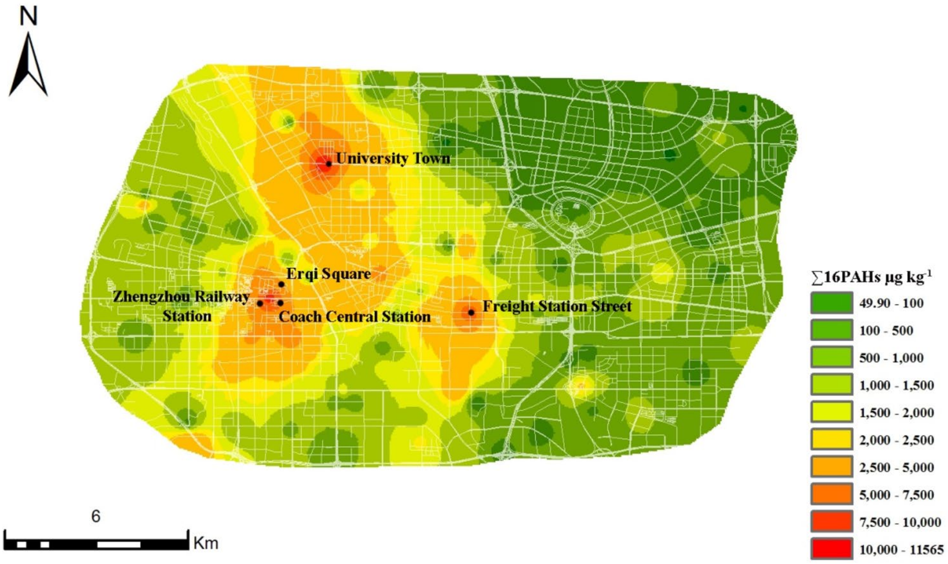

Fig. 1 Spatial distributionof $\sum_{16}$PAHs in urban soils of Zhengzhou City, China

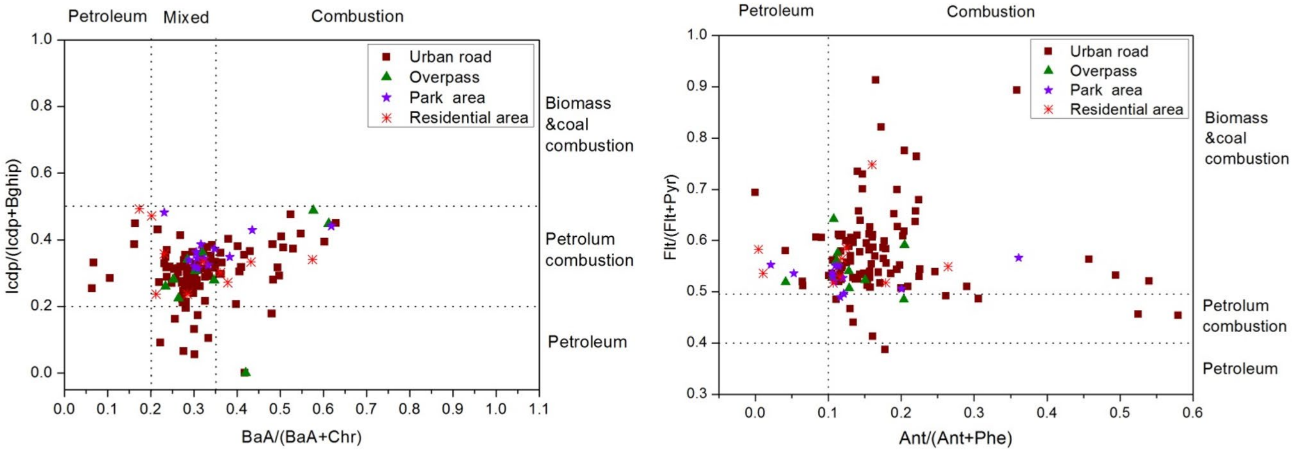

Fig. 2 Diagnostic ratios for source analysis of PAHs in the surface soil from the main district of Zhengzhou City, China including urban road, overpass, residential area and park

indexes a petroleum source, while > 0.35 implies petroleum combustion and 0.2–0.35 indexes sources from mix sources. Meanwhile, a ratio of IcdP/(IcdP + BghiP) < 0.2 indexes a petroleum source and 0.2–0.5 implies petroleum combustion and > 0.5 indexes sources from combustion of biomass and coal (Katsoyiannis et al. 2007). The diagnostic ratios obtained from the 130 samples are shown in Fig. 2. The results of this study indicate that the ratio of Ant/(Ant + Phe) ranged from 0.004% to 0.58%, 92.3% of which are higher than (0.1) The ratio of Fla/(Fla + Pyr) ranged from 0% to 0.91%, 90% of which are higher than 0.5. The ratio of IcdP/(IcdP + BghiP) ranged from 0 to 0.49 and the ratio of BaA/(BaA + Chr) ranged from 0.06% to 0.63%, 96.1% of which are higher than (0.2) These ratios indicated that PAHs in the urban soil were mainly originated from petroleum, coal and biomass combustion. Thus, both the combustion of coal and biomass and emissions from automobile transportation are the main sources of PAHs contaminants in urban soil.

Health risk assessments of the studied PAHs in the urban soil through the three main exposure pathways including ingestion, dermal contact and inhalation were performed for children, adolescence and adulthood (Peng et al. 2011). When the ILCR value is $\leq 10^{-6}$, 10^{-6} <to $< 10^{-4}$, and $\geq 10^{-4}$, the carcinogenicity level is expressed as negligible risk, potential risk, and high risk, respectively (Man et al. 2013). The ILCR values for children, adolescence, and adulthood in different urban areas is shown in Table 2. The ILCR value of ingestion or dermal was significantly higher than that of inhalation (Table S3). The total ILCR value of urban road was higher than that of the overpass, residential area, and

Table 2 Potential health risks (total ILCR) for urban soils of different land use types

Compounds	Area	Childhood		Adolescence		Adulthood	
		Male	Female	Male	Female	Male	Female
PAHs	Urban road	2.45E−06	2.52E−06	1.53E−06	1.59E−06	6.03E−06	6.60E−06
	Overpass	9.19E−07	9.46E−07	5.77E−07	6.00E−07	2.27E−06	2.48E−06
	Residential area	7.16E−07	7.37E−07	4.50E−07	4.67E−07	1.77E−06	1.94E−06
	Park	4.99E−07	5.14E−07	3.14E−07	3.26E−07	1.23E−06	1.35E−06

park. Meanwhile, the total ILCR values of the six groups of people divided by age and gender in urban road areas were all higher than the threshold for carcinogenic risk (1×10^{-6}), indicating that the urban soils of this area possessed potential cancer risk. Similarly, Wang et al. (2017b) found that the ILCR value exposed to PAHs in urban road soil of Nanjing was higher than that in residential area and park. Actually, the cancer risk of soil in the urban road area via ingestion and dermal contact pathways exceeded 1×10^{-6} in the present study (Table S3), which needs to be taken seriously. For the ILCR value, the distribution of ages and genders shows a decreasing trend of adulthood > childhood > adolescence and female > males. The highest ILCR in the male was recorded in the urban road, and the estimated values in childhood, adolescence and adulthood were 2.45×10^{-6}, 1.53×10^{-6}, and 6.03×10^{-6}, respectively. While for females, there were 2.52×10^{-6}, 1.59×10^{-6}, and 6.60×10^{-6}, respectively. These values are higher than those measured on Xi'an urban road (Bao et al. 2018), but less than those in urban green areas of the Beijing (Zhang et al. 2016).

In this study, surface soils of different urban functional area were collected from Zhengzhou City, China and the levels of PAHs, possible sources, and human health risks were analyzed. The $\sum PAH_{16}$ concentrations in the urban soil samples ranged from 49.90 to 11,565 μg kg^{-1}. Higher concentrations of PAHs were mainly concentrated in the middle of the city. The mean concentrations of PAHs decreased in the following order: urban road > overpass > residential area > park. Analysis based on the diagnostic rate demonstrated that PAHs originating mainly from pyrolysis sources such as traffic emissions and combustion of coal and biomass. Based on ILCR model, the present results indicated that PAHs in urban road areas had potential carcinogenic risks and carcinogenic risk decreased with road > overpass > residence > park and that female had a higher carcinogenic risk than male. The urban soil of Zhengzhou City, especially the land on both sides of the road has potential health risks to the residents and should pay further attention.

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