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Health Risk Due to Sixteen PAHs in Residential Street Soils from Industrial Region, Ghaziabad, Uttar Pradesh, India

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Abstract

The close proximity of soils to humans can significantly influence human exposure through ingestion, inhalation, and dermal absorption to polycyclic aromatic hydrocarbons (PAHs) in soil and consequently may cause health risks. PAHs are released to the environment mainly from pyrogenic sources and petrogenic sources. On the basis of their toxicity, some PAHs have been classified as probable carcinogens and a few others as possible carcinogens to humans, therefore, sixteen PAH compounds have been listed by the US Environmental Protection Agency (USEPA) as priority pollutants. Globally, several studies have been carried out on PAHs in soils and their health implications; however, a few references on human health risk due to PAHs through soil are available for rapidly industrializing India. This study was focused on human health risk of priority sixteen PAHs in residential soils from industrial region, Ghaziabad, Uttar Pradesh, India. On the basis of observed lower concentrations of total sixteen PAHs than the guideline values, the estimated average daily intakes (lifetime average daily dose) of PAHs through soil were lower than the recommended index dose for humans. Consequently, the estimated cancer risk (incremental lifetime cancer risk) was within the acceptable risk distribution range (10^{-6} -10^{-4}) for human adults and children. Observed PAHs concentrations were dominated by high molecular weight PAHs, and seven carcinogenic PAHs accounted for 52 % out of the $\sum$16 PAHs. Studied soils were classified as mild contaminated with PAHs, thus may be categorized with low health risk to humans residing in the vicinity.

1. Introduction

Majority of polycyclic aromatic hydrocarbons (PAHs) compounds are released to the environment through various anthropogenic and natural activities. Naturally, they are released from forest fires, volcanic eruptions, diagenesis of organic matter, and biochemical synthesis (Wilcke 2000). Anthropogenic activities includes various pyrogenic sources (incomplete biomass combustion processes) and petrogenic sources (petroleum products) (ATSDR, 1995).

In urban areas, majority of PAHs associated with anthropogenic activities through petrogenic sources (unburned petroleum and its products such as gasoline, kerosene, diesel and lubricating oil) and pyrogenic sources (incomplete combustion of carbon containing materials such as fossil fuels, wood and solid wastes) (Baek *et al*. 1991).

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Soil contamination with PAHs occurs mainly from atmospheric depositions of stationary sources of power plants and industries, and diffused sources from vehicular emissions, road asphalt and residential heating for cooking. (Maliszewska-Kordybach et al. 2008; Essumang et al. 2011; Hussain and Hoque, 2015). Soils are considered as major reservoir and sink for pollutants because of their quantity and holding capacity, (Wild and Jones, 1995). The close proximity of soils to humans can significantly influence human exposure through ingestion, inhalation, and dermal absorption to PAHs in soil and consequently can cause health risks. Human exposure to PAHs can also takes place via biomagnifications (indirectly transfer from soil to plants and animals) in food chain. Due to a rapid industrialization, concentrations of PAHs in urban soils have been increasing from various sources (Williams *et al.*, 2013). Their characteristic property of non-polar, low solubility, high affinity to particulate material, and resistance to degradation make these compounds to remain in the soils for long periods of time. Therefore, the soil is considered a major reservoir of PAHs and is a good indicator of environmental pollution and environmental risk for human exposure to PAHs (Wilcke, 2007; Sun *et al*. 2012).

On the basis of their toxicity, some PAHs have been classified by International Agency for Research on Cancer (IARC) as probable carcinogens and a few others as possible carcinogens to humans (IARC, 2006). Therefore, sixteen PAHs [naphthalene (Npt), acenaphthene (ANe), acenaphthylene (ANy), fluorene (Fle), phenanthrene (Phe), anthracene (Ant), fluoranthene (Flt), pyrene (Pyr), benz[a]anthracene (BaA), chrysene (Chr), benzo[b] fluoranthene (BbF), benzo[k] fluoranthene (BkF), benzo[a] pyrene (BaP), dibenzo[a,h] anthracene (DBA), indeno [1,2,3-cd] pyrene (Ind), and benzo[ghi] perylene (BghiP)] have been listed by the US Environmental Protection Agency (USEPA) as priority pollutants (USEPA, 2014).

Several studies have been undertaken worldwide to establish the relationship between human health and PAHs contaminated soil (Li *et al*., 2011; Obiri *et al*., 2013; Williams *et al*., 2013; Man, *et al*., 2013; Wang et al., 2014; Soltani et al., 2015). However, a few references on human health risk due to PAHs through soil are available for rapidly industrializing India (Kumar *et al*., 2013a, b; 2014a,b,c).This study was undertaken on quantification of the priority sixteen PAHs levels in residential street soils from industrial region of Ghaziabad, Uttar Pradesh, India for the estimation of probable lifetime cancer risk for humans through soil. Ecotoxicological health risk of PAHs in soils was also evaluated using recommended environmental soil quality guidelines (SQGs).

2. Materials and Methods

2.1. Sampling

The sampling area was industrial region of Ghaziabad district in Uttar Pradesh, India, a dynamic mixture of rural-urban settlement and characterized by the presence of several industrial activities. Area experiences a humid sub-tropical environment with ambient temperature between $27 - 45\ ^0C$ during summers and $4-25\ ^0C$ during winters.

Sub-surface (at the depth of ~5 - 10 cm) residential street soil was collected from twelve sampling location during February 2014. For each sampling location, three sub-samples (~500 g) in the radius of ~10 m were taken from the same location. After collection, unwanted materials were removed manually. Collected soils from sampling points at each location mixed thoroughly to ensure that the sample collected is the true representative samples of that location. A sufficient quantity of mixed soil was collected in cleaned wide mouth amber coloured glass bottles. All collected samples were stored in refrigerator at ~4 ^0C until further processing for extraction and analysis.

2.2. Sample Processing

Soil samples were air dried in clean and dark environment in the laboratory. Dried soils were smooth grinded, sieved though 1mm sieve and stored in glass bottle in dark at 4±2 °C until further processing for analysis.

USEPA methods were used for sample extractions and clean-up. Extraction of soil samples was carried out using acetone-hexane (1:1 v/v) solvent mixture in ultrasonic bath. Extracts were filtered, concentrated using rotary evaporator (Eyela, Tokyo, Japan) and subjected to activated silica gel (100–200 mesh) column chromatography. Aliphatic hydrocarbons and other unwanted polar compounds were removed with Pentane. Elution of PAHs from the column was made using methylene chloride/pentane (2:3) (v/v) solvent mixture. The eluted fraction containing PAHs was concentrated and solvent exchanged to acetonitrile for quantification by HPLC.

2.3. Instrumental Quantifications

Analysis of PAHs was performed using HPLC (Agilent 1100 Series) equipped with diode array detector (DAD, λ = 254 nm), quaternary pump and degasser. A 20 µl clean sample extract was separated for PAHs on LC-PAH Supelcosil™ (25cm x 4.6 mm, 5 µm film) analytical column and Eclipse XDB-C8 (4.6 x 12.5 mm, 5 µm) as guard column. Gradient flow of acetonitrile and water was used as mobile phase with linear flow of 60% acetonitrile and 40% water @1.0 ml/min to 100% acetonitrile in 42 min (Kumar et al., 2013a,b).

2.4. Quality Control Analysis

For quality control/assurance (QC/QA) analysis, procedural blanks (concentration, <DL "BDL"), duplicate samples (<10%), multi-level calibration curves (r^2, 0.999) and calibration verification (<5%) were undertaken. The peak identification was conducted by the accurate retention time of individual 16 PAH reference standard solutions. Method accuracy was checked by recovery study of fortified soil samples with the addition of known quantity of pure reference standards. Aliquots of soil spiked before extraction with 10 μg/kg soil for PAH compounds. The spiked and non-spiked soil samples were processed separately in duplicate as real samples, and values obtained in spiked and non-spiked samples were systematically compared. The percent recoveries were 82%-109% for 16 PAHs.

Method detection limit (DL) was obtained by processing the eight aliquots of a spiked sample with quantity of the standard materials to produce a valid quantifiable peak at signal to noise ratio >3:1 (S/N>3) (WDNR, 1994). The obtained standard deviation from eight replicate analysis was multiplied with 3 (t-students value for eight replicates at 99% confidence level) to obtain DL, which ranged between 0.09 - 0.21 (±0.03) μg kg-1. Concentrations of PAHs below detection limits were reported as <1 μg kg-1 (BDL) and taken as zero in calculations. All analysis was carried out in duplicate and the average of results was used in calculations. Moisture content of soils was determined separately to report data on dry weight basis. The results of the analysis are reported in μg kg-1 dry-weight (dw).

2.5. Health Risk Estimation

Due to sufficient evidence of carcinogenicity in experimental animals, BaP was considered to the potential reference mutagens and often used as a general indicator of PAHs and regarded as good index for PAH toxicity by the International Agency for Research on Cancer of World Health Organization (WHO) (IARC, 2006).Therefore, for other PAHs, BaP toxic equivalent factors (TEFs) derived and used for quantification of the cancer risk as benzo[a]pyrene (BaP) toxicity equivalency (BaP$_{TEQ}$) (Nisbet and LaGoy, 1992; Larsen and Larsen, 1998).

For human health risk assessment, soil ingestion pathway was considered as major exposure route of PAHs.In this study, incremental lifetime cancer risk (ILCR) was estimated from the lifetime average daily dose (LADD) of PAHs through soil. Due to non-availability of human exposure data for India, exposure parameters derived by US EPA (USEPA, 1989, 2014) have been used for risk assessment study. These exposure parameters have been used worldwide for similar studies in the literature. LADD and ILCR were estimated

from the following equations:

$$LADD \ (mg \ kg^{-1} \ d^{-1}) = (Cs \ x \ IR \ x \ F \ x \ EF \ x \ ED)/(BW \ x \ AT) \quad [1]$$

$$Cancer \ Risk = LADD \ x \ CSF \quad [2]$$

Where, Cs is the concentration of individual PAH in soil (μg kg-1), IR is the soil ingestion rate, F is the unit conversion factor, EF is exposure frequency (days/year), ED is the life time exposure duration (year), BW is the bodyweight (kg), and AT is the averaging time for carcinogens (days). CSF is oral cancer slope factor (7.3 mg/kg/day for BaP) (USEPA 2014). For this study, CSF for other 15 PAHs was calculated from CSF of BaP by multiplying with the TEFs.

Nemerow composite index (NCI) (P value) was used to assess soil environmental quality, which was based on environmental SQGs for Npt, Phe, Pyr, BaA, BbF, BkF, BaP, DBA and Ind (CCME, 2010). NCI calculations wereas following equation:

$$P = \{SQRT \ [(P_{i\text{-}mean})^2 + (P_{i\text{-}max})^2]/2\} \quad [3]$$

Where, $P_{i\text{-}mean}$ is the mean value of individual PAHs indexes and $P_{i\text{-}max}$ is the maximum value of individual PAHs indexes (Cheng *et al*., 2007). Soil quality classified into five environmental pollution grades which are safe ($P \leq 0.7$), warning ($0.7 < P \leq 1.0$), light pollution ($1.0 < P \leq 2.0$), moderate pollution ($2.0 < P \leq 3.0$) and heavy pollution ($P > 3.0$).
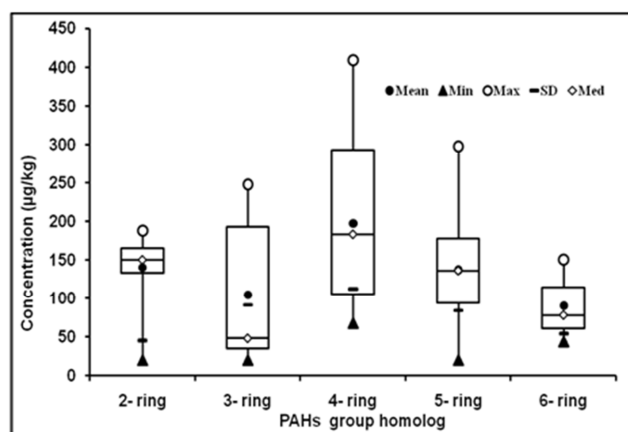
Fig. 1. Boxplot of PAHs concentrations in soil.

3. Results and Discussion

The concentrations of PAHs in residential street soils of the Ghaziabad industrial region are presented in Tables 1and Fig.1. The concentrations of ∑16PAHs ranged between240 - 1308 μg kg-1 with the mean of 574±304 μg kg-1. Concentration of acenaphthene, acenapthylene and fluorene was below detection limit at all the locations. Priority sixteen PAHs have been characterized by the aromatic rings present. PAHs with different aromatic rings were in increasing order

of 4-ring PAHs > 2-ring PAHs > 5-ring PAHs > 3-ring PAHs > 6-ring PAHs. The average concentration of PAHs with different rings was 197±112 µg kg^{-1}, 140±45 µg kg^{-1}, 137±84 µg kg^{-1}, 105±91 µg kg^{-1} and 91±54 µg kg^{-1}, for 4-ring, 2-ring, 5-ring, 3-ring and 6-ring PAHs, respectively, and their contribution accounted for 29%, 21%, 20%, 16% and 14% of $\sum$16PAHs. PAHs can be classified according to their molecular weights i.e. low molecular weight PAHs (LMW$_{PAHs}$) (<4 aromatic rings) and high molecular weight

PAHs (HMW$_{PAHs}$) (>4 aromatic rings). The observed concentrations of HMW$_{PAH}$ were comparatively higher (62 % of $\sum$16PAHs) (range, 84 – 930 µg kg^{-1};mean, 358±236 µg kg^{-1}) than LMW$_{PAHs}$ (38 %) (range,34 – 378 µg kg^{-1}; mean, 216±105 µg kg^{-1}).IARC suggested seven probable human carcinogens PAHs (BaA, BaP, BbF, BkF, Chr, DBA and Ind) (7PAH$_{carcinogen}$) ranged from 84 to 598 µg kg^{-1}, with a mean of 297±152 µg kg^{-1}, and accounted for 52 % of the $\sum$16 PAHs.

Table 1. Concentration and BaP toxicity equivalent (BaP$_{TEQ}$) of sixteen PAHs in residential street soils.

PAH compounds	Concentration (µg kg-1)			BaP$_{TEQ}$ (µg TEQ kg-1)		
	Range	Mean	%	Range	Mean	%
Naphthalene	20 - 188	140±45	12	0.02 -0.19	0.14±0.04	0.07
Acenaphthylene	<1 (BDL)					
Acenaphthene	<1 (BDL)					
Fluorene	<1 (BDL)					
Phenanthrene	20 - 196	76±64	7	0.01-0.10	0.04±0.03	0.02
Anthracene	<1 (BDL) - 156	144±17	12	0.07-0.08	0.07±0.01	0.04
Fluoranthene	<1 (BDL) - 160	119±39	10	4.10-8.00	5.93±1.96	2.95
Pyrene	<1 (BDL) - 100	73±28	6	0.04-0.10	0.07±0.03	0.04
Benzo(a)Anthracene	<1 (BDL) - 148	84±39	7	0.20-0.74	0.42±0.20	0.21
Chrysene	20 - 198	86±61	7	0.60-5.94	2.59±1.84	1.29
Benzo(b)Fluoranthene	20 - 178	83±65	7	2.00-18	8.31±6.51	4.13
Benzo(k)Fluoranthene	<1 (BDL) - 78	51±28	4	1.10-3.90	2.53±1.40	1.26
Benzo(a)Pyrene	<1 (BDL) - 192	88±75	8	20-192	88±75	44
Benzo(g,h,i)Perylene	<1 (BDL) - 82	77±7	7	1.44-1.64	1.54±0.14	0.77
Dibenzo(a,h)Anthracene	<1 (BDL) - 78	78±0	7	86-86	86±0	43
Indeno(1,2,3-Cd)Pyrene	<1 (BDL) - 72	58±20	5	4.40-7.20	5.80±1.98	2.88
$\sum$16PAHs	240 - 1308	574±304	100	2.68-254	65±84	100
$\sum$PAHLMW	34 - 378	216±105	38	0.02-0.28	0.17±0.07	0.26
$\sum$PAHHMW	84 - 930	358±236	62	2.52-253	65±84	99.7
$\sum$7PAHcarcinogen	84 - 598	297±152	52	2.52-244	63±81	97.0

Due to documented carcinogenicity and endocrine-disruptive activity of PAHs (Davis *et al*., 1993), the carcinogenic potency (BaP$_{TEQ}$) of 16 PAHs was estimated and presented in Table 1. The BaP$_{TEQ}$ for $\sum$16 PAHs ranged between 2.68-254 µg BaP$_{TEQ}$ kg^{-1}, with the mean 65 ± 84µg BaP$_{TEQ}$ kg^{-1}. BaP$_{TEQ}$ for $\sum$7PAH$_{carcinogen}$ ranged from 2.52-244 µg BaP$_{TEQ}$ kg^{-1} with an average 63±81 µg BaP$_{TEQ}$ kg^{-1} and accounted for 97% to $\sum$BaP$_{TEQ}$ of 16 PAHs. HMW$_{PAHs}$ were the major contributors and accounted for >99% to $\sum$BaP$_{TEQ}$, however, BaP$_{TEQ}$ of LMW$_{PAHs}$ was lowwith <1% contribution to the total carcinogenic potency of PAHs. Due to high TEF value for BaP and DBA, they significantly increase the ΣBaP$_{TEQ}$ with the contribution for more than 87% to $\sum$BaP$_{TEQ}$.

Environmental guidelines for PAHs in soil are not available in India, thus, soil quality guidelines (SQGs) from National Oceanography and Atmospheric Administration (NOAA), USA (NOAA, 1999) and Canadian government (CCME, 2010) were used for the assessment of ecotoxicological health effect of PAHs in soil. The observed levels of PAHs from this study were lower than the recommended SQGs (individual PAHs range, 700-10,000 µg kg^{-1}), suggesting least environmental health risk and adverse effects on the soil biota. On the basis of classification of soil contamination

with PAHs (Maliszewska-Kordibach, 1996), the studied residential street soils can be categorized as weakly contaminated with PAHs (574 ± 304 µg kg^{-1}). The Nemerow composite indices (NCI) for Ghaziabad residential soils ranged between 0.01-0.20 with the mean of 0.07, suggesting that the pollution level of residential soils due to PAHs is within safe category.

Health risk assessment was based on assumption that human adults and children exposed to PAHs through soil for during their life span of 70 years and 12 years, respectively. Estimated LADD and ILCR for human adults and children are presented in Table 2, Figure 1 and figure 2. The average LADD of $\sum$16PAHs and $\sum$7PAH$_{carcinogen}$ for human adults was 9.6x10^{-7}mg kg^{-1} d^{-1}and 5.0x10^{-7} mg kg^{-1} d^{-1};however, their LADD for children were5.6x10^{-7}mg kg^{-1} d^{-1}and 2.9x10^{-7}mg kg^{-1} d^{-1}, respectively. From oral studies, United Kingdom derived recommended index dose (RID) of BaP for a 70 kg adult as 0.02 µg kg^{-1}d^{-1} or 2.0x10^{-5} mg kg^{-1} d^{-1} (Environment Agency, 2002). On the other hand, USEPA developed an oral slope factor of 7.3 × 10^{-3} per µg BaP kg^{-1} bw d^{-1}. Explanatory means that ingestion of 1 µg BaP kg^{-1} bw d^{-1} or 1 x 10^{-3} mg kg^{-1} d^{-1} would pose a lifetime cancer risk of 7.3 × 10^{-3}. The estimated LADD in present study are much lower than dose

estimated by USEPA. Subsequently, average ILCR due to $\sum$16PAHs and $\sum$7PAH$_{carcinogen}$ for human adults was derived as 7.7×10^{-5} and 3.8×10^{-6}, respectively. The average ILCR for children was 4.5×10^{-5} and 2.2×10^{-6}, respectively. This estimated ILCR have been found within acceptable risk distribution range $(10^{-6}-10^{-4})$ (US EPA, 1989) for human adults and children.
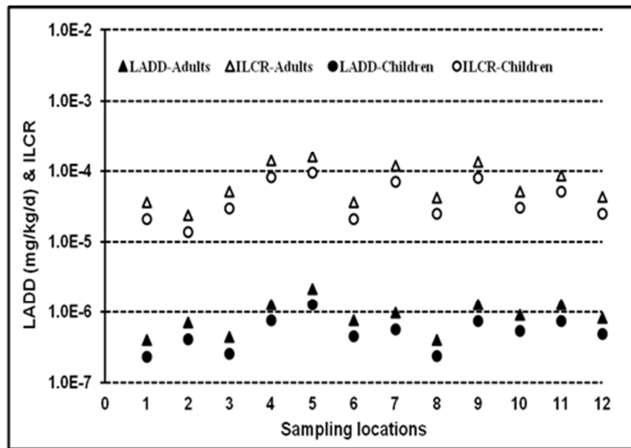

Fig. 2. Average daily intake of $\sum$16PAHs and their cancer risk to humans through soil.

Table 2. LADD ($mg\ kg^{-1}\ d^{-1}$) and ILCR due to sixteen PAHs for humans through soil.

PAH compounds	LADD		ILCR	
	Adults	Children	Adults	Children
Naphthalene	2.3×10^{-7}	1.4×10^{-7}	3.2×10^{-5}	1.9×10^{-5}
Acenaphthylene	*			
Acenaphthene	*			
Fluorene	*			
Phenanthrene	1.3×10^{-7}	7.4×10^{-8}	$3.5 10^{-5}$	2.0×10^{-5}
Anthracene	2.4×10^{-7}	1.4×10^{-7}	6.6×10^{-5}	3.9×10^{-5}
Fluoranthene	2.0×10^{-7}	1.2×10^{-7}	5.4×10^{-7}	3.2×10^{-7}
Pyrene	1.2×10^{-7}	7.1×10^{-8}	1.7×10^{-5}	9.8×10^{-6}
Benzo(a)Anthracene	1.4×10^{-7}	8.2×10^{-8}	3.8×10^{-6}	2.2×10^{-6}
Chrysene	1.4×10^{-7}	8.5×10^{-8}	6.6×10^{-7}	3.9×10^{-7}
Benzo(b)Fluoranthene	1.4×10^{-7}	8.1×10^{-8}	1.9×10^{-7}	1.1×10^{-7}
Benzo(k)Fluoranthene	8.4×10^{-8}	5.0×10^{-8}	2.3×10^{-7}	1.4×10^{-7}
Benzo(a)Pyrene	1.5×10^{-7}	8.6×10^{-8}	2.0×10^{-8}	1.2×10^{-8}
Benzo(g,h,I)Perylene	1.3×10^{-7}	7.5×10^{-8}	8.8×10^{-7}	5.2×10^{-7}
Dibenzo(a,h)Anthracene	1.3×10^{-7}	7.6×10^{-8}	1.6×10^{-8}	9.5×10^{-9}
Indeno(1,2,3-Cd)Pyrene	9.7×10^{-8}	5.7×10^{-8}	1.3×10^{-7}	7.8×10^{-8}
$\sum$16PAHs	9.6×10^{-7}	5.6×10^{-7}	7.7×10^{-5}	4.5×10^{-5}
$\sum$PAH$_{LMW}$	3.6×10^{-7}	2.1×10^{-7}	6.9×10^{-5}	4.1×10^{-5}
$\sum$PAH$_{HMW}$	6.0×10^{-7}	3.5×10^{-7}	8.2×10^{-6}	4.8×10^{-6}
$\sum$7PAH$_{carcinogen}$	5.0×10^{-7}	2.9×10^{-7}	3.8×10^{-6}	2.2×10^{-6}

*not calculated as PAH concentration was <1 µg kg-1 (BDL).

4. Conclusions

Study concluded that the residential soils in industrial region of Ghaziabad, Uttar Pradesh, India were mildly contaminated with priority sixteen PAHs. Estimated daily intakes of PAHs through accidental soil ingestion for humans were lower than the recommended index dose for benzo(a)pyrene. Cancer risk for human adults and children was also found within the acceptable safe risk range $(10^{-6} – 10^{-4})$. Due to insufficient data on exposure through other pathways, study concluded that human population residing in the area has extremely low health risk due to PAHs exposure through soils.

Acknowledgement

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