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Priority Polycyclic Aromatic Hydrocarbons in Industrial Soils from Western Corridor of Pune, India: Levels, Composition Profiles, Source Identification and Health Risk

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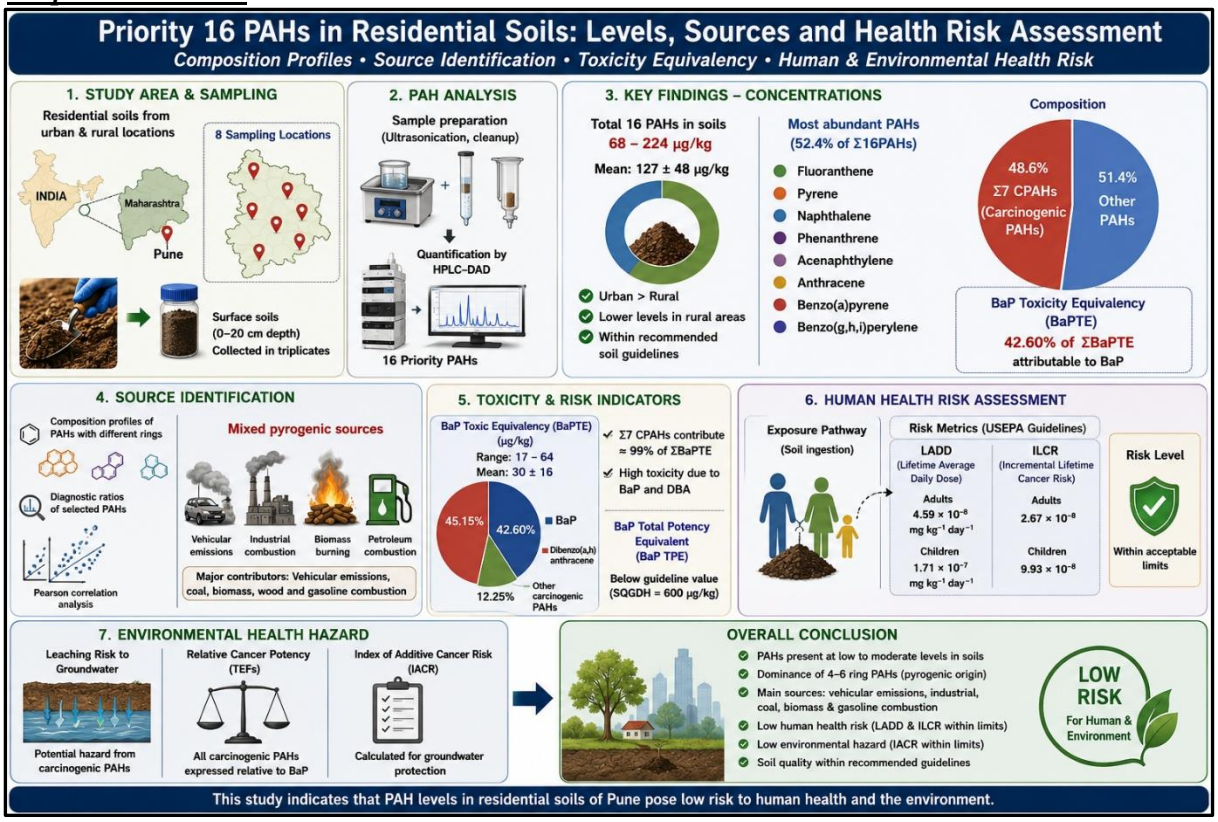
Graphical Abstract:



Abstract - Priority sixteen PAHs were assessed in residential soils from both urban and rural locations in this study in order to evaluate their composition profiles, potential sources, and impacts on human and environmental health. In rural areas, the concentrations of total 16PAHs in soils were relatively lower, with a mean of 127±48 µg/kg and a range of 68-224 µg/kg. Among the most common PAHs, fluoranthene, pyrene, naphthalene, phenanthrene, acenaphthylene, anthracene, benzo(a)pyrene, and benzo(g,h,i)perylene accounted for almost 52.4% of the total. 48.6% of ∑16PAHs were ∑7CPAHs (seven carcinogenic PAHs). Nevertheless, 42.60% of ∑BaPTE was attributable to ∑7CPAHs' benzo(a)pyrene toxicity equivalency (BaPTE).

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Recent measurements at other locations were compared with the measured amounts of PAHs and associated BaPTE. The correlation coefficient, diagnostic ratios of certain PAHs, and composition profiles of PAHs with various aromatic rings were utilized to identify potential PAH sources. The lifetime average daily dose (LADD) and incremental lifetime cancer risk (ILCR) of PAHs in soil for adults and children were calculated, discussed, and presented in accordance with USEPA guidelines. Additionally, the potential hazard to potable groundwater water quality from the leaching of carcinogenic PAHs from soil was assessed and provided, along with the cancer potency relative to BaP for all possibly carcinogenic PAHs and the index of additive cancer risk (IACR). The observed levels of PAHs in soils and their human health risk and environmental health hazard at different locations were assessed using recommended guidelines.

Key words: Priority PAHs, Soil, Source Identification, BaP Toxicity Equivalency, IACR, LADD, ILCR

INTRODUCTION

A class of chemical molecules with two or more benzene rings is known as polycyclic aromatic hydrocarbons, or PAHs. Numerous man-made sources, such as incomplete burning of coal, petroleum products, and biomass (pyrogenic sources) and petroleum products (petrogenic sources), release PAHs into the environment. By binding to DNA and forming DNA-PAH adducts, PAHs produce genotoxic effects, mutations, and the development of carcinogenesis (ATSDR 1995; Perera et al. 2011; Herbstman et al. 2012; Gurbani et al. 2013). Accordingly, the US Environmental Protection Agency and the European Union have designated sixteen PAHs as priority pollutants (USEPA 2014; EC 2001). Some priority PAHs have been classified by the International Agency for Research on Cancer (IARC) as probable carcinogenic to humans, while few other classified as possible carcinogens to humans (IARC 2006).

Long-distance transport of PAHs via the atmosphere is possible, and they can be separated into gaseous and particulate phases (ATSDR 1995; Chen et al. 2017). They are widely dispersed throughout the many environmental compartments, such as soil, plant, water bodies, and air (Wang et al., 2017). PAHs have a high affinity for particles and are adsorbed on dusts, sediments, and soils because of their distinctive low volatility and poor aqueous solubility characteristics (Aleksandra et al. 2019). Through volatilization, degradation, and leaching, soils may have a significant impact on the distribution and destiny of PAHs in the environment (Wilcke 2000; Li et al. 2018). The main causes of PAH concentrations in urban soils, where they may last for a long time, are industrial processes and automobiles. Because of their quantity and capacity to hold pollutants, soils may serve as both a source and a sink for many PAHs in the global circulation (Wild and Jones 1995). Human exposure is caused by the concentration of PAHs in soil and the soil's near proximity to people. Through ingestion, inhalation, or skin absorption mechanisms, PAHs in soils can affect human health (ATSDR 1995, 2005). Therefore, any risk evaluations including potentially hazardous persistent organic pollutants, such as PAHs, could take soil into account.

The impacts of PAH chemicals on human health through soils have been the subject of numerous research conducted globally in recent years (Chen et al. 2018; Lang et al. 2018; Ali et al. 2018; Gereslassie et al. 2018; Dumanoglu et al. 2017; Kasaraneni and Vinka 2016; Yu et al. 2014). According to Hafner et al. (2005), there is a connection between PAH emissions and energy use due to the demands of urbanization, industrialization, and population increase. According to Chawda et al. (2020), Gope et al. (2018), Singh & Agarwal (2018), Tarafdar & Sinha 2018a, Hazarika & Srivastava (2016), Sampath et al. (2015), and Kaur et al. (2013), research on the risk of PAHs to humans has been conducted in India, a developing nation.

However, there aren't many investigations on the effects of PAH exposure through soils in India (Ghosh & Maiti 2019; Tarafdar & Sinha 2018b; Kumar et al. 2015b, c). Levels of PAHs in Delhi soils have

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already been reported by several writers. Delhi's agricultural soils included 830–3880 µg/kg of total PAHs, according to Agarwal et al. (2009). No study was conducted on PAHs and their human health effect; therefore, this study was conducted on the distribution of priority sixteen PAHs in soils and their impact on human health in Pune, India, were the main topics of the current study. For the purpose of evaluating the composition profiles, potential sources, and health impacts on both persons and the environment in Pune, India's urban and rural areas, the levels of priority sixteen PAHs in soils were ascertained. The toxicity of prioritized sixteen PAHs in soils was assessed using the BaP toxicity equivalency (BaPTE), BaP total potency equivalent (BaP TPE), and index of additive cancer risk (IACR). Additionally, in accordance with USEPA requirements, the lifetime average daily dose (LADD) and incremental lifetime cancer risk (ILCR) for human adults and children resulting from PAHs in soil were determined.

MATERIALS AND METHODS

Solvents, Chemicals and Standards

Chemicals and solvents utilized in sample processing and analysis were analytical and HPLC grade, respectively (Fisher Scientific, India). For extract cleanup in column chromatography, activated silica gel (100–200 mesh) (Sigma-Aldrich, USA) was utilized. Supelco (Sigma-Aldrich, USA) supplied individual standard solutions of sixteen PAHs, including naphthalene (NPT), acenaphthylene (ANY), acenaphthene (ANE), fluorene (FLE), phenanthrene (PHE), anthracene (ANT), fluoranthene (FLT), pyrene (PYR), benzo(a) anthracene (BaA), benzo(b)-fluoranthene (BbF), benzo(k)uoranthene (BkF), benzo(a)pyrene (BaP), benzo(ghi)perylene (BghiP), dibenzo(a,h)anthracene (DBA), and indeno(1,2,3-cd)pyrene (Indp). Working standard solutions with appropriate concentrations were made following the serial dilution of stock solutions, and they were utilized in HPLC calibration and quality control studies.

Study Area

Soil Sampling

Soil samples collected in triplicates from eight locations at carefully chosen sampling sites in June 2023. Approximately 500g of subsurface (0–20 cm depth) soil samples were taken from three to five distinct spots within a 100–200 m radius of one another. To guarantee a representative sample of that point at the same place, the collected subsample at each location is joined and carefully mixed after undesired elements including stones, leaves, wood, and sticks have been manually removed. After that, 200g of the mixed sample was divided into duplicates and placed into clean, wide-mouth amber glass containers. The ice was then kept and brought to the lab.

Sampling Site

S.No.	Location	Location Code	Geo-Coordinate
1	Ambegaon	S-1	19.11314, 73.73250
2	Katraj	S-2	18.45289, 73.86543
3	Narhe	S-3	18.44623, 73.82644
4	Nanded	S-4	18.45990, 73.78988
5	Khadakwasla	S-5	18.44540, 73.78012
6	Dhayari	S-6	18.77712, 73.81017
7	Wadgaon	S-7	18.54919, 73.91859
8	Ambegaon Pathar	S-8	18.45884, 73.84264

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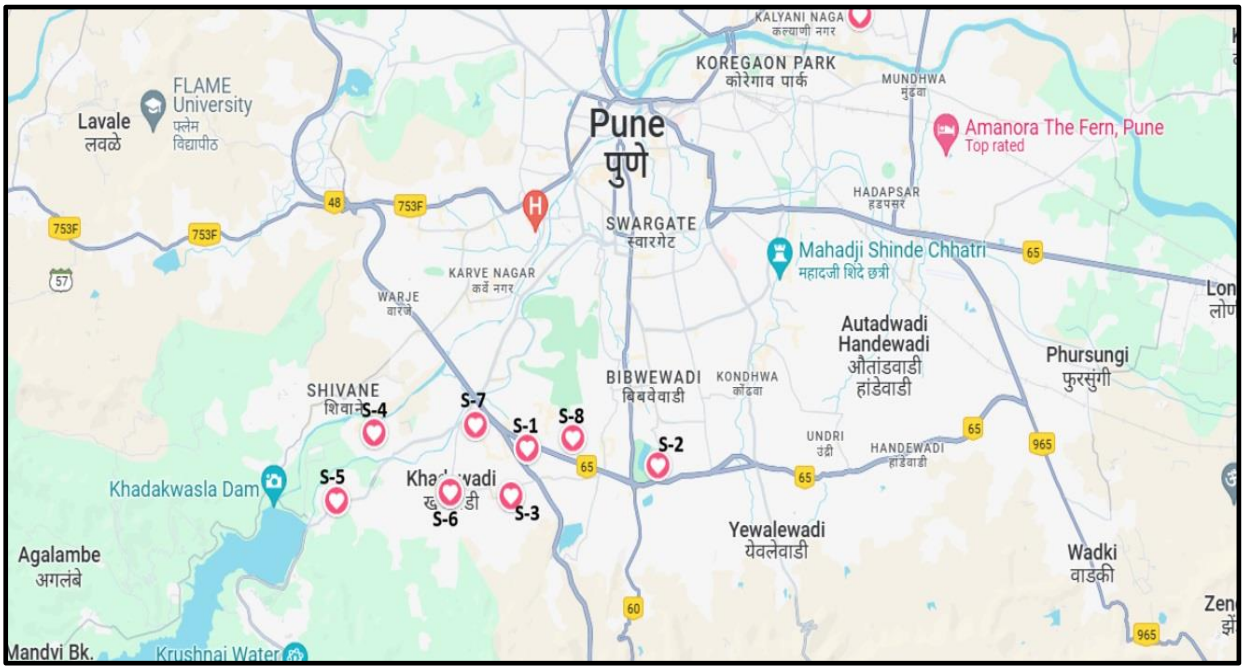

Analysis and Analytical Quality Control

The air-dried homogenized samples of 1mm size (~20 g) were extracted with acetone-hexane (1:1 v/v) using ultrasonication. A rotary evaporator (Eyela, Tokyo, Japan) was used to concentrate the sample extracts to around 2 ml at 40 °C under decreased pressure after they had been dried by passing through anhydrous sodium sulphate. The extracts were cleaned up using silica gel column chromatography in accordance with USEPA Method 3630C. HPLC (Agilent 1100 Series) with a diode array detector (DAD, λ=254 nm), LC-PAH SupelcosilTM (25cm x 4.6 mm, 5 µm film) analytical column, and Eclipse XDB-C8 (4.6 x 12.5 mm, 5 µm) as a guard column were used to quantify individual PAH constituents. For 42 minutes, the mobile phase consisted of 60% acetonitrile and 40% water with a gradient flow of 1.0 mLmin-1 to 100% acetonitrile (Kumar et al., 2012) Each analysis was performed with the necessary analytical quality control, which included calibration verification (SD, <10%), random duplicate analysis (SD, <10%), and analysis of method blanks (analyte concentrations were <MDL "method detection limit").The various PAH compounds were identified using five level calibra tion curves (r2, 0.999) with precise retention times for each standard.Every sample extract was measur ed twice (SD, <1%), and the average of the two analyses was employed in the computations.Analysis of fortified samples containing known quantities of 16 PAH standards and a surrogate standard (1-fluoronaphthalene) was used to conduct the accuracy/recovery research.

The acceptable range for matrix-spiked recoveries was 82% to 109% (±2–12%) for 16 PAHs and 94% for 1-fluoronaphthalene. The detection limits (DLs) were estimated using eight aliquots of a matrix-spiked sample that included a known quantity of the standard at a signal to noise ratio >3:1 (s/n >3). For sixteen PAH compounds, the estimated DLs varied from 0.09 to 0.21 (±0.03) µg/kg.

Toxicity Potential and Health Risk Assessment

BaP toxic equivalent factors (TEFs) or toxic equivalency (TE) of individual PAHs (Tsai et al. 2004) were used for the estimation of benzo(a)pyrene (BaP) toxicity equivalency (BaPTE) as:

$$BaPTE\ (\mu g/kg) = C \times TEF \quad (1)$$

Where, C is the concentration of individual PAH (µg/kg) and TEF is the corresponding toxic equivalent factor.

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Benzo(a)Pyrene Total Potency Equivalents (B[a]P TPE) were calculated to represent the human health protection from direct contact with soil contaminated with PAHs (CCME 2010). The total BaPTE of all carcinogenic PAHs is the B[a]P TPE. Based on an incremental lifetime cancer risk (ILCR) of 1 in 1,000,000 (10-6) from soil exposure, the suggested soil quality standards value for direct human contact (SQGDH) are 600 µg/kg (CCME 2010).

Additionally, the potential hazard to drinkable groundwater quality from the leaching of carcinogenic PAHs from soil was evaluated using the Index of Additive Cancer Risk (IACR) (CCME 2010). According to reports, the degradation of drinkable groundwater quality due to the leaching of carcinogenic PAHs from soils may increase the risk of cancer (CCME 2010). By dividing the concentration of each carcinogenic PAH by its soil quality standard for the preservation of drinkable water, the IACR's hazard index of PAHs was calculated. The hazard indices for all carcinogenic PAHs were then added up.

Given the toxicological significance of priority sixteen PAHs, unintentional ingestion of soil polluted with PAHs was thought to be the primary route of lifetime intake for estimating the risk to human health. An evaluation of the human health risks for Indian adults and children was conducted for this study, taking into account their exposure to PAH-contaminated soils on all days of the year for a total of 70 years and 12 years, respectively. Based on the computed lifetime average daily dose (LADD) of PAHs through soil, the incremental lifetime cancer risk (ILCR) was determined as follows (ATSDR 2005, USEPA 2019):

LADD (mg kg-1 d-1) = (Cs x IR x F x EF x ED)/(BW x AT) (2)

Cancer Risk = LADD x Cancer Oral Slope Factor (CSF) (3)

EF stands for exposure frequency (365 days per year), ED for lifetime exposure duration (70 years and 12 years for adults and children, respectively), BW for body weight (60 kg and 35 kg for adults and children, respectively), AT for averaging time for carcinogens (EFxED days), and Cs for the concentration of individual PAH in soil (µg/kg). According to USEPA 2019, CSF is the oral cancer slope factor. The CSF of BaP (7.3 per mg kg-1 d-1) was multiplied by the BaP toxic equivalent factors (TEFs) to get the CSF for other PAHs in this investigation.

Non-carcinogenic effects of PAHs as environmental health hazard were assessed by comparison of observed concentrations with soil quality guidelines for mammals and residential/park land use (CCME, 2010; Buckman 2008).

Further, the impact of PAHs on soil environmental quality was calculated as Nemerow composite index (*P* value), which has been used to evaluate PAHs pollution in soil (Sun et al. 2012). Accordingly, the NCI (*P* value) for this study was calculated as:

$$P\ (\text{NCI}) = \{\text{SQRT}\ [(P_{mean})^2 + (P_{max})^2]/2\} \quad (4)$$

Where, P_{mean} is the mean value of individual pollutants (PAH) and P_{max} is the maximum value of individual pollutant (PAH). Based on NCI (*P* value), the soil environmental quality categorized into five environmental pollution categories as, 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 ($P > 3.0$).

RESULTS AND DISCUSSIONS

Concentration Of PAHs In Soils

The observed concentrations (range and mean) of total PAHs in soils from Pune are presented in **Table 1**. The concentration of total PAHs in all soils ranged between 68-224 µg/kg with the mean of 127±48 µg/kg. The dominant PAHs were B(g,h,i)P (mean,16.9±13.5 µg/kg), PYR (mean, 13.7±8.69 µg/kg), DBA (mean, 13.5±4.79 µg/kg), BaP (mean, 12.7±13.1 µg/kg), BbF (mean, 12.5±7.71 µg/kg), PHE (mean, 9.70±5.59 µg/kg), FLE (mean, 8.64±14.9 µg/kg), BKF (mean, 7.61±6.05 µg/kg) and IPY (mean, 6.93±4.66 µg/kg), and accounted for 13.35%, 10.79%, 10.63%, 10.02%, 9.86%, 7.65%, 6.81%, 6.0% and 5.46%, respectively to ∑16PAHs (**Table 1**). The combined contribution of dominant PAHs was

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accounted for more than 80% to ∑16PAHs, suggested pyrogenic sources. However, dominance of fluoranthene and pyrene, suggested diesel-powered vehicles emission and biomass combustions as major sources of PAHs (Khalili et al. 1995). Similar results have been reported for PAHs sources in Delhi (Gadi et al. 2012).

Table:1 Concentrations (µg/kg) of PAHs and their BaP_{TE} in soils

PAHs	Concentration		%of ∑PAHs	BaP_{TE}		% of ∑PAHs
	Range	Mean±SD		Range	Mean±SD	
Naphthalene	BDL					
Acenaphthylene	BDL					
Acenaphthene	BDL-10.90	2.70±5.0	2.13	BDL		
Fluorene	BDL-44.50	8.64±14.9	6.81	BDL		
Phenanthrene	4.40-21.0	9.70±5.59	7.65	BDL		
Anthracene	BDL-7.00	3.85±2.89	3.04	BDL		
Fluoranthene	2.36-11.9	6.92±3.11	5.46	BDL		
Pyrene	3.90-24.7	13.7±8.69	10.79	BDL		
Benzo(a)Anthracene*	2.45-13.1	6.56±4.26	5.17	BDL-1.13	BDL	2.20
Chrysene*	1.50-8.90	4.61±2.83	3.64	BDL		
Benzo(b)Fluoranthene*	4.40-26.9	12.5±7.71	9.86	BDL-2.69	1.25±0.77	4.19
Benzo(k)Fluoranthene*	1.40-17.0	7.61±6.05	6.00	BDL-1.70	BDL	2.55
Benzo(a)Pyrene*	2.70-41.8	12.7±13.1	10.02	2.70-42	12.7±13.1	42.60
Benzo(g,h,i)Perylene	3.60-18.6	16.9±13.5	13.35	BDL		
Dibenzo(a,h)Anthracene*	6.40-21.1	13.5±4.79	10.63	6.40-21	13.5±4.79	45.15
Indeno(1,2,3-Cd)Pyrene*	BDL-13.3	6.93±4.66	5.46	BDL-1.33	0.69±0.47	2.32
∑16PAHs	68-224	127±48	100	17-64	30±16	100
2-3 ring	12-67	25±18	19.63	BDL		
4 ring	11-49	32±14	25.06	BDL-1.37	BDL	2.42
5 ring	14-79	33±19	25.89	4.78-45	15±13.6	49.34
6 ring	16-57	37±13	29.43	6.81-22.	15±4.9	48.04
*CPAH	34-113	64±24	50.78	17-64	30±16.	99

The concentrations of PAHs in the present study were compared with the recently measured PAHs in similar studies around the world including India (**Table 2)**. The average concentration of PAHs observed in present study (127±48 µg/kg) is comparable with the major cities of India, such as Bhopal, Korba, Ghaziabad and Gwalior. But, lower concentrations of PAHs (26.83 µg/kg) than this study were reported for Rettamalai, Tamil Nadu. However, elevated concentrations have been reported in other studies for soils from India including Dhanbad, and North-eastern region. Recently, elevated concentrations of seven PAHs (473-1132 µg/kg) in Delhi reported by Kumar et al. (2021).

On comparison of levels of PAHs in soils with similar soils from other countries, it was found that the observed concentrations were much lower than Banja Luka, Bosnia, Dingshu, Shengli, Nanjing, Lujia in China; Lagos, Nigeria and Tehran, in Iran; (**Table 2**).

Table 2: Comparison with PAHs in soil from different locations.

Locations	**Concentration (µg/kg)**		**Reference**
	Range	**Mean**	
Pune, MH	68-224	127	Present study
South India	ND-2934	464	Sakthivel et al. 2021
Delhi	473-1132	785	Kumar et al. 2021

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Delhi	213 - 851	550	Kumar et al. 2020
Chennai, TN	0.62-3649	64.3	Rajan et al. 2021
East India	1478 -27493	10945	Ghosh & Maiti 2019
Ukkadam	-	139.49	Kathavarayan et al. 2019
Avaniyapuram, TN	-	244.48	Kathavarayan et al. 2019
Rettamalai, TN	-	26.83	Kathavarayan et al. 2019
Koyambedu, TN	-	58.31	Kathavarayan et al. 2019
Nesapakkam, TN	-	33.44	Kathavarayan et al. 2019
Dhanbad, Jharkhand	1019 - 10856	3488	Suman et al. 2016
Korba, Chhitisgarh	7 – 2100	385	Kumar et al. 2014
Ghaziabad, UP	240 - 1308	574	Kumar et al. 2015a
Gwalior, MP	76 –1391	481	Kumar et al. 2015b
Bhopal, MP	-	122	Yadav et al. 2022
Banja Luka, Bosnia	356-11490	1990	Ilić et al. 2021
Nanjing, China	135-37400	2740	Wang et al. 2021
Dingshu, China	136-6800	1080	Wang et al. 2021
Lujia, China	164-1730	399	Wang et al. 2021
Montenegro	61 - 1457	272	Bigović et al. 2022
France	ND-31193	161	Froger et al. 2021
Tehran, Iran	169-655	396	Khoshand et al. 2017
Lagos, Nigeria	485-4513	1510	Ehigbor et al. 2020
Chile	21-4370	618	Deelaman et al. 2020

Toxic Fraction of PAHs and BaP Toxicity Equivalency (BaPTE)

Out of priority 16 PAHs, seven PAHs have been suggested the as probable human carcinogens (7CPAHs) by the International Agency for Research on Cancer (IARC 2006). Their concentrations in present study ranged between 34-113 µg/kg, with mean value of 64±24 µg/kg, and accounted for 50.78% of ∑16PAHs (**Table 1**). The average concentration of individual carcinogenic PAH was 6.56±4.26 µg/kg, 4.61±2.83 µg/kg, 12.5±7.71 µg/kg, 7.61±6.05 µg/kg, 12.7±13.1 µg/kg, 16.9±13.5 µg/kg and 6.93±4.66 µg/kg, respectively for BaA, CHR, BbF, BkF, BaP, DBA and IndP. Their contribution to ∑16PAHs was 4.6%, 3.2%, 3.9%, 4.0%, 5.2%, 4.6% and 3.7%, respectively (**Table 1**).

The World Health Organization (WHO) has identified benzo(a)pyrene (BaP), one of the priority 16PAHs, as a potential reference for the toxicity of the other 15 PAHs and frequently used as a generic indicator of PAHs. Table 1, and Figure 2, all show the predicted relative carcinogenic potential of BaPTE to the equivalent PAHs. With a mean of 30±16 µg/kg, the estimated concentration of ∑BaPTE for ∑16PAHs varied from 17 to 64 µg/kg. With a mean value of 30±16 µg/kg, the BaPTE for ∑7CPAHs ranged from 17 to 64 µg/kg and accounted for 99% of the total BaPTE. The carcinogenic potency of the 2-3-ring PAH molecules is minimal (BDL). However, the largest contributors to total BaPTE were 5-ring PAHs (49.34%) and 6-ring PAHs (48.04%), with 4-ring PAHs (2.42%) coming in second (Table 1). With a combined contribution of 87.75% to ∑BaPTE, BaP (42.60%) and DBA (45.15%) reflect the higher BaPTE values due to their high toxic equivalent factor (TEF), which greatly increased the ΣBaPTE. Consequently, DBA and BaP results indicated their significance in the soil. Furthermore, the carcinogenic potential of PAHs in soils is significantly influenced by other PAHs, including BbF, BkF, and BaA, which contribute 4.19%, 2.55%, and 2.20%, respectively, to ∑BaPTE (**Table 1**).

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Possible Source Identification of PAHs

PAHs with Different Rings

Two to three rings (NPT, ANY, ANE, FLE, PHE, and ANT), four rings (FLT, PYR, BaA, and CHR), five rings (BbF, BkF, and BaP), and six rings (BghiP, DBA, and IndP) were used to separate the sixteen PAHs. The quantity of aromatic rings that PAHs have is a good indicator of where they might have come from. Table 1 and Figure 1 list PAH homologs with various aromatic rings found in soils from several Pune sampling sites. All soil samples from Pune had concentrations of 2-3-rings, 4-rings, 5-rings, and 6-rings PAHs that ranged from 12-67 µg/kg (mean, 25±18 µg/kg), 11-49 µg/kg (mean, 32±14 µg/kg), 14-79 µg/kg (mean 33±19 µg/kg), and 16-57 µg/kg (37±13 µg/kg) and accounted for 19.63%, 25.06, 25.89%, and 29.43% to ∑16PAHs, respectively. These findings demonstrated that the majority of the PAHs at specific Pune locations were 2- and 4-ring PAHs, indicating a mixture of pyrogenic sources. According to reports, the main source of 3- to 4-ring PAHs in urban environments is vehicle exhaust. The vapor and particle phases that settle nearby can both contain these related PAHs.

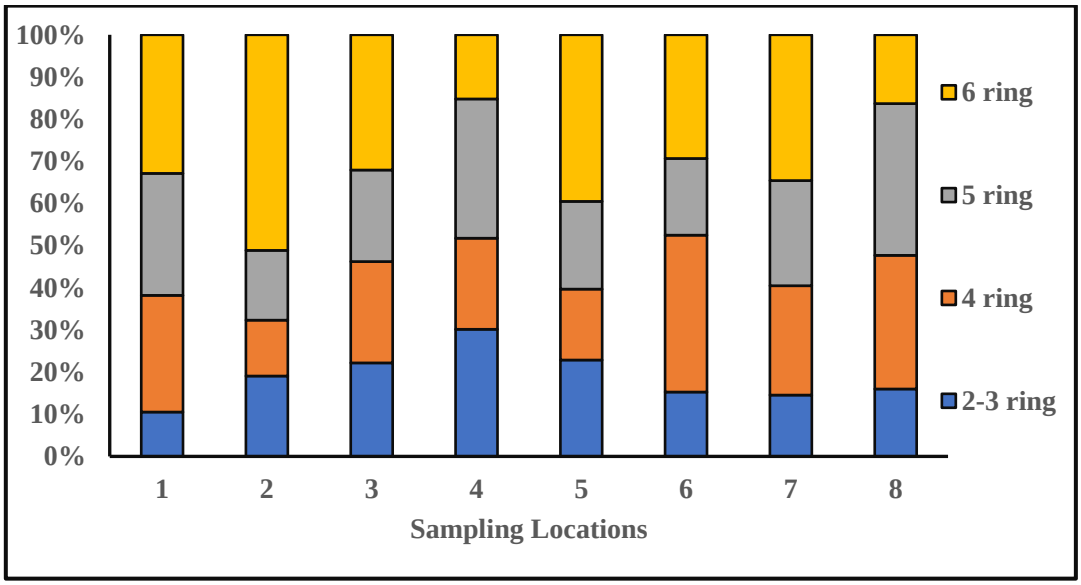

Figure: 1 Location wise Homolog pattern of PAHs with different rings

Depending on their molecular weights, PAHs in the environment can be categorized as either low molecular weight (LMW-PAHs) with <4 aromatic rings or high molecular weight (HMW-PAHs) with ≥4 aromatic rings. The octanol-water partition coefficient (Kow) and high volatility caused LMW-PAHs (2-3-ring PAHs) to separate into a gaseous phase in the environment and travel to different locations from the emission sources. However, HMW-PAHs (≥4-ring PAHs) are adsorbed to the particles, tend to deposit quickly, and stay nearer emission sources. They are then deposited on soils, vegetation, and aquatic bodies (Wang et al., 2017).

According to reports, pyrogenic sources, such as coal combustion and vehicle emissions, are often responsible for the majority of HMW-PAHs emitted into the environment. However, petrogenic sources and the burning of wood, grass, and industrial oil have been linked to the environmental domination of LMW-PAHs (Khalili et al. 1995; Marr et al. 1999; Wilcke 2007).

In order to identify potential sources of PAHs in the environment, ratios between LMW-PAHs and HMW-PAHs have been employed. Generally speaking, a ratio less than one indicated a pyrogenic source, whereas a ratio greater than one indicated a petrogenic origin source (Wilcke 2007). The study found that the average concentrations of ∑LMW-PAHs and ∑HMW-PAHs were 30.74±25.49 µg/kg

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(range: BDL-44.50 µg/kg) and BDL±21.1 µg/kg (range: 105-71.59 µg/kg), respectively, accounting for 16.59% and 83.42% of the total PAHs, indicating mixed origins in Pune (Table 1).

However, pyrogenic processes are also identified as major sources of PAHs in Pune due to the relatively higher fraction of HMW-PAHs and the resulting low ratio of LMW-PAHs to HMW-PAHs (<1.0). Because HMW-PAHs are adsorbed to the particles and have a propensity for fast deposition, their dominance can be explained. In urban settings, industrial and transportation emissions are typically regarded as a significant source of HMW-PAHs (ATSDR 1995; Kasaraneni and Vinka 2016). Vehicle emissions, construction activities, diesel generators, power plants, factories, and biomass burning are among the several pyrogenic sources of the majority of Delhi's air particulates, in addition to long-distance atmospheric transport of pollutants (Hama et al. 2020).

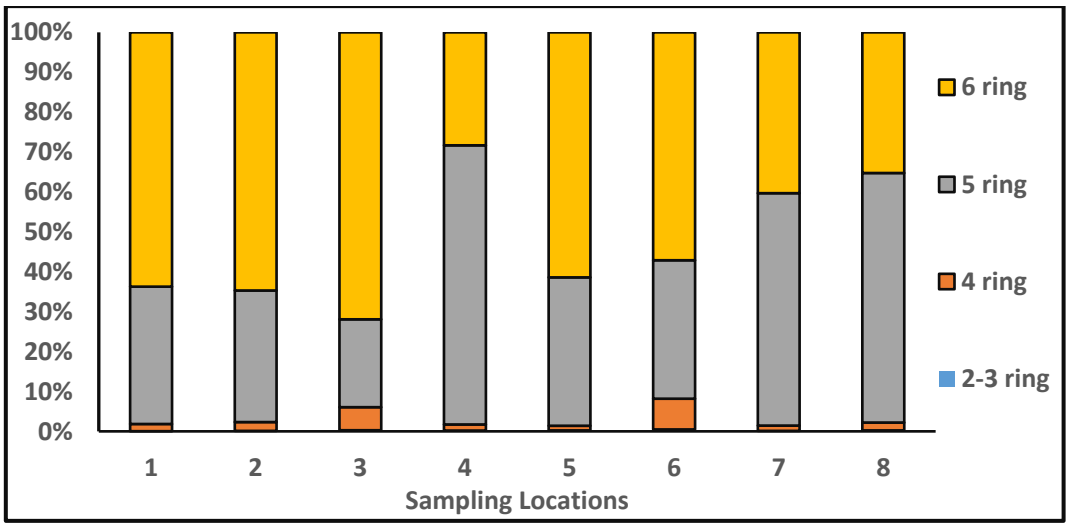

Figure: 2 Contribution of PAHs with different rings to ∑BaPTE

Diagnostic Ratios of Selected PAHs

To identify potential sources of PAHs in Delhi soils, molecular diagnostic ratios of specific PAH concentrations were also computed. For this investigation, specific molecular ratios of PAHs were computed, such as FLT/(FLT+PYR), BaA/(BaA+CHR), BbF/BkF, BaP/BghiP, BaP/(BaP+Chr), and IndP/(IndP+BghiP). Diesel, gasoline, wood, vehicle, and coal combustion emissions are just a few of the sources of PAHs that have been characterized using these molecular ratios (Yunker et al. 2002; Lee et al. 1995; Dickhut et al. 2000; Simcik et al. 1999; Chen et al. 2005; Khalili et al. 1995).

Table 3: Diagnostic ratio of PAHs for possible sources identification

Diagnostic ratios with their reported values for possible sources				
PAH ratio	Value	Possible sources	Reference	All locations
LMW/HMW	<1	Pyrolytic	Wilcke, 2007	0.66 (0.45-0.88)
	>1	Petrogenic		
	>0.1	Petroleum, biomass combustion		
FLT/(FLT+PYR)	<0.4	Petrogenic	Yunker et al. 2002	0.36 (0.23-0.46)
	0.4-0.5	Fossil fuel combustion		
	<1.0	Gasoline, diesel engine	Lee et al. 1995	
BaA/(BaA+CHR)	0.2-0.35	Petroleum combustion	Yunker et al. 2002	2.32 (0.30-6.42)
	>0.35	Biomass, coal combustion.		
BbF/BkF	1.30	Vehicular emission	Dickhut et al. 2000	3.51 (0.33-8.50)
	3.7	Coal combustion		

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BaP/BghiP	0.3 -0.78	Vehicular emissions	Simcik et al. 1999	1.30 (0.12-4.38)
	0.9-6.6	Coal combustion		
BaP/(BaP+CHR)	0.49	Gasoline	Khalili et al. 1995	0.68 (0.51-0.87)
	0.73	Diesel engine		
IPY/(IPY+BghiP)	<0.2	Petrogenic	Dickhut et al. 2000 Yunker et al. 2002	0.34 (0.00-0.77)
	0.2-0.5	Petroleum combustion		
	>0.5	Biomass, coal combustion		

*Range in parenthesis

According to Table 3, the average ratios of FLT/(FLT+PYR), BaA/(BaA+CHR), BbF/BkF, BaP/BghiP, BaP/(BaP+Chr), and IndP/(IndP+BghiP) for this study were 0.66, 0.36, 2.32, 1.30, 0.68, and 0.34, respectively. These values indicated mixed pyrogenic sources of PAH. According to Kaur et al. (2013), a BaA/Chr concentration ratio more than 0.40 suggests recent emissions and comparatively poor photochemical degradation, whereas a ratio less than 0.40 suggests the migration of older sources of PAHs.

The present study's measured BaA/Chr concentration ratio ranged from 0.30 to 6.42, suggesting that fresh and older air masses were transported onto Pune soils together with recent PAH emissions. Based on these findings of potential source identification, the study proposed that gasoline, coal, wood and biomass combustion, and mixed pyrolytic sources from diesel engine emissions are the main sources of PAHs in Pune soils (Figure 5, Table S5 and Table S6). However, it is impossible to ignore out petrogenic sources such car workshops and unintentional spills.

Pearson's Correlation Coefficient

Correlation analysis was performed for the assumption that two or more PAHs might be associated because of a shared origin, correlation analysis was carried out. Individual PAH relationships were calculated using Pearson's moment correlation coefficients, and the results are shown in Table S7. This study found a significant association (two tailed, $p<0.01$, $p<0.001$) between the LMW-PAHs and HMW-PAHs. Low temperature biomass combustion sources are linked to a notably strong correlation between LMW-PAHs like ANY, ANE, FLE, PHE, and ANT. FLE, PHE, ANT, CHR, and PYR all showed a strong association with the sources of biomass combustion. Significant relationships between FLT, BbF, PYR, BaA, CHR, BkF, BaP, BghiP, DBA, and IndP indicated high temperature combustion processes, including those in industries, automobiles, and coal combustion sources of emissions (Khalili et al. 1995; Marret al. 1999; Wilcke 2007).

Table 4: Pearson's moment correlation coefficient among PAHs

	FLE	PHE	ANT	FLT	PYR	BaA	CHR	BbF	BkF	BaP	BghiP	DBA	IPY
ANE	**0.63**	-0.46	0.00	0.16	0.00	-0.06	-0.18	**0.56**	-0.44	0.49	0.23	0.01	0.21
FLE		-0.50	**0.55**	**0.54**	0.22	0.19	0.15	**0.82**	-0.30	**0.83**	-0.32	0.26	0.01
PHE			-0.15	0.09	0.50	0.18	0.16	-0.45	**0.58**	-0.07	0.51	0.23	0.05
ANT				0.43	**0.55**	**0.60**	-0.06	**0.56**	-0.28	0.39	-0.19	-0.16	-0.28
FLT					**0.73**	0.38	**0.57**	**0.59**	**0.55**	**0.67**	-0.28	0.27	0.16
PYR						0.38	0.38	0.21	0.44	**0.54**	0.31	0.02	-0.21
BaA							-0.41	0.47	-0.17	-0.03	-0.17	0.06	0.51
CHR								0.03	**0.77**	0.48	-0.21	0.13	-0.30
BbF									-0.36	0.50	-0.32	-0.11	0.31
BkF										0.14	-0.06	0.31	-0.01
BaP											-0.03	0.43	-0.20
BghiP												-0.25	-0.23
DBA													0.29

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Significant correlations at $p<0.1$ are in bold

A significant correlation of BaA with BaP, BghiP and IndP has been reported for stationary source emissions (Kaur et al. 2013). Stationary sources have been identified as the source of industrial and coal combustion emissions near the urban region. These findings showed that PAHs in soils come from a variety of sources. Thus, the study came to the conclusion that the most important sources of PAHs in the soils of megacity Delhi might be pyrogenic sources, such as the burning of biomass and coal as well as vehicle emissions. According to Khanna et al. (2018), the primary sources of organic matter and elemental carbon in Delhi's environment are combustion activities. Similar PAH sources have been documented for a number of Delhi region environmental matrices (Khillare 2018; Hazarika & Srivastava 2016; Kumar et al.2015a; Jyethi et al. 2014; Agarwal et al. 2009).

Human Health Risk and Environmental Health Hazard Assessment

Human Health Risk

Human health risk assessment was predicated on the idea that both adults and children might come into contact with soil contaminated with PAHs. The LADD for adults and children resulting from 16PAHs in soil was calculated in order to determine the ILCR (Table 3, Table 4, and Table S8 – Table S11). Adult humans exposed to 16PAHs through soil had an average LADD of 4.59x10-8 mg kg-1 d-1, while children had an average LADD of 1.71x10-7 mg kg-1 d-1 (Table 3). The main cause of the overall average LADD for both adults and children was ∑HMW-PAHs. For human adults and children, the average LADD of ∑HMW-PAHs was 4.23x10-9 mg kg-1 d-1 and 2.73x10-7 mg kg-1 d-1, respectively.

Table5: LADD (mg/kg/d) and ILCR for Children due to PAHs in soil

PAHs	LADD			ILCR		
	Min	Max	Mean	Min	Max	Mean
Naphthalene	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
Acenaphthylene	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
Acenaphthene	0.00E+00	6.23E-11	1.54E-11	0.00E+00	8.53E-09	2.11E-09
Fluorene	0.00E+00	2.54E-10	4.94E-11	0.00E+00	3.48E-08	6.76E-09
Phenanthrene	2.51E-11	1.20E-10	5.54E-11	3.44E-09	1.64E-08	7.59E-09
Anthracene	0.00E+00	4.34E-10	2.20E-10	0.00E+00	5.95E-09	3.01E-09
Fluoranthene	1.35E-11	6.80E-11	3.95E-11	1.85E-09	9.32E-09	5.42E-09
Pyrene	2.23E-11	1.41E-10	7.82E-11	3.05E-09	1.93E-08	1.07E-08
Benzo(a)Anthracene	1.40E-09	7.49E-09	3.75E-09	1.92E-09	1.03E-08	5.13E-09
Chrysene	8.57E-11	5.09E-10	2.64E-10	1.17E-09	6.97E-09	3.61E-09
Benzo(b)Fluoranthene	2.51E-09	1.54E-08	7.14E-09	3.44E-09	2.11E-08	9.79E-09
Benzo(k)Fluoranthene	8.00E-10	9.71E-09	4.35E-09	1.10E-09	1.33E-08	5.96E-09
Benzo(a)Pyrene	1.54E-08	2.39E-07	7.26E-08	2.11E-09	3.27E-08	9.95E-09
Benzo(g,h,i)Perylene	2.06E-10	2.21E-09	9.67E-10	2.82E-09	3.02E-08	1.32E-08
Dibenzo(a,h)Anthracene	3.66E-08	1.21E-07	7.70E-08	5.01E-09	1.65E-08	1.05E-08
Indeno(1,2,3-Cd)Pyrene	0.00E+00	7.60E-09	3.96E-09	0.00E+00	1.04E-08	5.42E-09
∑PAHs	9.75E-08	3.67E-07	1.71E-07	5.31E-08	1.75E-07	9.93E-08
2-3 ring	6.74E-11	7.76E-10	3.40E-10	9.24E-09	5.28E-08	1.95E-08
4 ring	1.59E-09	7.81E-09	4.13E-09	8.93E-09	3.81E-08	2.49E-08
5 ring	2.73E-08	2.57E-07	8.41E-08	1.10E-08	5.78E-08	2.57E-08
6 ring	3.89E-08	1.28E-07	8.19E-08	1.29E-08	4.46E-08	2.92E-08

Table6: LADD (mg/kg/d) and ILCR for Human adults due to PAHs in soil

PAHs	LADD	ILCR

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	Min	Max	Mean	Min	Max	Mean
Naphthalene	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
Acenaphthylene	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
Acenaphthene	0.00E+00	1.68E-11	4.15E-12	0.00E+00	2.30E-09	5.69E-10
Fluorene	0.00E+00	6.85E-11	1.33E-11	0.00E+00	9.38E-09	1.82E-09
Phenanthrene	6.77E-12	3.23E-11	1.49E-11	9.27E-10	4.43E-09	2.04E-09
Anthracene	0.00E+00	1.17E-10	5.92E-11	0.00E+00	1.60E-09	8.11E-10
Fluoranthene	3.63E-12	1.83E-11	1.06E-11	4.97E-10	2.51E-09	1.46E-09
Pyrene	6.00E-12	3.80E-11	2.11E-11	8.22E-10	5.21E-09	2.88E-09
Benzo(a)Anthracene	3.77E-10	2.02E-09	1.01E-09	5.16E-10	2.76E-09	1.38E-09
Chrysene	2.31E-11	1.37E-10	7.10E-11	3.16E-10	1.88E-09	9.72E-10
Benzo(b)Fluoranthene	6.77E-10	4.14E-09	1.92E-09	9.27E-10	5.67E-09	2.63E-09
Benzo(k)Fluoranthene	2.15E-10	2.62E-09	1.17E-09	2.95E-10	3.58E-09	1.60E-09
Benzo(a)Pyrene	4.15E-09	6.43E-08	1.96E-08	5.69E-10	8.81E-09	2.68E-09
Benzo(g,h,i)Perylene	5.54E-11	5.94E-10	2.60E-10	7.59E-10	8.13E-09	3.57E-09
Dibenzo(a,h)Anthracene	9.85E-09	3.25E-08	2.07E-08	1.35E-09	4.45E-09	2.84E-09
Indeno(1,2,3-Cd)Pyrene	0.00E+00	2.05E-09	1.07E-09	0.00E+00	2.80E-09	1.46E-09
∑PAHs	2.62E-08	9.89E-08	4.59E-08	1.43E-08	4.71E-08	2.67E-08
2-3 ring	1.82E-11	2.09E-10	9.16E-11	2.49E-09	1.42E-08	5.24E-09
4 ring	4.28E-10	2.10E-09	1.11E-09	2.40E-09	1.03E-08	6.70E-09
5 ring	7.36E-09	6.92E-08	2.27E-08	2.97E-09	1.56E-08	6.92E-09
6 ring	1.05E-08	3.44E-08	2.21E-08	3.46E-09	1.20E-08	7.87E-09

The average LADD for adults and children through soil was found to be below the permissible limits. Based on oral trials of BaP, the suggested index dose for an adult weighing 70 kg is 0.02 µg kg-1 d-1, or 2.0x10-5 mg kg-1 d-1 (Environment agency 2002). Additionally, USEPA calculated an oral slope factor of 7.3x10-3 per µg BaP kg-1 bw day-1, meaning that consuming 1 µg BaP kg-1 bw day-1 would result in a 7.3×10-3 lifetime cancer risk (USEPA 2019).

Environmental and Human Health Hazard

The National Oceanic and Atmospheric Administration's (NOAA) soil quality guidelines (SQGs) for mammal protection and the Canadian Council of Ministers of the Environment's (CCME) SQGs for environmental and human health protection were used to evaluate the risk of PAHs' non-carcinogenic effects on environmental health (CCME 2010; Buckman 2008). In soils at various locations in Pune, the observed levels of ∑16PAHs (range: 68-224 µg/kg, mean: 127±48 µg/kg) and their individual compounds (1-44 µg/kg) were lower than the SQGs for residential and park soils (13-50x103 µg/kg) (CCME 2010) and for individual PAH compounds for mammals (99.4-1.48x106 µg/kg) (Buckman 2008). The mean concentrations of LMW-PAHs and HMW-PAHs were 30 µg/kg and 21 µg/kg, respectively, with observed quantities ranging from 1 to 44 µg/kg and 71 to 105 µg/kg. According to Buckman (2008), these were also less than SQGs for HMW-PAHs (1100 µg/kg) and LMW-PAHs (100x103 µg/kg). The study's findings indicated a low risk to human health and the environment.

Conclusion

In order to evaluate the composition profiles, potential sources, and health impacts on both the environment and people, this study was conducted on PAHs in soils from Pune, India's urban and rural areas. Low environmental health hazard was suggested by the fact that levels of ∑PAHs in soils from rural areas were lower than those in urban areas and below the SQGs for mammals and different land uses. Weekly polluted soils with PAHs were suggested by the Nemerow Composite Index (NCI) and soil classification, although they were placed in the safe category. The dominant PAH compounds in

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soils were Fluoranthene, Pyrene, Phenanthrene, Benzo(a)Pyrene, Diabenzo(a,h)Anthracene, Benzo(b)Fluoranthene, Benzo(k)Fluoranthene and Benzo(g,h,i)Perylene, and their combined contribution accounted for 74% to ∑16PAHs. Study indicated mixed pyrolytic sources from vehicular emissions from diesel engines; coal, wood & biomass combustions, and gasoline are the significant contributor of PAHs to soils in Pune.

LADD (lifetime average daily dose) of PAHs in soil for both adults and children, with different amounts and sources of PAHs depending on the area. The primary cause of both adults' and children's overall average LADD was ∑HMW-PAHs. Adults and children's estimated LADD and ILCR due to PAHs in soil were within the permissible limits. For the risk to human health from the carcinogenic effects of PAHs, the index of additive cancer risk (IACR), BaP toxicity equivalent (BaPTE), and BaP total potency equivalent (BaP TPE) were all below recommended levels.

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