

Distribution of PAHs in Surface Soils from Petroleum Handling Facilities in Calabar

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Abstract The level of concentrations of polycyclic aromatic hydrocarbons (PAHs) in surface soils from petroleum handling facilities (kerosene tank, generating plant, petrol stations, mechanic workshops, leaking pipeline and air port fuel dump) from Calabar metropolis southeastern Nigeria was determined by gas chromatography/ mass spectrometry. The results show that total polycyclic aromatic hydrocarbons (PAHs) varied from 1.80 to 334.43 mg/kg with a mean of 50.31 mg/kg. The lowest value of 1.80 mg/kg was obtained from petrol station while the highest value of 334.43 mg/kg was obtained from facility characterised by petrol stations and mechanic workshops. The ratio of phenanthrene/anthracene and fluoranthene/pyrene, varied from 0.43 to 27.72 and from 0.14 to 17.76 respectively. These ratios indicate various sources for the PAH. The two to three ring PAHs are the most abundant. Based on the PAH ratios and content alone it is not possible to distinguish between contribution from motor vehicle exhaust, gasoline spillage, used engine oil or petroleum production. However, considering the area of the study, it is very likely that the major source of soil contamination is originating from petroleum product.

Keywords PAHs · Soil · Petroleum · Contamination · Petrogenic · Pyrolytic · Calabar

1 Introduction

Polycyclic aromatic hydrocarbons (PAHs) are common contaminants of concern at petroleum contamination sites. They are widespread in the environment from both natural and anthropogenic sources (IARC, 1983; Menzie, Potocki, & Santodonato, 1992). Background concentrations of PAHs compounds resulting from atmospheric deposition may be significantly elevated in some surface soils, particularly in urban areas and along roadways. The main source of PAHs include incomplete combustion of organic material, untreated diesel, exhaust gas and waste oils (Bernes, 1996).

In soils and sediments PAHs are generally immobile because of their low water solubility (Rippen, 1992). In spite of the low solubility of PAHs, leaching and transformation in soils are obvious (Tebaay, Welp, & Brummer, 1993). Accumulation of PAHs in soils however may lead to future, contamination of groundwater and food chains. PAHs that enter the water body via gas exchange may partition into the organic rich particles (e.g., plankton) which can be ingested and may settle out quickly of the water column as faecal pellets or be recycled (ASTDR, 1995).

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Table 1 Concentrations of PAHs (mg/kg) and (%) TOC in surface soil

Facility ^a	Station no.	Nap	Any	Ace	Fln	Phe	Ant	Flu	Py	Ba(A)	Chr	BbF–BkF	BaP	Indeno	DahA	Bghip	Total	%TOC
A	1	1.34	0.04	0.15	0.35	0.36	0.02	0.06	0.11	0.03	0.04	0.07	0.04	0.27	0.01	0.27	3.14	2.3
	2	1.63	0.05	0.21	0.44	0.57	0.04	0.43	0.49	0.29	0.29	0.94	0.48	1.18	0.11	0.78	7.92	0.64
B	3	134.08	1.03	6.27	13.72	75.76	6.014	24.24	48.03	5.07	9.79	4.46	0.93	0.77	0	4.36	334.43	8.5
	4	3.38	0.06	0.55	1.10	0.83	0.10	0.23	0.35	0.90	1.12	0.15	0.06	0.22	0	0.02	8.26	2.1
C	5	67.71	0.48	3.68	8.23	40.21	3.05	15.16	29.11	2.56	4.04	2.01	1.57	3.01	0	0.16	188.13	6.1
	6	3.61	0.09	1.61	1.96	2.19	0.079	0.61	0.36	0.18	0.013	0.42	0.024	0.083	0.017	0.14	15.01	4.2
	7	12.53	9.83	3.59	5.72	7.89	0.45	2.59	2.64	0.31	1.54	0.07	0.01	0.002	0.002	0.043	47.22	5.2
D	8	5.41	0.85	0.37	0.99	2.34	0.18	1.12	2.14	0.45	0.13	1.42	1.14	1.11	0.07	1.58	19.3	3.5
	9	28.48	0.15	0.32	1.07	6.28	1.18	1.84	3.17	0.90	0.14	1.12	1.13	1.89	0.02	3.16	49.85	2.1
	10	30.85	0.20	0.65	2.70	8.23	0.76	2.50	4.28	1.57	0.89	2.11	1.67	3.64	0.16	6.61	66.82	4.5
E	11	2.87	0.57	0.41	1.02	1.76	0.25	1.10	2.01	0.89	0.57	1.06	1.21	1.31	0.11	1.15	17.31	1.21
	12	17.49	0.35	1.18	1.21	4.27	0.62	1.19	2.46	0.65	0.31	1.79	1.45	2.05	0.24	2.89	38.15	3.2
	13	57.80	0.38	3.35	13.41	35.59	2.63	11.60	23.03	6.10	3.24	5.76	4.37	3.59	0.33	2.48	173.72	6.21
E	14	1.12	0.45	0.23	0.31	2.14	0.95	0.35	0.27	0.38	0.14	0.41	0.05	0.01	0.001	0.10	7.56	2.3
	15	0.61	0.43	0	0.78	1.24	1.04	0.48	0.55	0.66	0.24	0.62	0.03	0.02	0.02	0.03	6.13	7.3
	16	1.71	0.10	0.71	4.62	1.76	2.39	0.57	1.12	0.25	1.75	0.06	0.18	0.001	0.001	0.004	15.14	2.1
E	17	1.21	0.02	0.53	0.61	3.81	0.30	0.77	2.37	0.35	0.64	0.36	0.68	0.12	0.55	0.13	12.45	18
	18	1.15	0.12	0.24	1.10	1.34	1.05	0.35	1.10	0.15	1.25	0.04	0.14	0.01	0.04	0.015	8.10	1.8
	19	1.65	0.23	0.11	0.56	0.61	0.21	0.03	0.21	0.08	0.03	0.09	0.07	0.38	0.10	0.54	4.7	0.85
E	20	0.07	0.11	0.001	0.48	0.25	0.58	0.005	0.15	0.12	0.02	0.001	0.004	0.001	0.002	0.001	1.80	0.34
	Ap	0.11	0.32	7.89	38.46	5.94	3.88	3.02	0.17	0.04	0.13	0.65	0.03	0.05	0.02	0.07	60.65	3.5
	Sp	0.81	0.02	0.20	0.49	5.04	0.63	2.30	6.86	1.07	1.39	1.26	0.46	0.55	0	0	21.08	3.1

^a Kerosene tank

B Generating plant

C Petrol station/mechanic workshop

D Mechanic workshop

E Petrol station

Available data on the PAH concentrations in urban soils focus on temperate cities. Knowledge on the PAH burden of soils in tropical cities is scarce (Wilcke et al., 1999). The study of Smith, Edelhofer, and Harrison (1995), showed that the PAH concentrations in urban soils of Lahore (Pakistan) are lower than in Birmingham (UK) although atmospheric concentrations are higher. PAHs concentrations from agricultural and forest soils in Ghana (57–54 $\mu\text{g}/\text{kg}$) suggest that background concentrations in tropical soils are lower than in temperate ones. Natural background concentrations of PAH in soils estimated by Edwards (1983), range between 1 and 10 $\mu\text{g}/\text{kg}$ for individual PAHs. PAHs concentrations in forest soils are much higher than in agricultural soils. This is due to the intercept effect of the leaves and the high carbon/organic content. High accumulation of PAHs is observed in the humus layer of forest soils (Wilcke, Muller, Kanchanakool, & Zech, 1998).

In Calabar the fast economic growth of the last decades resulted in obvious environmental problems, among them air pollution caused by the enormous daily traffic. This results in PAH emission and subsequent deposition. Between 1980 and today the number of petroleum handling facilities in Calabar metropolis have increased by more than 10-folds, thus leading to indiscriminate spillage and disposal of the petroleum products. In addition the waste generated by these facilities especially the used engine oils, spent lubricating oils, fuel additives and grease are disposed off on the ground surface, buried underground or dumped in drains, gutters and pits where no special controls are provided.

The present study is a preliminary assessment of the level of PAH in surface soils from petroleum handling facilities in Calabar metropolis.

2 Study Area

The study area Calabar (Southeastern Nigeria) the capital of Cross River State is situated on an area of about 200 km^2 with a population of about 300,000. It lies between latitude $4^{\circ}15'–5^{\circ}\text{N}$ and longitude $8^{\circ}15'–8^{\circ}25'$ (Figure 1). Two rivers drain the area namely Cross River and the Great Kwa River. The area falls within the semi equatorial climatic zone of Nigeria. It is characterized by mean annual rainfall in the range of 1,000–2,000 mm and mean monthly temperature of about 29°C (Iloje, 1991).

The area is underlain by tertiary-recent sediments known as Benin formation. The lithology is marked by lacustrine and fluvial sands, pebbles, clay lignite and alluvium.

3 Materials and Methods

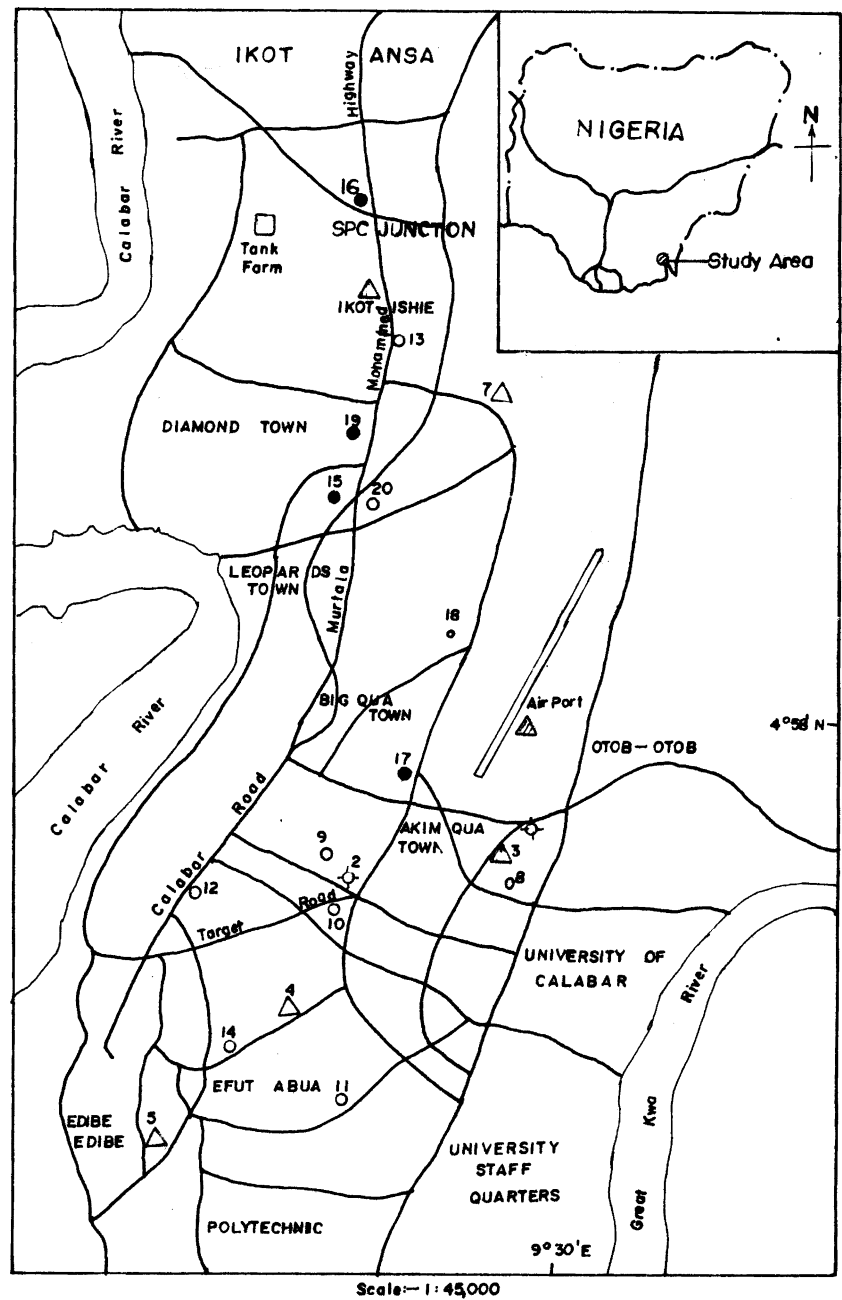
Surface soil samples were collected from various facilities such as kerosene tank (A), generating plant (B), mechanic workshops (D), petrol stations (E), airport fuel dump (Ap) and leaking pipeline (Sp). The locations designated (C) were characterized by mechanic workshops/petrol stations. The sample locations are shown in Figure 1, surface soil samples (0–15 cm) were taken with a stainless steel spatula from these facilities. The samples were wrapped with aluminium foils which were cleaned with toluene, before being transferred to the laboratory. In the laboratory the samples were air dried, crushed and sieved using 230 mesh sieves. After sieving they were immediately wrapped in aluminium foil and kept in a plastic bag until extraction.

Total organic carbon in the soil was determined using Walkley and Black (1934), titration method and Perkin-Elmer 240C elemental analyzer as describe by Yang, Liu, and Zhang (1998). The analytical results are presented in Table I. 15 g of the soil sample was extracted using accelerated solvent extractor (ASE C 300) Dionex, sunnyvale.

The extracts were then purified in a silica gel column using 10 g of activated silica gel to remove organic polymers, aliphatic and polar compounds.

Subsequently, the extracts were concentrated to a small volume (<5 ml) by rotary evaporation, and then further reduced to 1 ml by nitrogen gas. Prior to purification 5 μl of deuterated PAH internal standards namely deuterated naphthalene (Nap- D_8), deuterated acenaphthene (Ace- D_{10}), deuterated Phenanthrene, (Phe- D_{10}), deuterated chrysene (Chr- D_{12}) and deuterated Perylene (Perylene- D_{12}) with concentration of 187.34 mg/l . was added to the extracts. The extracts were analysed for naphthalene (Nap), acenaphthylene (Any), acenaphthene (Ace), fluorene (Flu), phenanthrene (Phe), anthracene (Ant), fluoranthene (Flu), pyrene (Py), benzo (a) anthracene (B(a)A), chrysene (Chr), benzo (b) fluoranthene (B(b)F), benzo (k) fluoranthene (B(k)F), benzo (a) pyrene (Bap), dibenzo (a, h) anthracene (DabA), Ideno (1,2,3-d) pyrene

Figure 1 Map of study area including sample locations.



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| | Keronsene Tank | A |
| | Generating Plant | B |
| | Mechanic Workshop/Petrol Station | C |
| | Mechanic Workshop | D |
| | Petrol Station | E |
| | Spilled Site | SP |
| | Airport Fuel Dump | AP |
| | Meteorological Station | |

(Ideno) and benzo (g, h, 1) perylene (Bghip) with gas chromatography coupled to mass spectrometry (HP5890/HP5972) autosampler HP 7673 and software HPG1304C at the hydrogeochemistry laboratory, University of Tubingen (Germany). A fused-silica HP-5 capillary column (30 m×0.25 mm, 0.25 µm film thickness) was used with helium as the carrier gas. A microliter volume was injected by the sampler by applying a hot splitless injection technique with a venting time of 1.5 min. The temperature program of the oven was started at 43°C for 5 min with initial flow of 1.5 ml/min and increased to 65–270°C. The total run time was 42 min. The mass spectrometer was operated in the electron impact (E1) mode with an ion source at 270°C and an electron voltage of 70 eV. Identification of the PAH compounds was performed by comparing GC retention time with those of authentic standards. Quantification of individual compound was based on comparison of peak areas with those of the recovery standards.

4 Results and Discussion

Concentrations of PAHs for the surface soil samples are presented in Table I. The total PAHs ranged from 1.80 to 334.43 mg/kg with a mean of 50.31 mg/kg. The lowest value of 1.80 mg/kg was obtained from facility E (location 20) and highest value of 334.43 mg/kg was obtained from facility C (location 3). The results show that the concentration levels of PAHs vary widely between sampling sites. It is observed that the most contaminated samples were from facilities C and D. The level of ΣPAH concentration is in the order C>D>AP>E>SP>B>A. The highest concentration of PAHs recorded from facilities C and D may be due to the volume of waste generated from these facilities which in most cases are disposed off directly on the ground surface. According to Ineson (1973), increased use of these petroleum products increases the potential pollution risk. The total PAH obtained at location 3 (334.43 mg/kg), location 5 (188.13 mg/kg), location 10 (66.82 mg/kg), location 13 (173.72 mg/kg) and Sp (60.65 mg/kg) exceeded that of the total mean (50.3 mg/kg). This is attributed to the volume of waste generated and the amount of product spilled on the surface soil. Soils are important sinks of atmospheric PAHs (Jones et al., 1989) and due to their low solubility they tend to accumulate in soil (Krauss,

Wilcke, & Zech, 2000). Soils of tropical environments have lower concentrations of PAHs than soils of temperate cities. This is attributed to the shorter accumulation time of the PAHs due to the tropical climate which enhances biodegradation, volatilisation, photo-oxidation and leaching into the groundwater (Wilcke, Muller, Kanchanakool, & Zech, 1999). Considering the individual PAHs in the surface soils of the study area, the order is Nap (33.94%)>Phe (18.83%)>Py (11.83%)>Flu (6.38%)>Ace (4.40%)>Chr (2.50%)>Ant (2.39%)>BbF–BkF (2.25%)>Bghip (2.22%)>Ba(A) (2.08%)>Ideno (1.87%)>Bap (1.42%)>Any (1.37%)>DahA (0.16%). In terms of ring type the two and three ringed PAH concentration is 128.96 mg./kg and 63.02 mg/kg respectively for the four and five ringed PAHs the mean concentrations are 20.44 mg/kg and 13.17 mg/kg in the study area. The PAH distribution pattern indicates the predominance of two and three ringed components making up to 69.93% while the others account for 30.07% thus the order is 3 ringed>2 ringed>4 ringed>5 ringed>6 ringed. The pattern of distribution of PAH abundance suggests a petroleum derived source in the surface soil of the study area. Naphthalene often constitutes a significant fraction of crude oils and petroleum products with higher lighter fractions, and hence might be used to detect PAH input from these sources and also exhaust from motor vehicles (Sporstol et al., 1983). The PAH concentrations in surface soils from the petroleum handling facilities in the study area is higher than those obtained by Wilcke et al. (1999) in the hydromorphic soils of Bangkok (12–380 µg/kg) and those obtained from agricultural and forest soils in Ghana (57–74 µg/kg; Smith et al., 1995). Spillage of petroleum and its refined products, both of which contain extreme complex mixtures of aromatic hydrocarbons (Youngblood & Blumer, 1975; Wakeham, 1977) may add to anthropogenic contribution of PAH, thus the spillage from these facilities significantly contribute to this high level of PAHs in the study area.

The processes controlling the level of PAHs in the soil are complex. Among important factors are chemical properties (especially their water solubilities), the composition of the soils such as organic carbon and clay content (Yang et al., 1998). The effect of soil organic carbon on the PAH concentrations was determined. Values obtained for percent organic carbon are presented in Table I. The organic carbon ranged from 0.34% (location 20) to 8.5% (location 3),

high total PAH concentration also correspond to high percent organic carbon (location 3 facility C). Soil organic matter is known to be most important sorbent of PAHs (Sims & Overcash, 1983).

The ratio values of phenanthrene / anthracene (Phe/Ant) and fluoranthene (Flu/Py) have been used to determine the sources of PAHs (Sicre et al., 1987; Budzinski, Jones, Bellocq, Pierard, & Garrigues, 1997; Soclo, Garrigues, & Ewald, 2000). According to Wise et al. (1988) and Benner, Gordon, and Wise (1989); Benner et al. (1990) petroleum often contain more phenanthrene relative to anthracene, thus a Phen/Ant ratio is observed to be very high in PAH of petrogenic pollution, but low in pyrolytic contamination cases (Gschwend & Hites 1981; Soclo et al., 2000). However, Gschwend and Hites (1981), Colombo, Pelletier, Brochu, Khalil, and Catoggio (1989) and Budzinski et al. (1997) suggested that the Phe/Ant ratio >10 and <10 , represented petrogenic and pyrolytic contamination while Yang (2000) and Liu et al. (2001), also attributed that the Phe/Ant ratio >15 and <15 , represented petrogenic and pyrolytic contamination respectively.

The ratio of Phe/Ant for this study varied from 0.43 to 27.72 (Table II) indicating different sources of PAH contamination. Most locations characterised mostly by mechanic workshops i.e., locations 1, 2, 3, 5, 6, 7, 8, 10 and 13, have Phe/Ant >10 indicating petrogenic input. Locations 14–20 (Petrol stations), SP and AP have Phe/Ant values <10 indicating pyrolytic input. Phe/Ant ratio may not only reflect the origin of the contaminant, but also the age of sample storage conditions, extraction procedure, etc. The older samples subjected to atmospheric weathering and UV light will show higher Phe/Ant ratio.

The Phe/Ant ratios for motor vehicle exhaust are in the range of 4.0 to 10.0 (Yang, Connell, Hawker, & Kayal, 1991). Locations 4, 9, 11, 12 (mostly mechanic workshops) and AP, have Phe/Ant ratio in this range. Motor vehicle exhaust contains PAH from both unburned fuel and pyrolytic processes, but the unburned fuel contribution is predominant.

Sicre et al. (1987) also suggested that Flu/Py ratio of <1 was attributed to petrogenic sources and values >1 to pyrolytic origin. Table II, shows that with the exception of locations 6, 14 and SP the ratio of Flu/py less than 1 indicating petrogenic inputs probably from oil contamination from these facilities. The ratios of Phe/Ant >10 and Flu/py <1 , represent

Table II Values of Phe/Ant and Fluo/Py ration for the surface soils

Station No.	Phe/Ant	Flu/Pyr
1	18	0.54
2	14.25	0.88
3	13.0	0.51
4	8.3	0.65
5	13.18	0.52
6	27.72	1.69
7	17.53	0.98
8	13	0.52
9	5.32	0.58
10	10.82	0.58
11	7.04	0.55
12	6.89	0.48
13	13.53	0.51
14	2.25	1.30
15	1.19	0.87
16	0.74	0.52
17	1.28	0.31
18	1.28	0.31
19	2.90	0.14
20	0.43	0.03
SP	1.53	17.76
AP	8.00	0.34

petrogenic origin and Phe/Ant <10 and Flu/Py >1 represent pyrolytic origin (Gschwend & Hites, 1981; Colombo et al., 1989). According to Yang (2000) and Liu et al. (2001), Phe/Ant ratio >15 and Flu/Py ratio <1 indicate petrogenic origin, while Phe/Ant ratio <15 and Flu/Py ratio >1 represent pyrolytic origin. From the above therefore, location 1 revealed a higher Phe/Ant ratio (18) and lower Flu/Py ratio (0.54) suggesting strong petrogenic input rather than pyrolytic. The high Flu/Py ratio at Sp depicting pyrolytic origin instead of petrogenic may be attributed to fire outbreak as vandals attempt to scooped the fuel from the leaking pipeline. Considering the area of study, it is very likely that the major source of soil contamination is originating from petroleum products. However, since the mechanic workshop area is showing one of the highest concentrations of PAH, it is possible that the exhaust from the equipment used in the facility is the important source of PAH at these locations. Thus based on the variations on the PAH ratios and content alone, it is not possible to distinguish between contribution from motor vehicle exhaust, gasoline spillage, used engine oils or petroleum production.

5 Conclusion

The distribution of PAHs in soils of the study areas from petroleum handling facilities were investigated. Petrol station/mechanic workshop show the highest level of concentration of total PAHs. The concentration of total PAHs is in the order petrol station/mechanic workshop>mechanic workshops>airport fuel dump>petrol station>leaking pipeline>generating plant>kerosene tank. Naphthalene, phenanthrene, pyrene and fluoranthene are the dominant PAH compounds. The two–three rings PAH are the most prominent. The abundance of two–three ring PAHs does not necessary indicate petroleum production input, but rather the impact of motor vehicle exhaust and other combustion sources. Motor vehicle exhausts contain PAH from both unburned fuel and pyrolytic processes, but the unburned fuel contribution is predominant. Taking into account the area of the study, it is very likely that the major source of soil contamination is originating from petroleum products. However, since the mechanic workshop area is showing one of the highest concentrations of PAH, it is possible that the exhaust from the equipment used in the facility is the important source of PAH at these locations. Thus, based on the PAH ratios and content alone, it is not possible to distinguish between contribution from motor vehicle exhaust, gasoline spillage, used engine oils or petroleum production. Due to the potential carcinogenic and mutagenic characteristics and potential exposure to humans periodic monitoring of persistent organic pollutants (PAHs) should be instituted as an indicator of environmental quality in the study area.

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