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Distribution of Polycyclic Aromatic Hydrocarbons and Heavy Metals in Soils from Municipal Solid Waste Landfill

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Authors' contributions

This work was carried out in collaboration between all authors. Author JCI conceived, designed, wrote the protocol, managed the litterature searches, and first draft of the manuscript. Authors SCU and CBL collected the sample, processed it and performed the Polycyclic Aromatic Hydrocarbons (PAHs) analysis. Author AUO performed the heavy metal analysis. Author CE performed the statistical analysis and formatted the final manuscript. All authors read, corrected, and approved the final manuscript.

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ABSTRACT

Aims: The distribution of polycyclic aromatic hydrocarbons (PAHs) and heavy metals in soils from Onitsha municipal solid waste landfill were studied.
Study Design: The Onitsha solid waste dumpsite were divided into two groups, the active site labelled (A), where waste dumping is still actively practised and the dormant

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site labelled (B) with passive or withdrawing waste dumping. Soil samples were obtained in triplicate at each site at the depth of 0-15 cm using calibrated soil auger. At each sampling site, the surface debris were removed and subsurface soil dug to 0-15 cm. Then 20g of soil sample from each depth was taken into a sterile container and transported to the laboratory for analysis.

Place and Duration of Study: The Analysis was done at the Department of Biochemistry, Anambra State University, Department of Applied Biochemistry, Nnamdi Azikiwe University, Awka and Spring Board Research Laboratory, Awka between February and sMarch 2014.

Methodology: Polycyclic Aromatic Hydrocarbons (PAHs) were determined simultaneously using high resolution gas chromatography/mass spectrometry (GC/MS) (GC/MS Shimadzzi QP 2010) while Heavy metals were analysed using Atomic Absorption Spectrophotometer (AAS).

Results: The result showed that Low molecular weight PAHs were not detected in both active and dormant dumpsite. High molecular weight PAHs (HMW PAHs) were predominant in soil from active site. Among the HMW PAHs, Benzo (a) pyrene was highest followed by Benzo(k) fluoranthene and Indeno (1,2,3-cd) pyrene and the least was 1,2 Benzo(a)anthracene. The medium molecular weight PAHs (MMW PAHs) Fluoranthene occurred sparingly in the active dumpsite. The result of heavy metal concentration in the soil were in the order of Fe > Cu > Zn > Cr > Pb > Ni > Cd.

Conclusion: PAHs and Heavy metal concentration were found to be very high and as such the practice of cultivating vegetables on soil from these dumpsites for agricultural purposes should be discouraged.

Keywords: PAHs; heavy metals; soil pollution; solid waste dump; toxicant.

1. INTRODUCTION

Nearly all human activities generate waste and the manner in which this is handled, stored, collected and disposed of can a pose threat to the environment and to public health [1,2]. Waste landfill open dump is the most common method of disposal of municipal solid waste (MSW) in Nigeria. This is a practice adopted as a substitute to ocean out fall of sewage, domestic and industrial wastes due to the effect of the latter on lives in the ocean [3]. Several fluxes of wastes contain a variety of contaminants which can pollute the soil in the area [4]. Soil is an environmental medium which is subjected to a number of pollutants due to different human activities [5]. Soil is one of the repositories for anthropogenic wastes. Biochemical processes can mobilize the chemical substances. The persistent organic pollutants, polycyclic aromatic hydrocarbons (PAHs) and heavy metals contamination in the soils is a major concern because of their toxicity and threat to human life and environment [6,7].

Onitsha, the largest and most populous city in Anambra State is noted for its high level of commercial activities [8]. It has several manufacturing businesses like, plastics, canned bottled drinks, pharmaceutical products, batteries, electrical wire and cables, paints, greases, motor oils and petroleum products. It is a home of different auto-spare parts both old and new, electrical and electronic products, building materials and hundreds of petrol stations. In addition, a huge artisan population exists in the city to render welding, soldering, brazing, sheet metal working, electrical and other services to the public. Also used consumer products such as electronics, light bulbs, aerosols, cans, lead foils, bottles, caps garbages and other materials are continually generated on a large scale by residents of the city.

A study conducted by Otti [9] on a proposal for solid waste management within Onitsha metropolis found organic wastes to be 27.3%, plastic/polythene 21.2%, cardboard paper 7.8%, metal 11.2%, glass 5.2%, textile 4.6%, ashes dust 14.4%, others 8.8%. These wastes contribute significant quantities of PAHs and heavy metals to the waste of the city. Polycyclic aromatic hydrocarbons (PAHs) (Table 1) are one of the most common classes of organic pollutants detected in open dumpsite [10,11]. They are introduced into the landfill along with petroleum hydrocarbon residues [12] and by deposition from the atmosphere [13], but they can also be formed on the spot in municipal landfill fires [14]. They can occur in the air either attached to dust particles or as solids in soil or sediments [15]. The occurrence of various heavy metals such as As, Cd, Pb, Ni, Zn, Fe, Cu, Cr etc in MSW are of concern and was reported by many workers [16-18].

Since these contaminants affect the environmental qualities in addition to affecting plant and animal health in and around open dump sites, monitoring of soil qualities especially PAHs and heavy metals distribution in dump sites becomes necessary in order to facilitate recommendation of suitable remedial measures [19,20]. There are very few reports on PAHs and heavy metals status in soil from active and dormant solid waste dump site in Anambra State. This study aimed at determining the distribution of PAHs and heavy metals in soils from MSW dump. A significant importance of this work will be in providing baseline information for further soil quality monitoring studies.

Table 1. Classification of polycyclic aromatic hydrocarbon (PAHs) based on molecular weight [21]

PAHs	Chemical formular	Molecular weight	Number of rings
Low molecular weight (LMW)			
Naphthalene	$C_{10}H_8$	128.00	2
Acenaphthylene	$C_{12}H_8$	152.00	3
Acenaphthene	$C_{12}H_{10}$	154.21	3
Fluorene	$C_{13}H_{10}$	166.20	3
Phenanthrene	$C_{14}H_{10}$	178.20	3
Anthracene	$C_{14}H_{10}$	178.20	3
Medium molecular weight (MMW)			
Fluoranthene	$C_{16}H_{10}$	202.26	4
Pyrene	$C_{16}H_{10}$	202.30	4
High molecular weight (HMW)			
Chrysene	$C_{18}H_{12}$	228.30	4
Benzo(a)anthracene	$C_{18}H_{12}$	228.89	4
Benzo(k)fluoranthene	$C_{20}H_{12}$	251.30	5
Benzo(k)fluoranthene	$C_{20}H_{12}$	251.30	5
Benzo(a)pyrene	$C_{20}H_{12}$	251.30	5
Indono(1,2,3-Cd)pyrene	$C_{22}H_{12}$	276.30	6
Benzo(g,h,i)pevylene	$C_{22}H_{12}$	276.30	6
Dibenzo(a,h)anthracene	$C_{22}H_{12}$	278.35	6

2. MATERIALS AND METHODS

2.1 Site Description

The present study focused on Onitsha municipal solid waste open dump site located at latitude 06° 6^1 N and longitude 06° 48^1E opposite National Metallurgical Institute along Onitsha-Owerri road Onitsha, Anambra State, Nigeria (Fig. 1). The dumpsite is surrounded

by factories; gasoline stations, motor parks, auto spare parts market, auto repair workshops, electrical/ electronic market, hospitals and houses.

The dumpsite (Fig. 2) contains mixtures of both organic and inorganic waste materials, such as food wastes, papers, cardboards, metals, tins, glasses, ceramics, battery wastes, textile rags, plastics, sewage might soils and other miscellaneous materials such as bricks, ash, fine dust, rubber and wood waste etc.

2.2 Field Reconnaissance and Sampling

The goal of the field reconnaissance survey was to ascertain the level of pollution on the solid waste dumpsite. Sampling was subsequently carried out by mapping out plots on the dumpsite using the grid system. The dumpsite were divided into two groups, the active site labelled (A) where waste dumping is still actively practised and the dormant site labelled (B) with passive or withdrawing waste dumping. Soil sample were obtained in triplicate at each site at depth of 0-15 cm using calibrated soil auger. At each sampling site, the surface debris was removed and subsurface soil dug to 0-15 cm. Then 20 grams of soil samples from each depth was taken a sterile container and labelled. The entire sample was transported to the laboratory for analysis.

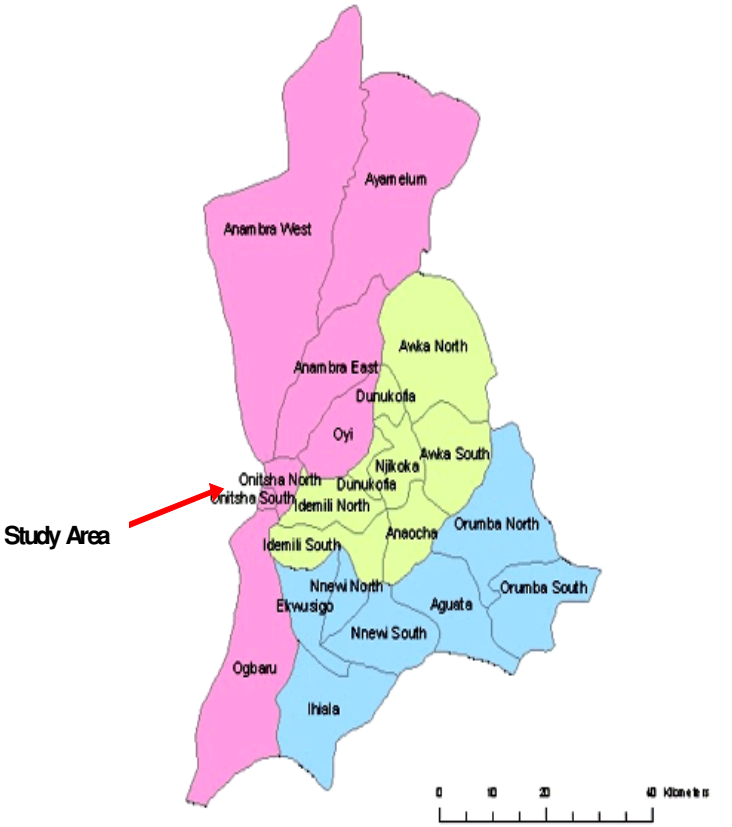

Fig. 1. Map of Anambra State, Nigeria showing the study area

Fig. 2. The municipal solid waste dump site located along Onitsha-Owerri road, Anambra State, Nigeria

2.3 Polycyclic Aromatic Hydrocarbons (PAHs) Analysis

Sixteen non-substituted PAHs (ns, PAHs) with two to five aromatic rings Naphthalene, Acenaphthylene, Acenaphthene, Fluorene, Phenanthrene, Anthracene, Fluoranthene, Pyrene, Chrysene, 1,2-benzoanthracene, Benzo(b)fluoranthene, Benzo(k)fluoranthene, Benzo(a)pyrene, Indeno(1,2,3-cd)pyrene, 1,25-Dibenzoanthracene, 1,2-Benzo(a)perylene were determined simultaneously using high resolution gas chromatography/mass spectrometry (GC/MS) as reported by Tang et al. [20].

Before GC/MS analyses, 10g each of the soil samples were extracted with 50 ml extraction solvent mix of acetone and dichloromethane (50:50 v/v) using an ultra-sonic bath and the extraction was subsequently repeated with 50 ml of the extraction solvent. The sample extract was concentrated using rotary evaporator and then reconstituted with Cyclohexane and reconcentrated to 2 ml and transferred into Cyclohexane and purified using a Silica gel column. The final fractions containing PAHs was eluted with 25 ml of dichloromethane pentane mix (2:3 v/v) and analyzed GC/MS (Shimadzzi QP 2010) equipped with a 30 m *x* 0.25 mm ID fused silica capillary column coated with 0.25 µm thick film of HP-5.

2.4 Heavy Metal Analysis

Heavy metals were determined using Atomic Absorption Spectrophotometer as described by Adelekan and Alawode [18]. The fresh soil was spread on a clean plastic sheet placed on a flat surface and air dried in open air in the laboratory under room temperature for 24 hrs.

Afterwards the soil was sieved using 90.5 mm sieve. After sieving, 0.5 g was weighed from the sieved soil of each sample into a Pyrex 250 ml council flask. Ten (10) ml of Perchloric and Nitric acid mixture in ratio 1:2 was added to the sample and was digested on a hot plate at 105℃ for 30 minutes in a fume cupboard. After digestion the flask was left to cool and the digest was transfered into a 50 ml vial and made up to 50 ml mark with deionized water. The resulting solutions were then analysed using Atomic Absorption Spectrophotometer AAS (GBC Avanta ver 2.02).

2.5 Statistical Analysis

Data obtained were expressed as mean ± standard deviation (SD) and One Way Analysis of Variance (ANOVA) was used to compare obtained means. Values with $P \leq 0.05$ were considered statistically significant.

3. RESULTS AND DISCUSSION

3.1 Results

The analysis of the heavy metal concentrations in soil from Active and Dormant dumpsite is presented in (Table 2).

Table 2. Heavy metal concentrations (mg/kg) in Soil from active and dormant dumpsite

Heavy metals	Active site	Dormant site
Fe	7,460.00±10.00[a]	457.18±0.50[b]
Zn	362.75±0.43[a]	214.43±0.51[b]
Cu	1,976.50±0.50[a]	322.47±0.50[b]
Cr	94.67±0.15[a]	29.41±0.52[b]
Pb	50.00±0.50[a]	28.42±0.52[b]
Cd	5.90±0.10[a]	1.85±0.30[b]
Ni	30.10±0.10[a]	11.10±0.09[b]

**Values are mean±standard deviation of triplicate determinations. Values in the same row bearing the same superscript letter are not significantly different at p<0.05*

The result of the PAHs analysis is presented in (Table 3).

3.2 Discussion

The result of heavy metal concentration in soil from both active and dormant site of MSW dump indicated that heavy metal concentration in active site were significantly higher at ($p<0.05$) than heavy metal concentration in soil from dormant site (Table 2). The heavy metal concentrations were on the order of Fe > Cu > Zn > Cr > Pb > Ni > Cd. The differences in concentration of heavy metals in both sites may be attributed to difference in age of the dump site. The active site still receives waste on daily basis while waste dumping has been suspended at the dormant site for over five years. Other factors include plant uptake, soil pH, organic matter content and other soil characteristics affects the amount of heavy metal leaching [18].

Table 3. PAHs concentration (mg/kg) in soil from active and dormant dumpsite

PAHs constituents	Active site	Dormant site
Low molecular weight		
Naphthalene	0.02±0.01	ND
Acenaphthylene	ND	ND
Acenaphthene	ND	ND
Fluorene	ND	ND
Phenantherene	ND	ND
Anthracene	ND	ND
Sub total low molecular weight	0.02±0.01	ND
Medium molecular weight		
Fluoranthene	0.02±0.02	ND
Pyrene	ND	ND
Sub total	0.02±0.02	ND
High molecular weight		
Chrysene	0.04±0.02	0.01±0.01
1,2 Benzo(a)anthracene	0.02±0.00	ND
Benzo(b) fluorenthene	0.03±0.02	0.01±0.00
Benzo(c) fluoranthene	0.05±0.01	0.03±0.02
Benzo(a)pyrene	0.11±0.01	0.04±0.02
Indeno(1,2,3CD)pyren	0.05±0.01	0.02±0.01
1,2,5 Dibenzoanthracene	0.04±0.01	0.01±0.01
1.2 Benzo(a)perylene	0.04±0.02	0.01±0.00

**Values are mean±standard deviation of triplicate determination. ND = Not Detected; detection limit =0.001mg/kg*

Both dumpsites had the highest Fe concentration (475.18±0.74 – 7,460.00±10.00 mg/kg). This value was higher than 15.52 - 103.46 mg/kg as reported by Tripathi and Misra [21] in dump site at Allahabad India and the value 56.12 - 280.16 mg/kg reported by David et al. [22] in dump site in Part Harcourt, Rivers State, Nigeria. Concentration of Zn in soil ranged from 214.43±0.51 - 362.75±0.43 mg/kg which was higher than the values 18.21 - 108.00 mg/kg reported by Tripathi and Misra [22] and 21.18 - 128.16 mg/kg reported by David et al. [23]. Cu values ranged from 322.47±0.50 – 1,979.50±0.50 mg/kg in this study and were higher than values reported in literature by Tripathi and Misra [22] and David et al. [23]. Elevated levels of these metals in soil from the dump site may be attributed to deposition of solid waste, incidental accumulation, atmospheric depositions and deposition of agricultural chemicals [24].

Fe, Zn and Cu are essential trace metals required by the body for metabolic activities. Fe and Zn are cofactors to some metabolic enzymes in both plants and animals. In case of women, pregnancy and lactation increases the demand for Fe, Zn and Cu [25,26]. Decrease in the store of these essential trace metals may aggravate Cd and Pb toxicity [27]. High levels of zinc above maximum Zn tolerance for human health could produce zinc toxicity which could result to low copper status, altered ion function and reduced immune function while high levels of Cu and Fe may result to nausea, vomiting, diarrhea and abdominal pains [28,29].

Concentrations of Cr in soil from active dump site (94.67±0.15 mg/kg) were significantly higher ($P<0.05$) than the value from the dormant site (29.41±0.52 mg/kg). The values at the active site were higher than the values 6.25 - 75.55 mg/kg reported by Adelekan and

Alawode [18] in dumpsite within Ibadan in Southwest Nigeria but lower than the values 900 – 2000 mg/kg reported by Adefemi and Awokunmi [30]. The values of Cr obtained from this study were higher than Maximum Allowable Limits (MAL) of Cr concentration 60 mg/kg in Germany and Sweden and 30 mg/kg in Denmark and Netherlands [31]. The high concentration of Cr in active dumpsite may be attributed to increase in industrial and municipal waste which contained high concentration of Cr. Accumulation of Cr in the human body can possess health risk.

The value of Pb from the study ranged from 28.42±0.52 – 50.00±0.50 mg/kg. The values of Pb from the active dump site was high compared to the MAL of 40.00 mg/kg set for countries like Denmark, Netherland and Sweden [31] but lower than the limit 70 mg/kg, 100 mg/kg set for Germany and France respectively [31]. The increase in Pb in the active dumpsite may be due to atmospheric deposition owing to its very close surrounding of high vehicular traffic highway and dumping of waste with high Pb content. Environmental protection Agency (EPA) [32] reported lead as a probable human carcinogen. Elevated levels of Pb in human reduces plasma copper which may lead to irreversible damage of the brain, alter the flexibility of red blood cells (RBC), increase cell fragility leading to increased risk of haemolysis [28,33].

Concentrations of Cd in soil obtained from active site were significantly higher at $P<0.05$ than the dormant site. This may be attributed to differences in the age of the dumpsite and biodegradation of waste in dormant dumpsite. The value of Cd from the study ranged from 1.85±0.30 - 5.90±0.30 mg/kg. These values were higher than the MAL for Cd 1.00 mg/kg in Spain and Germany and 0.5 mg/kg in Denmark and Netherland [31]. Indiscriminate dispose of Cd containing waste on the dumpsite and atmospheric deposition may contribute to the high level of Cd in the dumpsite. Accumulation of Cd in humans may lead to acute cadmium poisoning which may cause high blood pressure, kidney damage, destruction of testicular tissues and destruction of red blood cells [34]. Adelekan and Alawole [18] reported that cadmium, lead and mercury can be harmful to animals and humans at relatively low concentrations and thus should receive close scrutiny in relation to application of municipal solid waste composts to Agricultural soil. The concentration of Ni in the dumpsite ranged from 11.10±0.09 to 30.10±0.10 mg/kg.

Concentration of Ni were found to be significantly highest ($P<0.05$) in soil from active dumpsite than in soil from dormant dumpsite. The values from the active site (30.10±0.10 mg/kg) were higher than the values (5.65-19.40 mg/kg) reported by Adelekan and Alawode [18] in soil from dumpsite in Ibadan. The value of Ni found in the active site was higher than the MAL of 15 mg/kg found in countries like Denmark and Netherland but within MAL of 50 – 70 mg/kg as in Austria [31] and 75 mg/kg as in Australia and New Zealand [35]. Elevated level of Ni in soil from active dump site above that of dormant site could be as a result of continuous dumping of Ni compounds and alloys at the active dumpsite and biodegradation of waste in the dormant site over time. Accumulation of Ni in human through inhalation, ingestion or skin absorption may result to cancer of the lung larynx and prostrate. Others include dermatitis, bronchial asthma, myocardial infarction, respiratory failures and birth defects [33,36].

Result of polycyclic aromatic hydrocarbon (PAHs) distribution in the MSW dumpsites were presented in (Table 3). Low molecular weight PAHs (LAW PAHs) were not detected in both soils of active and dormant dumpsite. High molecular weight PAHs (HMW PAHs) were predominant in soil from active site along the HMW PAHs, Benzo (a) pyrene was highest followed by Benzo (k) fluoranthene and Indeno (1,2,3-cd) pyrene and the least was 1,2

Benzo(a)anthracene. The medium molecular weight PAHs (MMW PAHs) Floranthene occurred sparingly in the active dumpsite. The presence of HMW PAHs compared to LMW PAHs may be due to the recalcitrant nature of the HMW PAHs, age of dumpsite, weather and climatic condition. Low molecular weight PAHs such as Naphthalene, Acenaphthylene Acenaphthene, Fluorene, Anthracene and Phananthrene are transformed rapidly by many bacteria and fungi while high molecular weight PAHs however are more recalcitrant in the environment and resist both chemical and microbial biodegradation [37].

The predominance of HMW PAHs in the active site compared to dormant site may be attributed to differences in biological diversity, abundance and the age of pollution given that the dormant site was not receiving waste. Therefore PAH constituents must have volatilized and disappeared from the soil but the high molecular weight constituents still abound but in much lower concentration than in active dumpsite. It is well known that light PAHs compound occur in soil in lower proportions than heavier ones due to their physicochemical properties such as high solubility, volatility, biodegradability and lower sorption ability to soil organic portion [38,39]. This also corroborates the report of Reid et al. [40]; Semple et al. [41] and Dick et al. [42] which stated that the fate of PAHs in the soil is affected by factors such as biological diversity and abundance, the nature of the soil minerals and organic matter and pollutant properties such as solubility, polarity, hydrophobicity, lipophilicity and molecular structure. This might explain the differences in PAHs concentration between soil samples that are not in close proximity as well as variation in PAHs concentration of the same site.

PAHs adsorb to organic matters, percolate the soil and contaminate the aquifers. They are taken up by aquatic species and plants and passed through food chain and bioaccumlate in human on long term exposure. This may lead to human health disorder such as organ dysfunction, reproductive, neurobehavioral and genetic disorders [43]. The international agency for research on cancer (IARC) and Environmental protection Agency (EPA) has determined the following Benzo (a) anthracene, Benzo (a) pyrene, Benzo (b) fluoranthene, Benzo (k) fluoranthene, Indeno (1,2,3-cd) pyrene as possible human carcinogen [44].

Correlation coefficient was analysed to understand the relationship between HMW PAHs and heavy metal concentration. Various notable significant correlations were recorded and matrix of correlation coefficient between different quantitative variables is presented in (Table 4). The result of correlation can prove useful in understanding the relationship of metal concentration with each other and with PAHs distribution in the soil. The correlation study of high molecular weight PAHs and heavy metals from the study sites revealed that there is a significant positive correlation among the heavy metals Pb, Cd, and Cu and among the HMW PAHs Bbf, Chy, Ind and Dah at $P<0.01$ level indicating that a positive linear relationship existed among the heavy metals and among the HMW PAHs suggesting that the source of the heavy metals and HMW PAHs may be the same and could be attributed to waste dump due to human activities. Also there are strong positive relationship between HMW PAHs (Bbf, Baa, Bkf, Chry Indp and Dah) and heavy metals (Pb, Cd, Cu) at $P< 0.01$ level. This showed that the HMW PAHs increased with increase in heavy metal level. The observation that significant negative correlation exist between Bap and Cr indicated that Benzo (a) pyrene increased with decrease in Chromium level. Benzo (a) pyrene one of the most recalcitrant PAHs in soil adsorbs to the soil matrix and thus is physically unavailable to degradative bacteria and fungi. Carl et al. [37] reported that the formation of non extractable bound residues is a significant sink of PAHs in soils.

Table 4. Correlation of HMW PAHs and heavy metal concentration in soil from MSW dumpsite

	Fe	Zn	Cu	Cr	Pb	Cd	Ni	Chry	Baa	Bbf	Bkf	Bap	Indp	Dah	Bapy
Fe	1														
Zn	-.866	1													
Cu	-.500	.000	1												
Cr	-.655	-.1189	-.982	1											
Pb	-.500	.000	.1.000*	-.982	1										
Cd	-.500	.000	- 000*	-.982	1.000*	1									
Ni	.500	.000	-1.000*	.982	-1.000*	-1.000*	1								
Chry	-.500	.000	1.000*	-.982	1.000*	1.000*	-1.000*	1							
Baa	.000	.500	-.866	.756	-.866	-.866	.866	-.866	1						
Bbf	.000	.500	-.866	.756	-.866	-.866	.866	-.866	-1.000*	1					
Bkf	-.500	.000	1.000*	-.982	1.000*	1.000*	-1.000*	1.000*	-.866	-.866	1				
Bap	-.655	.189	.982	-1.000*	.982	.982	-.982	.982	-.756	-.756	.982	1			
Indp	.000	.500	-.866	.756	-.866	-.866	.866	-.866	-1.000*	1.000*	-.866	-.756	1		
Dah	.000	.500	-.866	.756	-.866	-.866	-.866	.866	1.000*	1.000*	-.866	-.756	1.000*	1	
Bapy	-.500	.000	1.000*	-.982	-1.000*	1.000*	1.000*	-1.000*	-.866	-.866	-1.000*	.982	-.866	-.866	1

**Chry = Chrysene, Bbf = Benzo(b)fluoranthene, Bkf = Benzo(k)fluoranthene, Bapy = Benzo(a)pyrene, Indp = Indeno(1,2,3-cd)pyrene, Dah = 1,25-Dibenzoanthracene, Bap = 1,2-Benzo(a)pevylene. (* = correlation is significant at the 0.01 level)*

4. CONCLUSION

The absence of proper management practice and lack of adequate capacity for final deposition of solid waste has been a serious problem in urban areas. The recommendation of the study therefore includes the prevention of further waste dumping at the dumpsite and relocation of dumpsites out of the city. Also proper monitoring and remediation plans should be put in place to reduce the chances of ground water pollution by leaching of these contaminants and the practise of cultivating vegetables on these dumpsite or the introduction of municipal solid waste to agricultural soil should be discouraged.

CONSENT

Not applicable.

ETHICAL APPROVAL

Not applicable.

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COMPETING INTERESTS

Authors declare that there are no competing interests.

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