



# CONCENTRATION OF POLYAROMATIC HYDROCARBON (PAH'S) IN SOIL AND WATER FROM INDISCRIMINATE DISPOSAL OF PHARMACEUTICAL WASTE IN LAGOS NIGERIA



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**Abstract:** The Levels and distributions of the concentrations of poly-aromatic hydrocarbons (PAH's) in soil and water samples were assessed using Gas Chromatography and Mass Spectrometer (GC-MS). PAH's are important for public health and the environment due to their carcinogenic, mutagenic characteristics and harmful effects on soil and vegetation. Samples were collected in wet and dry season to determine the concentration of some (PAH'S) compounds to ascertain their concentrations and compare with allowable standards. Areas covered in this research include; Abule-Egba, Epe, Olusosun and Shagamu in Lagos Nigeria. About 16 PAH's were analysed in the four areas. The results obtained showed high distribution of Benzo(a)anthracene ( $0.47326 \pm 0.45635$  ppm) and Benzo(a)pyrene ( $0.34547 \pm 0.39769$  ppm) in Epe area during the wet season. The percentage of Benzo(k)fluoranthene in soil was found to be 49% while 39% was found in water though higher than the permissible limit as stated by US Occupational Safety and Health Administration (OSHA) as  $0.2 \text{ mg/m}^3$ .

**Keywords:** Polyaromatic hydrocarbon, pharmaceutical waste, organic pollutants, Lagos-Nigeria

## Introduction

Polycyclic aromatic hydrocarbons (PAHs) are a large group of organic compounds with two or more fused aromatic (benzene) rings made entirely from carbon and hydrogen (WHO, 2002; 2010) and do not contain heteroatom or carry substituent and are one of the wide spread organic pollutants (Samat, 2010). PAHs can be classified into low molecular weight PAHs (LMW PAHs), a group that includes PAHs with a number between 2 and 3 rings; intermediate molecular weight PAHs, which includes those PAHs with 4 aromatic rings; and high molecular weight PAHs (HMW PAHs), which includes those PAHs with a number of rings between 5 and 6 (WHO, 2002). They belong to the group of persistent organic pollutants (POPs) which are organic pollutant contaminants that are resistant to degradation and can remain in environment for a long period with potential to cause adverse environmental effects (Manz and Wenzel, 2001). PAHs are compounds susceptible to suffering LRAT, reaching at other environmental matrices such as water and aquatic systems, soils and sediments (Wei and Jen, 2007). Naphthalene is the simplest example of a PAH, some of the more common are Acenaphthene

Dibenzo(a,h)anthracene, Anthracene Fluoranthene, Benzo(a)anthracene Fluorene, Benzo(a)pyrene Indo (1,2,3cd) pyrene, Benzo(b)fluoranthene Naphthalene, Benzo(k)fluoranthene Phenanthrene Chrysene Pyrene. They can have a faint, pleasant odour (Martinez *et al.*, 2004). A few PAHs are used in medicines and to make dyes, plastic, and pesticides (Manz & Wenzel, 2001). Over 100 compounds existing in indoor air have been identified to date; two of the more common ones are benzo (a) pyrene and naphthalene (Martinez *et al.*, 2004).

Environment PAHs exert their toxicity following biotransformation to toxic metabolites, which can be bound covalently to cellular macromolecules such as protein, DNA and RNA, which causes cell damage, mutagenesis and carcinogenesis (ECSC, 2002). They have a relatively low solubility in water, but are highly lipophilic (Tamet *et al.*, 2001), when dissolved in water or absorbed on particulate matter. PAHs are the most toxic among the hydrocarbon families (Zeng & Vista, 2012) and can undergo photodecomposition when exposed to ultraviolet light from solar radiation (Law and Hii, 2006). PAH may also be degraded by some microorganisms in soil (Tam *et al.*, 1983) and their pollutants have high molecular mass, PAHs of 4 and more condensed aromatic rings are considered to be more dangerous than 2 and 3 rings PAHs in view of their potentials (Zheng *et al.*,

2004). The movement of PAHs in the environment depends on properties such as how easily they dissolve in water and how easily they evaporate in the air (Katsoylannis *et al.*, 1998). As persistent organic pollutants (POPs), some of them are susceptible to dispersion on a global scale because in addition to having environmental persistence, (Katsoylannis *et al.*, 1998).

Lagos, a cosmopolitan city with high industrial activities and urban migration that has developed with dumpsites sighted around residential areas that exposes human, animal and vegetation to environmental health risk, routes of the contamination are soil, water and crops. The study was carried out in four selected local government areas in Lagos and environs.

## Materials and Methods

### List of apparatus/instrument

Rotary Evaporator, Separating funnels, Beakers (100 cm<sup>3</sup>), measuring cylinder (10 cm<sup>3</sup>), conical flask, vials 2-mL for GC auto sampler with 4-mL, screw cap, round bottom flask, burette (100 cm<sup>3</sup>), pipette, clamps, Spatula, Sensitive Weighing balance (capable of accurate weighing to 0.01 g), Soil particles, water samples, Ultrasonic bath, fused silica capillary column (see instrument condition for size), Gas Chromatography Agilent 7890 A GC-  $\mu$ ECD GC-MS, micro syringes 10, 25, 50, 100, 500, and 1000  $\mu$ L, What man Filter paper (No. 41).

### List of chemical/reagents

Dichloromethane (DCM), Acetone, Hexane, Sodium Sulphate (NaSO<sub>4</sub>), Distilled water and all solvents used were GC-MS grade, Chloroform/Ethyl Ether, Methylene, PAH's standards 99% purity from Sigma-Aldrich and Agilent Technologies from local vendors Synapse Technologies, Helium Gas for GC-MS (Authorized Agilent Tech representative, Nigeria).

### Methods

#### Sampling

The areas of sampling was cleared from debris and dug between 10 and 15 cm deep using soil ager to remove top layer soil while carefully reaching the desired depth between 15 and 20 cm to obtain composite samples. A caution in the process of digging and packaging the samples was applied as contamination such as dust, inert gases in the atmosphere which may lead to erroneous results can pose a threat during the process (Samat, 2010). PAH can photo chemically decomposed under strong ultraviolet light or sunlight and some can be lost during atmospheric sampling (Tamet *al.*,

2001). Samples which contained unknown pollutants that are volatile were sampled, the samples were well mixed and sub-samples were collected (Weber *et al.*, 2014).

**Sample collection (Vane *et al.*, 2007)**

**Soil**

A small sample to adequately represent each site was taken in duplicate by scooping from the soil auger, mixed and reduced to fine homogeneous powder into foil paper and sealed in a zip lock bag and placed all in one whirlpack bag while maize was taken from its stalk and wrapped with foil paper and preserved for further processing. They were air dried in a room temperature away from light for 10 days

**Water**

Water samples were collected in silica glass bottles from borehole (≈150ft depth), well, and river/stream from four dumpsites and control sites using EPA Method 1998b (2013). They were labelled and stored in refrigerator at temperature of 5-10°C for analysis until extraction and analysis for 10 days.

**pH analysis/temperature**

The pH was measured at the sampling time using a pH meter model ATC pH-911 pen type with backlit display and temperature device model TDS 3-3. A total of 62 of the samples were measured for the analysis.

**Study area**

Lagos is located within the low-lying coastal zone of south-western Nigeria with a population of over 20 million people in 2016 and a total of 1,171.28 km<sup>2</sup> (452.23 square miles) representing 0.4% of Nigeria’s territorial landmass according to (Kumar, 2012). It lies approximately between longitude 2<sup>o</sup>42’ E and 3<sup>o</sup> 42’ E and latitude 6<sup>o</sup> 22’ N and 6<sup>o</sup> 52’ N and generates the highest amount of waste in Nigeria at 6 million tonnes per annum ( Luo *et al.*, 2004).

**Water (liquid-liquid extraction)**

This procedure was adopted using US.EPA 3510C 1991 method. A separatory funnel was used for liquid-liquid extraction to determine level of PAH’s in the samples using GC-MS instrument. A 50 ml Dichloromethane was added to 100 ml water in a separating funnel and shaken vigorously in ratio 1:1 and allow to settle (for efficiency, start with 50 ml water and 50 ml DCM, and later added 50 ml water/DCM). A 50 ml of 30% DCM was again added to the same separating funnel to make up 100ml of DCM and vigorously, all samples were spiked with 2 ml of DAH’s Solvent was filtered through 20 – 30 g of NaSO<sub>4</sub> to trap any possible water. A volume of 2 – 3 ml was reduced using Rotary Evaporator by adding 2 – 5 ml of hexane and transferred in to 2 ml vial ready for GC-MS analysis.

**Chromatographic conditions and MS-detection**

The GC-MS analysis was carried out using Agilent 7890 A GC- μECD gas chromatograph with an Agilent 7693 Auto sampler equipped with chem.-station software under the following conditions: Injected volume 2 mL/splitless, Column:Agilent 19091J-413: 1910.59288, HP-55% Phenyl methyl siloxanand 325°C: 30 m x 320 μm x 0.25 μm, Average Velocity 59.564 cm/sec, Front Detector μECD; Carrier gas: Helium (99.999%), Pressure 19.392 psi; Temperature program: (°C) – 100 run Time =28 min.

**Results and Discussion**

A total of ∑16PAH’s were analysed and observed to be distributed across the four study sites, few of the results that

adequately represent study areas will be discussed in this article. Figs. 1 – 4 shows chromatographic representation of levels and distribution of PAH’s in four selected dumpsites with respect to each compound of PAH, while Tables 1 – 9 show the PAH of all the study area under consideration.

Figure 1 shows summary distribution of Benzo(a)anthracene in soil at 0-20 cm depth, water and maize samples within Abule-Egba, Epe, Olusosun and Shagamu dumpsites. Benzo(a)anthracene in Fig. 2 shows that in wet season at Epe, the soil have highest mean concentration at control site (0.34547±0.39769 mg/kg) while Olusosun had the lowest (0.04492 ± 0.0429 mg/kg). Epe soil in wet season at control site seems to have high deposit of Benzo(a)pyrene 0.34547±0.39769 mg/kg and deposited more at Shagamu (0.3091 ± 0.25115 mg/kg) and Olusosun dry season (0.10925±0.02809 mg/kg). Water had significant concentration of Benzo(a)pyrene (0.10635±0.01956 mg/kg) in Epe at dry season control site. Benzo(a)pyrene was not detected in maize during wet season but had values above detection limit (0.2749 ±0 mg/kg) which suggests high concentration of unknown compound beyond the detection of the instrument. Fig. 3 shows distribution of Benzo(g,h,i)perylene in soil at 0-20 cm depth, water with a significant presence of Benzo(g,h,i)perylene in soil sample detected in dry season at Abule-Egba dump site (0.50824 ±0.18095 mg/kg) but above detection limit at the control site (0.29206 ± 0 mg/kg). Epe recorded significant mean concentration of Benzo(g,h,i)perylene in wet season at control site (0.46598 ±0.24709 mg/kg) and low in dry season active dumpsite site (0.35525 ±0.08936 mg/kg) but was above detection limit in wet season active site (0.54523 ± 0 mg/kg). The highest mean concentration of Benzo(g,h,i)perylene was detected in Olusosun control site in dry season (0.58631 ±0.41613 mg/kg) but low in active site (0.37893 ± 0.07 mg/kg) and absent in wet season at control site. The distribution of Benzo(g,h,i)perylene in Shagamu in dry season dump site was as low as (0.39668 ±0.09072 mg/kg) and control site (0.40633 ±0.01422 mg/kg) while in wet season dumpsite was (0.31787 ±0.02382 mg/kg) and control site was (0.34717 ±0.01857 mg/kg). The mean concentration of Benzo(g,h,i)perylene in maize sample in wet season control site was above detection limit (0.30974 ± 0 mg/kg) which implies high concentration beyond detection by instrument. The water samples analysed has the presence of Benzo(g,h,i)perylene compound in a low concentration in dry season at Abule-Egba dump site (0.30343 ± 0.01607 mg/kg) and wet season control site (0.33529 ± 0.03035 mg/kg) but absent in wet season dump site and above detection limit in dry season control site. In Epe, the water samples have low concentration of Benzo(g,h,i)perylene in dry season dump site (0.33094 ± 0.008 mg/kg) ,control site (0.36161 ± 0.02409 mg/kg) and wet season control site (0.3037 ± 0.00738 mg/kg) but it was above detection limit in wet season active site (0.40667 ± 0 mg/kg). A significant mean concentration of Benzo(g,h,i)perylene was detected in dry season at Olusosun control site (0.37035 ± 0.11072 mg/kg) but low at dump site (0.32773 ± 0.05045 mg/kg) and wet season control site. Shagamu Benzo(g,h,i)perylene concentration was above detection limit in dry season dump site (0.37103 ± 0 mg/kg), control site (0.32596 ± 0 mg/kg) and wet season active site (0.29206 ± 0 mg/kg).

**Table 1: Benzo(a)anthracene water concentration by sampling sites**

Site Location	Dry Season		Wet Season	
	Active (mg/kg) (Mean ± SD)	Control (mg/kg) (Mean ± SD)	Active (mg/kg) (Mean ± SD)	Control (mg/kg) (Mean ± SD)
Abule-Egba	0.20881 ± 0	0.20881 ± 0	-	0.20881 ± 0
Epe	0.20881 ± 0	0.20881 ± 0	0.20881 ± 0	0.20881 ± 0
Olusosun	0.20881 ± 0	0.2089 ± 0.00013	0.44529 ± 0	0.20881 ± 0
Shagamu	0.20881 ± 0	0.20881 ± 0	0.44529±0.33442	-

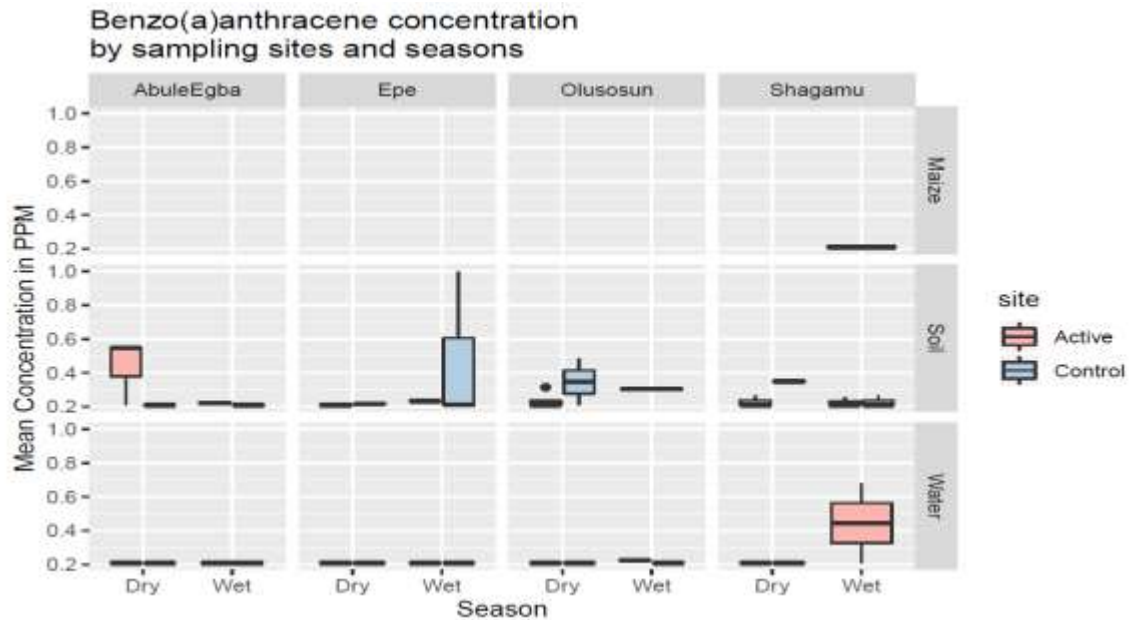


Fig. 1: Benzo(a)anthracene concentration by sampling sites

Table 2: Benzo(a)pyrene soil/maize concentration by sampling sites

Site Location	Dry Season		Wet Season		Maize (mg/kg) (Control)
	Active (mg/kg) (Mean±SD)	Control (mg/kg) (Mean±SD)	Active (mg/kg) (Mean±SD)	Control (mg/kg) (Mean±SD)	
Abule-Egba	0.13723±0.04305	0.09252 ± 0	0.12103±0.03863	0.11565±0.01812	-
Epe	0.13963±0.06662	0.11235±0.01581	0.10283 ± 0	0.34547±0.39769	-
Olusosun	0.10925±0.02809	0.20044±0.15262	0.23955 ± 0	-	-
Shagamu	0.09668±0.00464	0.24791 ± 0.1048	0.16003 ± 0.051	0.3091 ± 0.25115	0.2749±0

Table 3: Benzo(a)pyrene water concentration by sampling sites

Site Location	Dry Season		Wet Season	
	Active (mg/kg) (Mean ± SD)	Control (mgk/g) (Mean ± SD)	Active (mgk/g) (Mean ± SD)	Control (mg/kg) (Mean ± SD)
Abule-Egba	0.11005±0.0248	0.17218±0.13345	-	0.18095±0.00566
Epe	0.20881 ± 0	0.10635±0.01956	0.22368 ± 0	0.10597±0.01638
Olusosun	0.09928±0.00956	0.10245±0.01405	0.0996 ± 0	0.12577±0.03182
Shagamu	0.09556 ± 0	0.19798 ± 0	0.15972±0.09504	-

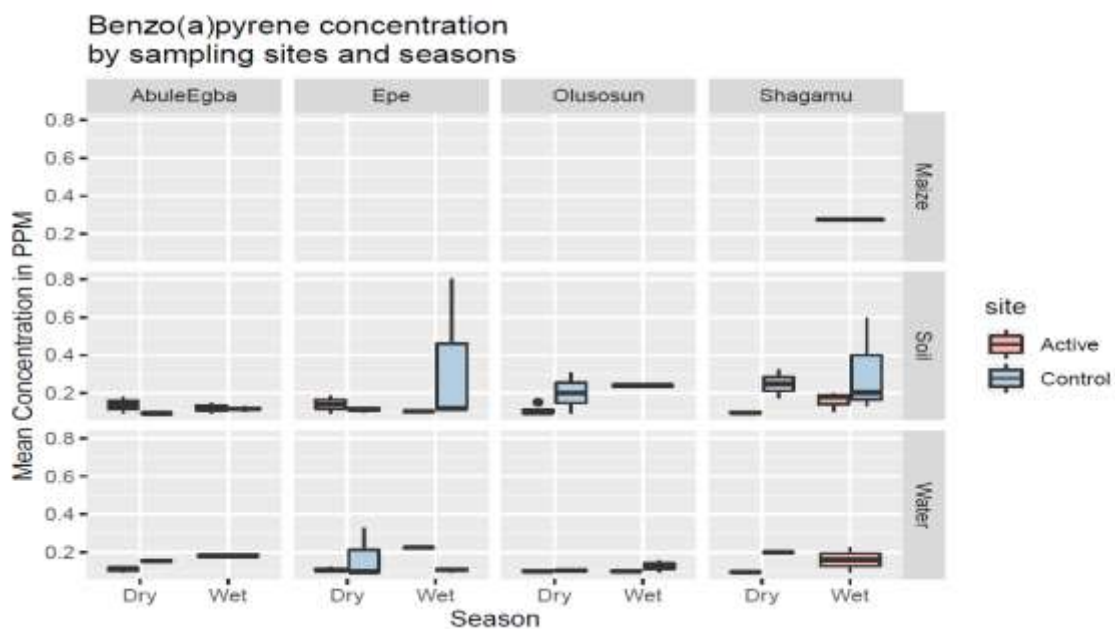


Fig. 2: Benzo(a)pyrene concentration by sampling sites

Table 4: Benzo(g,h,i)perylene soil/maize concentration by sampling sites

Site Location	Dry Season		Wet Season		Maize (mg/kg) (Control)
	Active (mg/kg) (Mean ± SD)	Control (mg/kg) (Mean ± SD)	Active (mg/kg) (Mean ± SD)	Control (mg/kg) (Mean ± SD)	
Abule-Egba	0.50824±0.18095	0.29206 ± 0	0.30227±0.00258	0.43445±0.17823	-
Epe	0.35525±0.08936	0.32706±0.03521	0.54523 ± 0	0.46598±0.24709	-
Olusosun	0.37893 ± 0.07	0.58631±0.41613	0.40566 ± 0	-	-
Shagamu	0.39668±0.09072	0.40633±0.01422	0.31787±0.02382	0.34717±0.01857	0.30974± 0

Table 5: Benzo(g,h,i)perylene water concentration by sampling sites

Site Location	Dry Season		Wet Season	
	Active(mg/kg) (Mean ± SD)	Control (mg/kg) (Mean ± SD)	Active(mg/kg) (Mean±SD)	Control (mg/kg) (Mean ± SD)
Abule-Egba	0.30343±0.01607	0.3364 ± 0	-	0.33529±0.03035
Epe	0.33094 ± 0.008	0.36161±0.02409	0.40667 ± 0	0.3037 ± 0.00738
Olusosun	0.32773± 0.05045	0.37035± 0.11072	0.33846 ± 0	0.33979± 0.02351
Shagamu	0.37103 ± 0	0.32596 ± 0	0.29206 ± 0	-

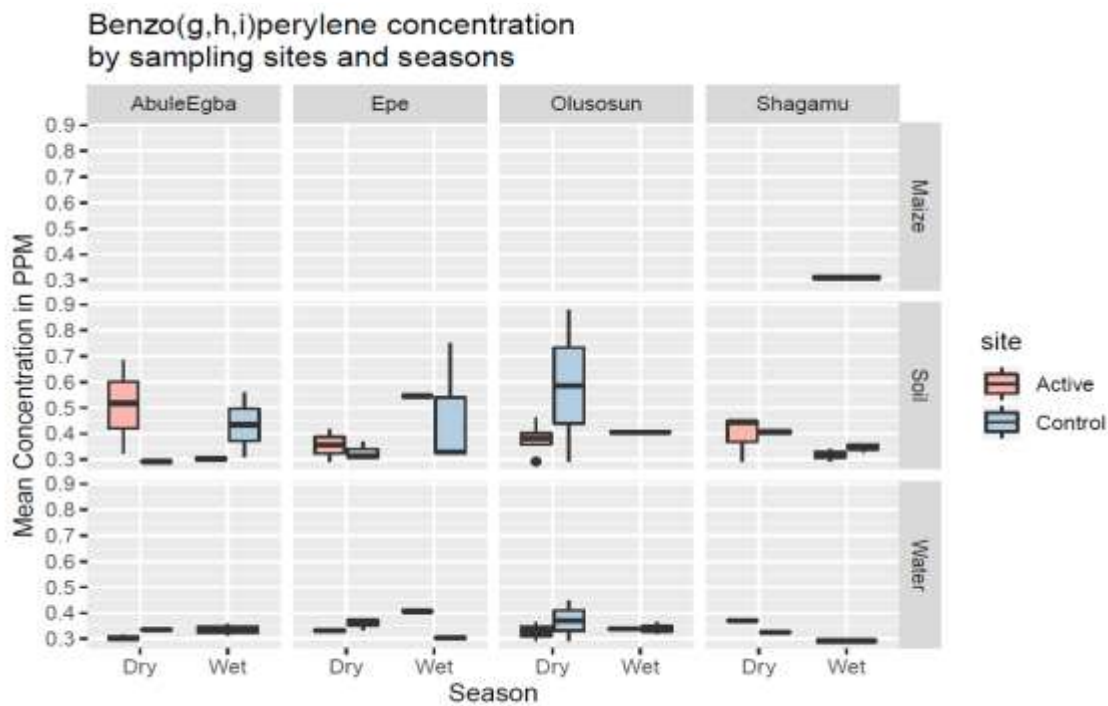


Fig. 3: Benzo(g,h,i)perylene concentration by sampling sites(ppm)

Table 6: Benzo(k)flouranthene soil concentration by sampling sites

Site Location	Dry Season		Wet Season		Maize(mg/kg (Control)
	Active(mg/kg) (Mean± SD)	Control (mg/kg) (Mean± SD)	Active(mg/kg) (Mean± SD)	Control (mg/kg) (Mean± SD)	
Abule-Egba	0.31981±0.08625	0.2159 ± 0	0.21709±0.00169	0.26427±0.05201	-
Epe	0.22226±0.00899	0.22876±0.01076	0.22638 ± 0	0.43572±0.35683	-
Olusosun	0.2352 ± 0.02027	0.33955±0.17487	0.27622 ± 0	-	-
Shagamu	0.23032±0.01305	0.26797±0.07364	0.26084±0.04722	0.25193±0.01503	0.38759±0

Table 7: Benzo(k)flouranthene water concentration by sampling sites

Site Location	Dry Season		Wet Season	
	Active (mg/kg) (Mean ± SD)	Control (mg/kg) (Mean ± SD)	Active(mg/kg) (Mean ± SD)	Control(mg/kg) (Mean ± SD)
Abule-Egba	0.21942± 0.00497	0.22253 ± 0	0.2159 ± 0	0.27125± 0.05113
Epe	0.24072± 0.02781	0.22558± 0.00503	0.40667 ± 0	0.22075± 0.00145
Olusosun	0.2262 ± 0.01457	0.23132± 0.02181	0.22938 ± 0	0.23061± 0.00923
Shagamu	0.21815 ± 0	0.22587 ± 0	0.31274± 0.13696	-

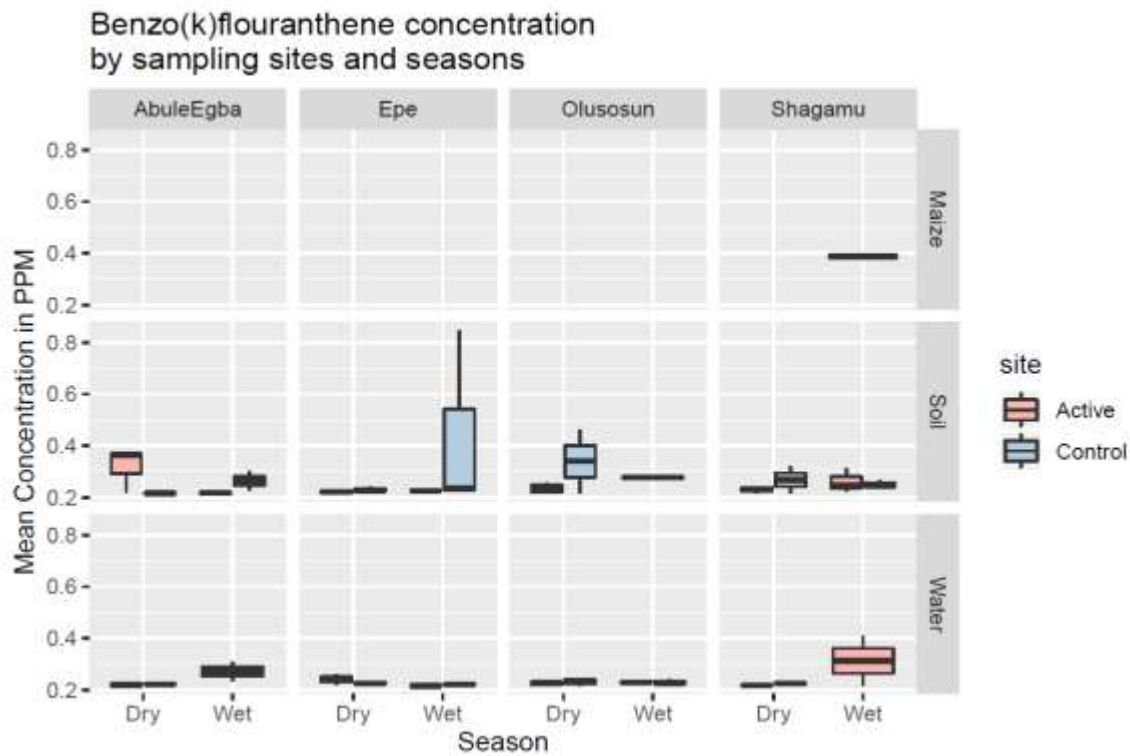


Fig. 4: Benzo(k)flouranthene concentration by sampling sites

Figure 4 shows the summary distribution of Benzo(k)flouranthene mean concentration in analysed soil in dry season at Abule-Egba dump site was significant at  $(0.31981 \pm 0.08625 \text{ mg/kg})$  but above detection limit at control site  $(0.2159 \pm 0 \text{ mg/kg})$ . In both seasons, a significant concentration of Benzo(k)flouranthene was seen to be widely distributed in wet season control site at Epe  $(0.43572 \pm 0.35683 \text{ mg/kg})$  and was above detection limit in the dump site. The control site in dry season at Olusosun has a more significant presence of Benzo(k)flouranthene  $(0.33955 \pm 0.17487 \text{ mg/kg})$  but with low mean concentration in dry season dump site  $(0.2352 \pm 0.02027 \text{ mg/kg})$ . The mean concentration of Benzo(k)flouranthene in maize sample was above detection limit with values that could not be detected by the instrument due to high concentration  $(0.38759 \pm 0 \text{ mg/kg})$ . The mean concentration of Benzo(k)flouranthene in water sample in all the study areas were detected but generally low. Fig. 4 also shows the summary distribution of Naphthalene in soil in dry season at Abule-Egba dump site

with values  $(0.37426 \pm 0.32431 \text{ mg/kg})$  but was not detected in dry season control site, wet season dump site but low concentration was observed in wet season control site  $(0.11433 \pm 0.16168 \text{ mg/kg})$ . Epe has a similar presence of Naphthalene though in a significant concentration  $(0.15671 \pm 0.22162 \text{ ppm})$  in dry season dump site but absent at the control site. In wet season at Epe dump site, Naphthalene was above detection limit  $(0.0455 \pm 0 \text{ mg/kg})$  with a low concentration at the control site  $(0.01252 \pm 0.02169 \text{ mg/kg})$ . The mean concentration of Naphthalene in dry season at Olusosun dump site was significant  $(0.12783 \pm 0.21678 \text{ mg/kg})$  but lower at the control site  $(0.05557 \pm 0.07859 \text{ mg/kg})$ . In water samples, Naphthalene compound was only detected in wet season dump site in a significant concentration  $(0.25522 \pm 0.36093 \text{ mg/kg})$  and absent in Abule-Egba, Epe, Olusosun and Shagamu dry season dump and control sites, respectively.

Table 8: Naphthalene soil/maize concentration by sampling sites

Site Location	Dry Season		Wet Season		Maize (mg/kg) (Control)
	Active (mg/kg) (Mean± SD)	Control (mg/kg) (Mean± SD)	Active (mg/kg) (Mean± SD)	Control (mg/kg) (Mean± SD)	
Abule-Egba	$0.37426 \pm 0.32431$	$0 \pm 0$	$0 \pm 0$	$0.11433 \pm 0.16168$	$0 \pm 0$
Epe	$0.15671 \pm 0.22162$	$0 \pm 0$	$0.0455 \pm 0$	$0.01252 \pm 0.02169$	-
Olusosun	$0.12783 \pm 0.21678$	$0.05557 \pm 0.07859$	$0.25582 \pm 0$	-	-
Shagamu	$0 \pm 0$	$0.497270.31056$	$0.16561 \pm 0.25779$	$0.05299 \pm 0.09178$	$0 \pm 0$

Table 9: Naphthalene water concentration by sampling sites

Site Location	Dry Season		Wet Season	
	Active(mg/kg) (Mean ± SD)	Control (mg/kg) (Mean ± SD)	Active(mg/kg) (Mean ± SD)	Control(mg/kg) (Mean ± SD)
Abule-Egba	$0 \pm 0$	$0 \pm 0$	-	$0 \pm 0$
Epe	$0 \pm 0$	$0 \pm 0$	$0 \pm 0$	$0 \pm 0$
Olusosun	$0 \pm 0$	$0 \pm 0$	$0 \pm 0$	$0 \pm 0$
Shagamu	$0 \pm 0$	$0 \pm 0$	$0.25522 \pm 0.36093$	-

## Conclusion

Sixty-seven (67) samples were collected and analysed for level of PAH's in the environment as well as their ecological and health risk. The results of the study shows the levels of  $\Sigma$ 16PAH's in the environment have been widely identified and distributed in dumpsites as well as control sites and further shows PAH compounds were detected at the dumpsites across all seasons. The results of soil in wet season ranged from highest 1551817.7  $\mu\text{g}/\text{kg}$  at Epe control site to the lowest at Abule-Egba dumpsite 87621.1  $\mu\text{g}/\text{kg}$  while in dry season soil, total PAH's was highest at Abule-Egba dumpsite 1581407.57  $\mu\text{g}/\text{kg}$  to lowest at Epe dumpsite 222459.27  $\mu\text{g}/\text{kg}$ . Maize samples analysed was observed to contain high levels of compounds of PAH with values close to PAH in soil 158758.5  $\mu\text{g}/\text{kg}$ . In wet season at Olusosun, water ranged from 112294.37  $\mu\text{g}/\text{kg}$  to low at Epe control site 74454.07  $\mu\text{g}/\text{kg}$ . In dry season PAH was high at Epe control site 177941.37  $\mu\text{g}/\text{kg}$  and low at Abule-Egba 43472.67  $\mu\text{g}/\text{kg}$ . It was generally observed that there was high deposit of PAH's in wet season soil due to their low solubility; they tend to deposit into sediments and maize through the root from soil as well as water in dry season, both seasons presented heavy pollution with high ecological and human risk to the environment. In conclusion, the control sites soil and water seems to be susceptible to high risk to PAH exposure it is worth to note that the risk levels of PAH in Maize at Shagamu and soil in Epe community should be closely monitored to reduce the high risk of exposure. This study however will provide an overview options for actions to be taken to minimize the indiscriminate disposal of pharmaceuticals into the environment and evaluates their suitability for inclusion into the strategy approach to international chemical management (SAICM) in the global plan for action especially to developing countries.

## Conflict of Interest

Authors declare that there is no conflict of interest reported in this work.

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