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**LEVELS OF POLYAROMATIC HYDROCARBONS IN BOREHOLE WATER OF  
SELECTED SETTLEMENTS IN THE COASTAL AREA OF ONDO-STATE, NIGERIA**

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**ABSTRACT**

PAHs are persistent toxic contaminants in the environment which pose health risk to human and animals when ingested. This study was used to assess the quality of 264 borehole water samples of selected settlements covering 33 locations divided into four districts (identified as I, II, III & IV) between 2017 and 2019. PAHs were extracted from the water samples using standard method. GC-FID was used for the analysis of PAHs. PAHs concentrations (mg/L) ranged from ND to  $7.77 \pm 1.54$  and  $0.01 \pm 0.00$  to  $7.10 \pm 2.18$  for wet and dry seasons respectively. There were significant differences ( $p < 0.05$ ) between the two seasons for concentrations of Naphthalene, Acenaphthylene, Fluorene, Phenanthrene, Fluoranthene, Benzo(b)fluoranthene and Benzo(a)pyrene. The results obtained from this study suggest no significant risk to this population given the toxicity of these PAHs with exception to district I is of public health concern as Fluoranthene, Benzo(b) fluoran and Benzo(a) pyrene are known carcinogens. This contamination in district 1 is of public health concern as several PAHs are known carcinogens. It is recommended that advanced technological engineering be applied to contain the presence of these pollutants in drinking water sources of residents of the area.

**Keywords:** GC-FID, borehole water, PAHs, toxicity and WHO

## INTRODUCTION

Polycyclic aromatic hydrocarbons (PAHs) are a large group of organic compounds with two or more fused aromatic rings. They have a relatively low solubility in water, but are highly lipophilic. PAHs can leach from soil into water. Water contamination also occurs from industrial effluents and accidental spills during oil shipment at sea. Groundwater sources of PAH contamination could also be from seepages from waste disposal sites, oils spills, surface and underground storage tank leakages, agro activities, effluents discharges, and deliberate dumping (Ogbuaguet *et.al.*, 2011). Contaminated groundwater sources pose potential risk to the local water consumers as well as the natural environment (Ritter *et.al.*, 2002).

The environmentally significant PAHs are those molecules that contain 2 to 7 benzene rings. PAHs are divided into two groups based on their physical, chemical, and biological characteristics (Martinez *et.al.*, 2004). The lower-molecular-weight (LMW) PAHs, for example, the 2 to 3 rings of PAHs such as naphthalenes, fluorenes, phenanthrenes, and anthracenes, have significant acute toxicity to aquatic organisms. The high-molecular-weight (HMW) PAHs, containing 4 to 7 rings, from chrysenes to coronenes, do not cause acute toxicity but are known to be carcinogenic (Neff, 1979).

Several PAHs have been classified by the World Health Organization (Wanget *et.al.*, 2007), the Agency for Toxic Substances and Disease Registry (Kim *et.al.*, 2013), and USEPA (1993) as carcinogenic, while others have been confirmed to be at least, mutagenic to mammals. Concentrations of benzo(a)pyrene in drinking water are generally lower than those in untreated water and about 100-fold lower than the U.S. Environmental Protection Agency's (EPA) drinking water standard. (EPA's maximum contaminant level [MCL] for benzo(a)pyrene in drinking water is 0.2 parts per billion (ppb).

Oil exploration in the Niger-Delta region of Nigeria since mid-20th century has brought about environmental degradation to this region. Few people are fully aware of their daily dependence upon crude oil. Awareness is growing as the fragilities of modern civilization, stemming from population growth and economic expansion become recognizable not only as world and national problem but also as personal problem. However, as human dependency on crude-oil increases the dangers that accompanied it also increases. During the transportation of crude oil through tankers and pipelines, oil spills occur and can cause greater ecological damage (Burgherr, 2007). Non availability of potable water to settlements necessitates heavy reliance on rivers for domestic, agricultural or recreational purposes.

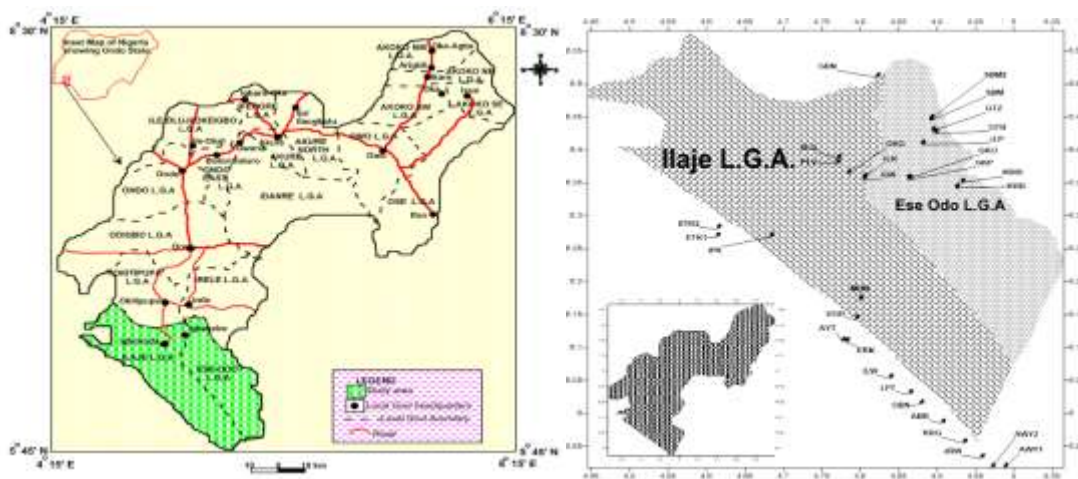
The study areas were oil producing towns of Ondo State in the southern Nigeria. Their rivers empty to the Atlantic Ocean and to some other parts of the country. The river is known for sea foods (Ololade *et.al.*, 2008) which mean that its pollution may have national and global health effects. It is against this suspicion and for the non-existence of research on the possible contamination of the groundwater aquifer of this area that the current investigation was conducted with objectives aimed at the determination of the presence and levels, and spatial variations of some PAHs in borehole water sources of the coastal areas of Ondo state.

## 2. MATERIALS AND METHODS

### 2.1 Description of the study Areas

The study areas comprise mainly selected communities in Ilaje and Ese-odo Local Government Areas of Ondo state, Nigeria. Ondo State comprises of eighteen (18) Local Government Areas, as presented in Figures 1 and 2. Ilaje Local Government consists of over four hundred towns and villages covering an area of 3,000 square kilometers (Adeleke *et.al.*, 2015). This area constitutes one of the major oil producing areas in the state and part of the Niger Delta region of Nigeria. The estimated population of Ilaje LGA is about 254,235 according to National Population Census of 2006. The area is purely a riverine community having two major climatic seasons namely; wet or rainy which begins in March or April and end in October while, dry season starts in November and end in March. The people of Ilaje belongs primarily to the Yoruba ethnic group and are divided into seven political wards namely Ugbo1, Ugbo 2 and Ugbo 3; Aheri, Etikan and Mahin 1 and Mahin 2.

The predominant occupation of the people in this area is fishing although artisans and petty traders are also found. There is no pipe-borne water and the few hand-dug wells are not potable (brackish water) due to sea salt intrusion, oil and domestic contaminations. Therefore, the only source of potable water in this area is from boreholes. However, Ese-odo LGA has a landed area of 762km<sup>2</sup> with a population of 154,978 (NPC, 2006). It lies between longitudes 2°24 to 3°24 and latitudes 6°22 to 6°42. The mean annual total rainfall is >2000mm with a mean monthly temperature of 28-29°C (Adeleke *et.al.*, 2015).



**Fig 1: Map of Ondo state showing the study Areas Fig 2: The Digitized map of all sampling points in the coastal area of Ondo-state**

**Table 1: Ilaje and Ese-odocoastal communities divided into districts**

DISTRICT I	DISTRICT II	DISTRICT III	DISTRICT IV
Alagbaka-Akure water scheme	Awoye (AWY I)	Etikan I (ETK I)	Ojuala (JLP)
Owena- Ondo water works	Ayetoro (AYT)	Etikan II (EKT 2)	Ugbotu-Zion (GTZ)
Igbaraoke- Ilesha water works	Ikorigho (KRG)	Igbokoda (GBN)	Ugbotu-Bolorunduro (GTB)
	Ilowo (ILW)	Ilara (ILR)	Igbobimi (IGK)
	Ilepete (LPT)	Ipare (IPR)	Igbekebo-Okoto (GKO)
	Jirinwo (JRW)	Mahin (MHN)	Igbekebo-Pekagha (GKP)
	Erunna (ERN)	Okoga (OkG)	Sabomi I (SBM)
	Obe-Rewoye (ABR)	Palm Village (PLV)	Sabomi II (SBM 2)
	Obe-Rebiminu (AWY 2)	Salem Quarters (SLQ)	Kiribi I (KRB I)
	Obe-Nla (OBN)		Kiribo II (KRB 2)
	Ugbonla (UGB)		

## 2.2 Sample Collection

Thirty boreholes waters were sampled within the selected communities of the coastal area which are grouped into districts I, II and III as presented in Table 1. Samples for PAHs analysis were collected in amber bottles in each season of 2014 and 2015 respectively. Again, samples were taken from three different urban water supply schemes (water works) which include Owena-Ondo, Owena-Ilesa and Alagbaka-Akure as district I to serve as control, a total of two hundred and sixty-four samples were taken each season respectively in all the communities of the study area.

## 2.3 Laboratory Analysis

### 2.3.1 Sample Preservation

A gas chromatography coupled with flame ionization detector (GC-FID model HP 5890); utilizing the column chromatograph for cleaning of sample extracts was used in the analysis of samples. Analytical grade reagents used include N hexane as solvent, silica gel as desiccant, conc. H<sub>2</sub>SO<sub>4</sub> as preservative for samples, and reagent water that was prepared by passing tap water through a carbon filter bed containing about 0.5kg activated carbon, using a water purification system. A PAH standard mixture containing 500 ppm each of fluoranthene, pyrene, benzo (a) anthracene, chrysene, benzo (b) fluoranthene, and benzo (k) fluoranthene was added to the samples before extraction and used as internal standard. Stock solutions

were used to prepare working standard solutions for calibration.

### **2.3.2 Extraction Procedure for PAHs in water**

PAHs in borehole water samples were extracted according to the method of USEPA 3510C (Liquid-liquid extraction method) 500ml of water sample was poured into a separating flask. Then, 40ml of dichloromethane ( $\text{CH}_2\text{Cl}_2$ ) was added into the flask and shaken, with pressure released at intervals. The sample was allowed to stand until two distinct layers were formed in the flask.

The lower layer (organic extract) of the sample was filtered into a beaker through a filter paper containing glass wool and anhydrous sodium sulphate. The process was repeated two times with 20ml of extracting solvent,  $\text{CH}_2\text{Cl}_2$ , added. The extracts were combined and concentrated by evaporation at room temperature overnight in a fume cupboard by covering with perforated aluminium foil. Sample clean-up was done by USEPA method 3630C.

A 600 X 19 mm clean-up column was prepared. The hole was blocked with glass wool, 3g of activated silica gel (60 resin) was added and the column was topped with sodium sulphate. The column was rinsed by diluting with 20ml on hexane and discarded. This concentrated extract was loaded onto prepared column and diluted with 50ml in hexane. The diluents were then concentrated to 1ml using rotary evaporator under a gentle stream of pure nitrogen. Then 1ml of the extract was transferred into a well labelled vial and stored at  $4^\circ\text{C}$  prior to gas chromatograph flame ionization detector (GC-FID).

### **2.3.3 Instrumentation and Analytical Conditions**

An agilent 5890 series II gas chromatograph fitted with flame ionization detector (GC-FID), with on-column capillary injection systems and Hewlett-Packard Model 7673 auto samples. The fused silica capillary column DB-5, 30m X 0.25mm internal diameter and 0.25  $\mu\text{m}$  film thickness (5% phenyl-silicone), 95% methyl silicone). High purity nitrogen was used as carrier gas at flow rate of 1ml/min and 30ml/min respectively. The separation was performed in split less mode with an oven temperature programmed from  $80^\circ\text{C}$  was held for 2mins, increased to  $190^\circ\text{C}$  at  $5^\circ\text{C}/\text{min}$ , and then increase to  $25^\circ\text{C}$  at  $10^\circ\text{C}/\text{min}$ . The injector and detector temperatures were at  $250^\circ\text{C}$  and  $300^\circ\text{C}$  respectively.

### **2.4 Statistical Analysis**

Inferential statistics were used for analyses of the data generated. Analysis of variance (ANOVA) and Pearson correlation were performed on the data using SPSS 20.0 for significant variations and inter-element relationships. Results were presented as the mean  $\pm$  standard error. Correlation Analysis was done using SAS (1999).

**Table 2: Seasonal mean values of Poly Aromatic Hydrocarbons (PAHs) in borehole waters in the Coastal Areas of Ondo State during the Raining Season.**

PAH (mg/L) Congeners	WET				Mean±S.E
	I	II	III	IV	
Naphthalene	BDL	BDL	0.03±0.02	BDL	0.01±0.01
Acenaphthalene	0.65±0.45	BDL	BDL	BDL	0.06± 0.05
Acenaphthene	0.74 ±0.32 <sup>a</sup>	0.39± 0.07 <sup>b</sup>	0.33± 0.10 <sup>b</sup>	BDL	0.35± 0.05
Flourene	6.25± 4.24 <sup>a</sup>	0.03± 0.01 <sup>b</sup>	0.07± 0.04 <sup>b</sup>	0.21± 0.08 <sup>b</sup>	0.61± 0.45
Anthracene	26.64± 2.35 <sup>a</sup>	0.18± 0.08 <sup>b</sup>	0.21± 0.17 <sup>b</sup>	0.05± 0.03 <sup>b</sup>	2.66± 0.23
Phenanthrene	1.03± 0.10 <sup>a</sup>	0.20 ±0.11 <sup>b</sup>	0.22± 0.12 <sup>b</sup>	0.37± 0.02 <sup>b</sup>	0.36± 0.12
Flouranthene	0.23± 0.03 <sup>a</sup>	0.01± 0.01 <sup>b</sup>	0.26± 0.03 <sup>a</sup>	0.45± 0.22 <sup>b</sup>	0.14± 0.08
Pyrene	0.02± 0.00 <sup>a</sup>	0.02± 0.00 <sup>a</sup>	0.25± 0.02 <sup>a</sup>	0.14± 0.01 <sup>a</sup>	0.14± 0.08
Benzo (a) anthracene	3.35± 0.25 <sup>a</sup>	0.18± 0.02 <sup>b</sup>	0.57± 0.06 <sup>a</sup>	0.19± 0.02 <sup>a</sup>	0.53± 0.29
Chrysene	8.81± 0.83 <sup>a</sup>	7.17 ±3.85 <sup>a</sup>	2.95 ±1.39 <sup>a</sup>	0.01± 0.00 <sup>b</sup>	7.77 ±1.54
Benzo (b) fluoranthene	37.01± 1.45	0.48± 0.04 <sup>b</sup>	1.71± 0.12 <sup>b</sup>	3.01± 0.21 <sup>b</sup>	4.22± 0.32
Benzo (k) fluoranthene	0.64± 0.39 <sup>a</sup>	0.99± 0.17 <sup>a</sup>	0.94± 0.03 <sup>a</sup>	0.71± 0.05 <sup>b</sup>	0.88± 0.132
Benzo (a) pyrene	0.64 ±0.40	BDL	BDL	0.80± 0.03 <sup>a</sup>	0.06± 0.04
Dibenzo (a,h) anthracene	19.82± 11.27	0.15± 0.02 <sup>b</sup>	BDL	BDL	1.85± 1.31
Benzo (g,h,i) perylene	0.03± 0.00	BDL	BDL	BDL	0.002± 0.00
Indeno (1,2,3-cd) pyrene	0.35± 0.03 <sup>a</sup>	BDL	0.002± 0.00	BDL	0.004 ±0.00

Means with similar superscript along the row indicate no significant ( $p>0.05$ ) difference in their mean values  
 Sampling Points I, II, III and IV indicate Districts.  
 BDL: Below detection Limit.

**Table 3: Seasonal mean values of Poly Aromatic Hydrocarbons (PAHs) in borehole waters in the Coastal Areas of Ondo State during the Dry Season.**

PAH(mg/L) Congeners	DRY				Mean±S.E
	I	II	III	IV	
Naphthalene	BDL	BDL	0.03±0.02	BDL	0.01±0.00
Acenaphthalene	0.61±0.04 <sup>a</sup>	BDL	BDL	0.13 0.02	0.10± 0.06
Acenaphthene	0.74 ±0.32 <sup>a</sup>	0.42± 0.07 <sup>b</sup>	0.33± 0.10 <sup>b</sup>	0.23 0.07 <sup>b</sup>	0.36± 0.05
Flourene	6.25± 1.24 <sup>a</sup>	0.02± 0.01 <sup>b</sup>	0.07± 0.04 <sup>b</sup>	0.05± 0.03 <sup>b</sup>	0.61± 0.45
Anthracene	26.64± 2.32 <sup>a</sup>	0.20± 0.08 <sup>b</sup>	0.21± 0.16 <sup>b</sup>	7.06± 2.64 <sup>b</sup>	4.89± 0.31
Phenanthrene	1.03± 0.03 <sup>a</sup>	0.09 ±0.05 <sup>b</sup>	0.22± 0.02 <sup>a</sup>	0.72± 0.32 <sup>a</sup>	0.42± 0.14
Flouranthene	0.23± 0.01 <sup>a</sup>	0.01± 0.01 <sup>b</sup>	0.26± 0.01 <sup>a</sup>	0.14± 0.02 <sup>a</sup>	0.14± 0.08
Pyrene	BDL	0.02± 0.02 <sup>a</sup>	0.25± 0.01 <sup>a</sup>	0.19± 0.01 <sup>a</sup>	0.14± 0.09
Benzo (a) anthracene	3.52± 0.24 <sup>a</sup>	0.01± 0.01 <sup>c</sup>	0.57± 0.05 <sup>a</sup>	0.22± 0.02 <sup>b</sup>	0.55± 0.30
Chrysene	8.81± 0.82 <sup>a</sup>	10.11 ±0.00 <sup>a</sup>	2.95 ±1.39 <sup>a</sup>	3.00± 0.21 <sup>b</sup>	5.67 ±1.71

Benzo (b) fluoranthene	37.01± 3.14 <sup>a</sup>	0.02± 0.02 <sup>c</sup>	1.72± 0.12 <sup>c</sup>	9.79± 0.90 <sup>b</sup>	7.10± 4.18
Benzo (k) fluoranthene	0.64± 0.04 <sup>a</sup>	0.93± 0.18 <sup>a</sup>	0.94± 0.04 <sup>a</sup>	0.84± 0.29 <sup>b</sup>	0.88± 0.13
Benzo (a) pyrene	0.64 ±0.32 <sup>a</sup>	BDL	BDL	0.09± 0.01 <sup>b</sup>	0.09± 0.05
Dibenzo (a,h) anthracene	19.83± 1.27 <sup>a</sup>	BDL	BDL	3.55± 0.24 <sup>b</sup>	2.99± 1.73
Benzo (g,h,i) perylene	0.03± 0.00 <sup>a</sup>	BDL	BDL	0.01± 0.00 <sup>a</sup>	0.10± 0.00
Indeno (1,2,3-cd) pyrene	0.03± 0.00 <sup>a</sup>	BDL	0.01± 0.00 <sup>a</sup>	0.01± 0.00 <sup>a</sup>	0.01 0.00

Means with similar superscript along the row indicate no significant ( $p>0.05$ ) difference in their mean values  
Sampling Points I, II, III and IV indicate Districts.  
BDL: Below detection Limit.

## DISCUSSION

The concentrations and descriptive statistics of the occurrence of 16 congeners of PAHs analysed in this study showed that borehole samples in the Coastal area of Ondo state, Nigeria contain relatively low amount of PAH. During the wet and dry seasons, the mean concentrations of Naphthalene, Acenaphthylene, Acenaphthene, Fluorene, Anthracene, Phenanthrene, Fluoranthene, Pyrene, Benzo(a) anthrac, Chrysene, Benzo(b) fuoran, Benzo(a)pyrene, Dibenzo(a,h)ant, Benzo(g,h,i)per and Indeno (1,2,3) are as presented in Table 4. The concentrations were below detection limit in some districts during the study. It can be seen that 16 out of the 16 target US EPA priority PAH compounds reported in this study were frequently detected in all the borehole water samples including the control samples. There were significant ( $p<0.05$ ) differences in the concentrations of PAHs such as Acenaphthene, Fluorene, Anthracene, Phenanthrene, Pyrene, Benzo(a)anthracene, Chrysene, Dibenzo (a,h)anthracene and Indeno (1,2,3)cd in the waters from the borehole in the coastal districts during wet season. These concentrations were significantly ( $p<0.05$ ) higher in district I than all others during wet season. Acenaphthene, Fluorene, Pyrene, Benzo (a) anthrac, Chrysene and Dibenzo (a,h) in district I were excessively higher than those recorded for other districts in dry season for the coastal area of Ondo State.

In general, the low molecular weight PAHs (LMW: 2- and 3-ring PAHs) were less dominant, accounting in some cases for as low as 2% of the total PAHs, this trend contrasted the results reported by Farooq *et.al.*, (2011) and Thavamani *et.al.*, (2012), both reported that molecular weight PAHs (LMW: 2- and 3-ring PAHs) were dominant, accounting in some cases for as high as 90% of the total PAHs, for surface water samples from Greece, where Naphthalene and Phenanthrene accounted for 82% of the total PAHs. However, the low molecular weight PAHs such as Naphthalene, Fluorene, Anthracene and Phenanthrene are toxic to aquatic organisms while the high molecular weight PAHs such as Chrysene, Benzo(b) fouran are not, though are known to be carcinogenic (Gan and Ng, 2012; Fang *et.al.*, 2014; Khairy *et.al.*, 2014; Griffith *et.al.*, 2015; Soltani *et.al.*, 2015; Yebra-Pimentel *et.al.*, 2015). High significant values reported in district I, control samples (urban water supply schemes) might be due to elevated concentrations in drinking water due to coal-tar lined pipes as reported by Davi *et.al.* (1994).

Reporting on district basis, study showed that Naphthalene, Benzo (a) pyrene and Benzo (g,h,i)per were not detected in Districts II and IV in wet season. Significant ( $p<0.05$ ) differences in the concentrations of Anthracene, Benzo (b) fuoran and Dibenzo(a,h)ant: 26.64±2.35mg/L and 26.64±2.32mg/L,

37.01±1.45mg/L and 37.01±3.14mg/L as well as 19.82±11.27mg/L and 19.83±1.27mg/L were observed in District I in both seasons.

Similar trend of PAHs distribution in borehole water was observed in both seasons with few exceptions in District IV for Anthracene (0.37±0.02mg/L and 7.06±2.64mg/L), Benzo (b) fuoran (0.71±0.05mg/L and 9.79±0.90mg/L), Benzo (a) pyrene (0.00±0.00mg/L and 0.09±0.01mg/L) and Dibenzo (a,h) ant (0.00±0.00mg/L and 3.55±0.24mg/L) for wet and dry seasons respectively. The PAH concentrations were significantly ( $p < 0.05$ ) higher in district I than all others during wet season and also for the PAHs Acenaphthene, Fluorene, Pyrene, Benzo (a) anthrac, Chrysene and Dibenzo (a,h) in district I were excessively higher than those recorded for other districts in dry season for the coastal area of Ondo State.

## CONCLUSION

The results of PAHs congeners of the borehole water samples were within the WHO permissible levels with the exception of anthracene, acenaphthene, fluorine, anthracene, chrysene, benzo(b) fuoran, dibenzo(a,h) anthracene all found in district 1 in both seasons. The results of all PAHs congeners showed no significant difference within the season except for pyrene.

In conclusion, most of the borehole water met the PAHs requirements as stipulated by WHO. However, there is need to put in place facilities for potable water in district 1 (water from water works) to reduce exposure of PAHs to humans through consumption of water from this source.

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