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Determination of Polycyclic Aromatic Hydrocarbons (PAHs) Concentration in Borehole Water from Orhuworhun Town, Delta State, Nigeria.

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ABSTRACT

The quest for quality water and its importance have made it necessary to assess the potability of the water supply. This investigation was carried out to evaluate the concentration of PAHs in borehole water obtained from Orhuwhorun town in Delta State, Nigeria. The water sample was collected from a resident borehole and stored in a sterile bottle to avoid contamination. The water sample was analyzed for PAHs with the use of a gas chromatography mass spectrometer (GC-MC). The total concentration of PAH in water (Σ16 EPA PAHs), which are 16, includes chrysene (Chry), benzo[b]fluoranthene (BbF), acenaphthene (Ace), benz[a]anthracene (BaA), acenaphthylene (AcPy), anthracene (Ant), benzo[k]fluoranthene (BkF), benzo[ghi]perylene (BghiP), benzo[a]pyrene (BaP), Indeno[1,2,3-cd]pyrene (IndP), fluoranthene (Flu), dibenz[a,h]anthracene (DahA), naphthalene (Nap), pyrene (Pyr), phenanthrene (Phe), and fluorene (Flu). The mean concentration in this study ranges from 22.729 to 102.109 mg/L. The concentration of dibenz[a,h]anthracene (DahA) was observed to be the highest, while the concentration of Chry was found to be the lowest amongst all Σ16 PAH congeners detected. The result of this study was compared with the U.S. EPA Standard Limit, and all PAH congeners were found to exceed the standard limit, which shows that the sample is not fit for drinking. This significant concentration of PAH in the water can be attributed to various anthropogenic activities and untreated waste containing PAH. Therefore, there is an urgent need for regulatory agencies to make sure that industries check for environmental compliance.

Keywords: Water quality; concentration; PAHs; Environment; Pollutants; Remediation

1. Introduction

Water is one of the most essential natural resources needed for life as we know it. It is the responsibility of the government to supply drinkable water in the majority of urban areas across the world, including Nigeria. Most of the time, the obligation is not fulfilled to a sufficient degree, which forces the residents of those places to search elsewhere for their water needs. It's possible that the substitute is unhealthy (Chika and Prince, 2020). One of the numerous anthropogenic activities that release pollutants into the ecosystem as a result of fast industrialization and urbanization is the production of polycyclic aromatic hydrocarbons, or PAHs (Qiao et al., 2018; Mojiri et al., 2019). Transportation fuel, power plant emissions, oil spills, coal mining, and other human-caused sources are among the sources of PAHs, which are produced when organic matter is not completely burned. One of the major sources of PAHs in the environment includes biologic, petrogenic, and pyrogenic sources (HącWydro et al., 2019). Plants, algae, microbes, phytoplankton, and gradual changes in organic matter can all produce persistent organic pollutants of biogenic or diagenetic origin (Gennadiev and Tsibart, 2013; Rocha and Palma, 2019). More than 400 different forms of PAHs have been found so far. These include low molecular weight (2-rings or 3-rings) and high molecular weight (4-rings or more rings) benzene rings joined in linear, angular, or cluster patterns (Pan et al., 2006). Colorless, pale-yellow solids, or white make up the majority of PAHs (Pogorzelec and Piekarska, 2018). Owing to the intrinsic, enduring, and extensively harmful characteristics of PAHs, eliminating them from the environment has become a worldwide issue. Adeniji et al. (2019) claim that PAH pollutants are ubiquitous and present in both terrestrial and aquatic ecosystems, as well as the atmosphere. The rate of PAH deposition in soil or sediments has been found to have accelerated due to their enhanced hydrophobicity and low water solubility. As a result of their strong adsorption onto soil particles, PAHs ultimately find their way into the soil ecosystem (Lu et al., 2011; Kuppusamy et al., 2017). According to García-Sánchez et al. (2018), PAH contamination has a negative impact on human health and welfare, as well as the health and welfare of living things worldwide, either directly or indirectly. Since the contaminated matrix and the surrounding conditions are the two main factors in appropriate PAH remedial procedures, they are always crucial (Kuppusamy et al., 2017). With varied degrees of success, several remediation methods involving biological, chemical, physical, and more recently established combined methods have been used consistently. Recently, effective PAH pollution reduction has also been documented using integrated remediation techniques.Amongst the various remediation strategies, microorganism-based methods for restoring the ecology of PAH-polluted environments have been extensively studied (Kuppusamy et al., 2017; Malla et al., 2017; Mehetre et al., 2019).

The waste motor oils, lubricants, and organic solvents that the people of Orhuwhorun town in Delta State, Nigeria, routinely discard or accidentally release into the environment may contaminate their workshops and neighboring areas with petroleum hydrocarbon compounds. According to reports, a significant portion of the harmful contaminants often discharged from cars and industrial activities in most Nigerian cities are waste oils and organic solvents used in most workshops. The mixture of chemicals found in used or waste motor oil includes petroleum hydrocarbon compounds, additives, decomposition products, metallic components from engine wear, chloro-dibenzofurans, and chlorinated biphenyls. These pollutants may find their way to water sources underground and may affect public health upon ingestion. The health of locals who drink water from sources near these areas and the steel company may be severely harmed by these PAHs. Therefore, the purpose of this investigation is to establish the level of groundwater pollution, the quantity of PAHs, and the appropriateness of borehole water from Orhuwhorun town in Delta State, Nigeria, in comparison to EPA-approved standards.

2. Materials and Methods

2.1 Description of the Study Area

The municipality of Orhuwhorun is situated in Delta State, Nigeria's northeastern Udu Local Government Area. The hamlet is located roughly 15 kilometers from Delta State's commercial hub. The settlements that border the land are all around it. Usieffrun (Urhiephron) is located in the west, Ekete is in the north, Owenrode is in the south, and Gini is in the east. Certain communities, such as Ovwian, Arhagba, and Igbogidi, define limits to Orhuwhorun's southwest and southeast. Presented in Figure 1 is the map of Delta State, Nigeria.

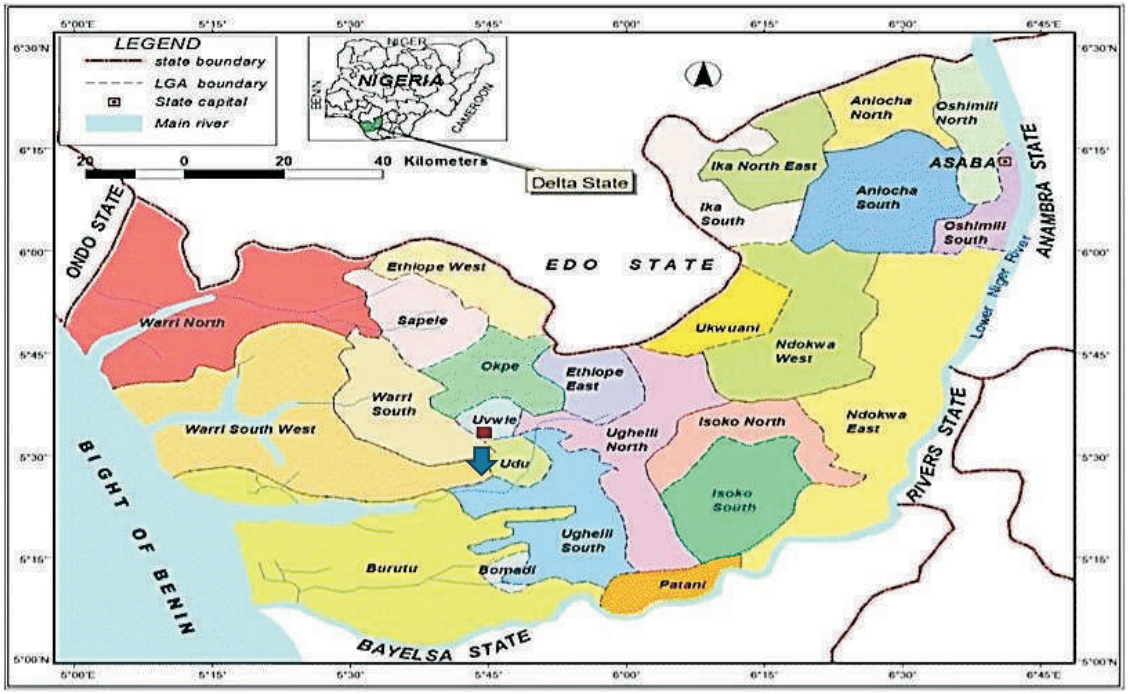

Figure 1: Map of Delta State, Nigeria

2.2 Reagents and standards

All chemicals used in this study were of analytical grade and of the highest purity. The reagents included hexane, dichloromethane, GC-grade alumina (used as a desiccant), concentrated sulfuric acid (H2SO4), organic-free reagent water, and anhydrous sodium sulfate.

2 3 Gathering of samples

Water samples were collected from the study locations over a three -month period (January ,February ,and March)from a 150 -foot -deep borehole in a residential area near the city of Orhuvworhu .The samples were placed in an ice chest immediately after collection to preserve their integrity .Subsequently ,the samples were transferred to pre -labeled and pre -cleaned physicochemical containers for laboratory analysis .

2 4 Extraction

Prior to extraction ,the sample vials were thoroughly cleaned using detergent ,rinsed with water ,and treated with solvent to eliminate any residual impurities .The water samples were filtered using Whatman filter paper to remove particulate matter .The extraction was conducted following EPA Method 3510C with minor adjustments .

Using the liquid -liquid extraction technique ,100 mL of the water sample was vigorously shaken with 50 mL of dichloromethane (DCM) The mixture was then poured into a separatory funnel and allowed to sit for three minutes ,with periodic venting to release excess pressure .After a minimum of 10 minutes ,the organic layer (DCM extract)and aqueous phase were separated ,and the DCM extract was collected in a 250 mL Erlenmeyer flask .The extraction process was repeated with an additional 60 mL of DCM ,and the extracts were combined in the Erlenmeyer flask .

The combined organic extracts were concentrated to approximately 2 mL by gently heating under a nitrogen stream using a rotary evaporator . A glass chromatographic column was prepared by packing it with activated silica gel ,1 g of Na_2SO_4 ,and DCM to condition the column .The concentrated extracts were dissolved in 5 mL of n -hexane ,loaded onto the column ,and eluted with 50 mL of n -hexane .The eluents were then reconcentrated to 0 5 mL using the rotary evaporator and prepared for gas chromatography (GC)analysis .

2.5 Water PAHs evaluation using GC-MS

A 10 mL polypropylene cartridge column was carefully packed with 1 gram of silica gel that had been activated at 130 °C for one hour. The cartridge was then preconditioned with 6 mL of dichloromethane (DCM). The concentrated extract was loaded onto the column, and the eluate was collected in a 50 mL pear-shaped flask placed beneath the column. The column was further eluted with an additional 10 mL of DCM. The

combined eluate was then concentrated using a rotary evaporator set to 38 °C until it was nearly dry. The residue was redissolved in 1 mL of methanol (CH_3OH) and transferred to a 2 mL standard vial for gas chromatography (GC) analysis.

For the GC analysis, capillary columns coated with VF-5 (30 m + 10 m EZ Guard column, 0.25 mm internal diameter, 0.25 µm film thickness) were used. The injector and detector temperatures were set to 250 °C and 300 °C, respectively. The oven temperature program was as follows: hold at 60 °C for 2 minutes, ramp at 25 °C/min to 180 °C and hold for 1 minute, then ramp at 5 °C/min to 310 °C. Helium was used as the carrier gas at a flow rate of 1.4 mL/min. The inlet mode was splitless with a linear velocity of 30 cm/sec and a detector make-up gas flow rate of 29 mL/min. The concentrations of the analytes were expressed in mg/L, as reported by Okechukwu et al. (2021).

3. Results and Discussion

3.1 PAHs concentration

Table 1 displays the concentration results of the water sample analyzed. Figure 2 displays the GC/MS analysis; the minimal detection limit was 0.001 ppm. Additionally, the mean levels were computed.The water sample recorded different mean concentrations of 16 priority PAH congeners as shown in Table 1. The result indicates the presence of Flu which is evidence that the water sample is contaminated with PAHs (Liapis et al., 2008). The US EPA Standard Limit for each of the different PAH congeners is also displayed.

Table 1: Mean PAHs concentration obtained from study area.

	Concentration (mg/L)	%	US EPA (ppm)
Nap	34.073	4.153	0.002
AcPy	51.867	6.322	0.002
Ace	51.003	6.217	0.002
Flu	34.347	4.187	0.002
Phe	36.896	4.497	0.002
Ant	26.815	3.269	0.002
Flu	46.109	5.620	0.002
Pyr	55.636	6.782	0.002
Chry	22.729	2.770	0.002
BaA	28.114	3.427	0.002
BbF	71.036	8.659	0.002
BkF	96.079	11.711	0.002
BaP	33.544	4.089	0.002
IndP	102.109	12.446	0.002
BghiP	29.043	3.540	0.002
DahA	101.002	12.311	0.002

The mean concentration of the S16 PAHs examined, and their corresponding US EPA standard limits are displayed in the report from Table 1. PAHs concentrations in the water sample collected ranged from 29.043 mg/L to 102.109 mg/L. From the results, Nap, which is a low-molecular-weight PAH, indicates a high concentration of about 34.07 mg/L. This may be due to leachates and surface water runoffs (Aralu et al., 2023; Peretomode & Eyenubo, 2023). The calculated mean concentration of the sample shows IndP having the greatest mean concentration, and the lowest mean concentration of PAH is related to Chry, which has been found to be a very insistent PAH and has been linked to the incomplete combustion of fuel (Peretomode & Eyenubo, 2023). It has been discovered that lower molecular weight PAHs occur primarily in lower concentrations due to dissolution and high volatility (Onyidinma et al., 2021). The average mean concentration ranged from 22.729 to 102.109 mg/L. The outcome showed that every S16 PAH found was above the various 0.002 ppm US EPA regulatory limits. The high concentration of PAHs recorded can be related to the prevalence of mechanical and industrial activities, which are major sources of livelihood in the location. There may be fuel and petroleum products spillage from transport vehicles and storage facilities clustered in the location and leachates (Aralu et al., 2023). Similar high concentrations have been established in earlier studies (Ogunfowocan et al., 2003; Opuene, 2009). In general, the total concentration of PAHs found in this study is far greater than the 0.00005 mg/L WHO recommended limit. Furthermore, the reported individual PAH values are above the US EPA's threshold of 0.002 parts per million, suggesting that the water is contaminated. The US EPA regulation limit for drinking water was exceeded by the concentration of S16 PAH congeners found in groundwater, referring to a similar study reported by Mojiri et al. (2019). The Wang et al. (2021) research, which discovered low to slightly moderate amounts of PAH in their study region, is contradicted by our investigation. This study found a value between 0.12 and 0.63 ppm, but their study found a concentration of 0.0003 ppm for PAHs. has demonstrated that the ratios of the different compounds, based on the characteristics of their composition and distribution pattern related to their source, can be used to identify the sources (pyrogenic or pyrolytic) of PAHs (Kafilzadeh et al., 2011; Daniel, 2015; Kafilzadeh, 2015).

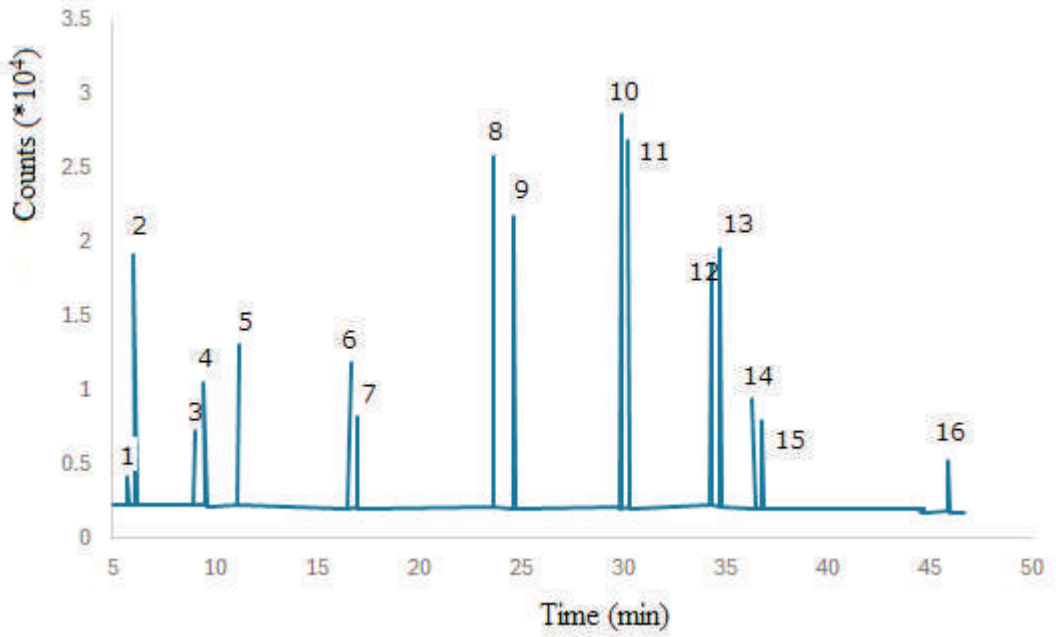

Figure 2. Chromatogram of water sample with (1) Pyr, (2) Nap, (3) BaA, (4) Acy, (5) BghiP, (6) DahA, (7) Flu, (8) BkF, (9) BbF, (10) Flu, (11) Ant, (12) Chry, (13) BaP, (14) Phe, (15) IndP, and (16)Ace

3.2 Distribution of PAHs

Table 2 displays the distribution patterns of the 16 PAH homologs by ring size found in the water sample from Orhuwhorun town in Delta State, Nigeria. The analysis's findings indicate that the sample site contains the poly aromatic hydrocarbon rings 2R, 3R, 4R, 5R, 6R, and 7R (Table 2).

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Table 2: Distribution of PAHs

Rings	Percentage (%)
2R	6.154499
3R	14.38924
4R	10.92741
5R	26.51334
6R	9.392314
7R	32.62318

The PAHs found in the sample were discovered to include both low- and high-molecular-weight PAHs (Araru et al., 2021; Adeniji & Okoh, 2019). The percentage distribution of PAHs with two rings is 6.15%, with fluoranthene as a major component. The three rings were 14.39% with fluorine and phenanthrene as the major components, four rings were 10.93% with pyrene as the major component, five rings were 26.51% with benzo(a)pyrene, benzo(k) fluoranthene, and benz(b)fluoranthene as the major components, and six rings were 9.39%. The maximum PAH value recorded was 7 rings being 32.62%, as illustrated in Figure 2. The column (Figure 2) clearly shows that 3-ring, 4-ring, 5-ring, and 7-ring were the dominant contributors to PAH in the investigated location. Similarly, 2-ring and 6-ring were the least and contributed the least to the total sum of the 16 PAH. The results from Table 1 show that concentrations of low-molecular-weight PAH in the sample are lower than concentrations of PAH with high molecular weights. This signifies that more PAHs with high molecular weights are present in the samples than PAHs with low molecular weights. This result is consistent with a similar report by Edet et al. (2021) but differs from other studies that have been reported by Aderonke et al. (2020) and Adedosu et al. (2015). Whereas the authors (Aderonke et al., 2020) attributed the dominant high molecular weight PAHs to solid wastes from dump sites and the partial combustion of organic materials, low molecular weight PAHs were attributed to oil spills.

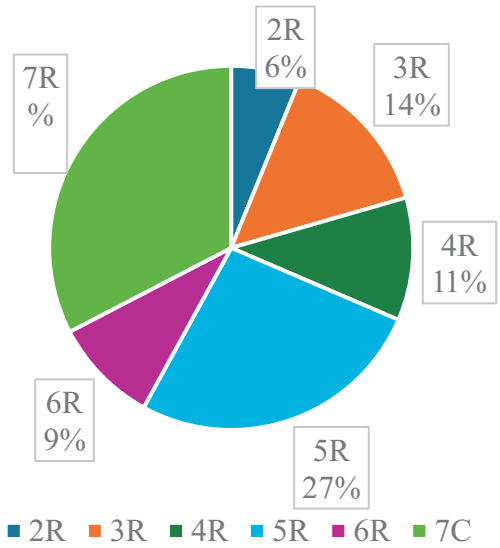

Figure 3: Percentage distribution of PAH in Borehole Water Based on Ring Numbers.

The concentration of the S16 PAHs in the sample sites has been found to be above the US EPA regulation limit. This implies that the samples are polluted, and exposure (intake) via the gastrointestinal tract may result in both non-carcinogenic and carcinogenic health risks (Olawoyin et al., 2012; Omokpariola & Omokpariola, 2021). Reports have shown that carcinogenic PAHs (BaA, BaP, BbF, BkF, Chry, and DahA) are more prominent in parts of the body such as the bladder, lungs, and liver (ATSDR, 2023). Research (Olawoyin et al., 2012; Okechukwu et al., 2021) has also shown that children are more at risk than adults; however, PAHs ingested via the human body lead to degradation of the body tissues and a lack of proper function of the nervous system (Iwegbue et al., 2016).

Conclusion

In Nigeria, the contamination of water by PAHs is turning into a serious public health issue. Because of the worryingly high amount of PAHs, this study has shown that home water in a few chosen areas of Orhuwhorunin Delta State, Nigeria, is not generally safe or suitable for consumption. Moreover, the samples' PAH concentrations are substantially beyond the permitted limits. Anthropogenic activities in the Orhuwhorun community's neighborhood are the primary cause of the PAHs that have been found, and this has contaminated groundwater. These potentially harmful chemicals' existence justifies initial worry and necessitates a more thorough periodic evaluation of PAHs at the sites. Groundwater needs to be regularly monitored by putting an EMP in place. To reduce potential contamination and health problems, Orhuwhorunin Delta State, Nigeria, has to regularly check its groundwater by adopting an EMP. The regulatory agency should also make sure that industries check for environmental compliance.

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