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# Assessment of Heavy Metals in Water and Fishes of Oyo Field and Ilaje Coastal Waters, Ondo State, Nigeria

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**ABSTRACT**

The study, aimed at assessing comparative concentration of heavy metals at the shoreline and about 70 km offshore, determined levels of heavy metals (Pb, Cd, As, Cr and Hg) in water and Tilapia bravemanus at the shoreline of Ilaje Local Government Area of Ondo State, Nigeria (SP1) using AAS technique. Same metals were assessed in water and Euthynnus alleteratus at Oyo Oil Field, about 70 km offshore of Ondo State (SP2). Trends of heavy metals in water and fish at SP1 were Hg > Cd > Cr > As > Pb and Hg > Cd > Cr > As > Pb respectively. While trends in water and fish at SP2 were Cr > Hg > Cd > As > Pb and Cr > Cd > As > Hg > Pb respectively. Bioaccumulation factor (BAF) in SP1 ranged from 217.43 to 1773. In SP2 it ranged from 31.34 to 331. Heavy metal contamination in water at SP1 was higher than at SP2. Similarly, heavy metal bioaccumulation in Tilapia brevimanus was higher than in Euthynnus alleteratus. Based on the outcome of the study, Tilapia brevimanus and Euthynnus alleteratus can be used as bio-indicators of heavy metal contamination in SP1 and SP2 respectively.

## 1. INTRODUCTION

Water is an essential requirement for the sustenance of life. Many studies have been conducted, and more are still being conducted, to improve sustainable management of water resources. Most of the studies are carried out to determine the level of contaminants in water and how to improve its suitability for various uses [1-2]. Increasing contamination of water bodies through anthropogenic activities is becoming a global challenge [2-4]. Toxic heavy metals are among persistent contaminants of water bodies. They are distributed in the environment through natural processes such geological weathering, atmospheric deposition or through anthropogenic activities such as agricultural, municipal and industrial discharges [5]. Once introduced into the environment, they migrate through food chain [6].

Recent threat to coastal waters is oil and gas pollution. The major activities include drilling, exploration and production, and all these activities introduce toxic metal-contaminated wastes into the environment. Produced water is the largest waste generated by the oil industry, and it contains various heavy metals, which, unlike hydrocarbons, are non-biodegradable [7-8]. According to the Directorate of Petroleum Resources (DPR) in Nigeria, about 2, 571,113.90 barrels of crude oil were spilled on the environment between 1976 and 1988, with estimated 17.2 billion $m^3$ of natural gas flared annually as a result of oil and gas production activities [9-11].

The fate and transport of these heavy metals often involve aquatic environment where they bio-accumulate and bio-magnify, especially metals with high uptake and low elimination rates in the tissues of fish [12-13]. As the metals migrate through the food chain, they end up in humans through consumption of the aquatic organisms [14]. Although some of the heavy metals, such as, Zn, Cr, Co, Mo and Fe play important physiological roles in the body of living organisms [15], others are nonessential metals, also known as xenobiotic or foreign elements, with no proven biological function. They include Al, Cd, Hg, Sn and Pb [16].

The main factors affecting bioaccumulation of heavy metals in fish are concentration in water and period of exposure. However, environmental factors such as pH, alkalinity, temperature, hardness, oxygen concentration and dissolved organic matter may also affect bioaccumulation and toxicity in fish [17]. In some cases, the concentrations are not high enough to pose apparent toxicological effects such as sickness or death but can be remote causes of low production, leading to population decline and eventual extinction [18].

The bioaccumulation factor (BAF), which is basically the ratio of centration of heavy metals in the body of an organism to concentration in the ambient environment, helps to determine affinity of heavy metals to tissues of the organisms, and this increases the chances of diseases. Many diseases have been associated with toxic heavy metals. Arsenic exposure has been associated with clinical conditions such as cardiovascular and peripheral vascular disease [19]. Pb ions have been reported to compete with essential metallic cations for binding sites, inhibiting enzyme activity, or altering the transport of essential cations such as calcium [20]. Decreases in olfactory function has been attributed to airborne cadmium particulate matter, especially in the work environment [21]. Cr is one of the essential heavy metals, however high concentration of it has been associated with adverse health effects. Another important factor responsible for adverse effect of Cr is the oxidative state. The hexavalent form ($Cr^{+6}$) has been reported as the most toxic species. Both the hexavalent and trivalent

($Cr^{3+}$) are considered important environmental pollutants in water sources [22]. Hg is one of the most toxic heavy metals because of its predisposition to methylation. Methyl mercury compounds are the most toxic mercury compounds. Dimethyl mercury has an oral $LD_{50}$ of about 0.08 mg/kg for rat. An estimated exposure of 1 mg/m$^3$ for 3 months has resulted in death in humans [23].

A study by Olusola and Festus [3] on Ondo coastal waters determined Cr, Cd, Pb, Cu, Zn and Ni in water, fish and sediments. Similarly, study by [6] around the same area determined Pb, Zn, Cu, Cr and Ni in water and sediments. Both studies were restricted to the shoreline and estuary, and Hg, a toxic pollutant in aquatic environments was not considered. And only two xenobiotic heavy metals (Cd and Pb) were included in the studies. This may be due to the difficulty of analysing metals such as As and Hg.

Heavy metals contamination of water bodies remains a global environmental challenge. Apart from the toxic effects, they are persistent pollutants that are most times only detected after causing harm to living organisms. As stated above, previous studies on heavy metals at Ondo coastal water were restricted to the estuaries and shoreline, and the levels of mercury were not considered. This study aims to assess heavy metal levels deeper offshore, and equally determine levels of mercury.

## 2. MATERIALS AND METHODS

### 2.1 Study area

Oyo Field is about 70 km off the coast of Ilaje Local Government Area of Ondo State, Nigeria, with a water depth of about 410 m. It lies approximately between latitude 5°30' – 6° 15' N and longitude 3° 50' – 4° 35' E. Ugbonla is a coastal town of Ilaje Local Government Area. Ilaje has a total land area of 2,300 square km, and a population of 290,615, according to 2006 NPC census [24], and lies within 5°45' - 6°15' N and 4°30' - 5°00' (Figure 1). The activities in the area include fishing, peasant farming, artisan trade etc. Though not directly linked to the local economy, offshore oil exploration is the main industrial activity and a potential point source pollution of the aquatic environment.

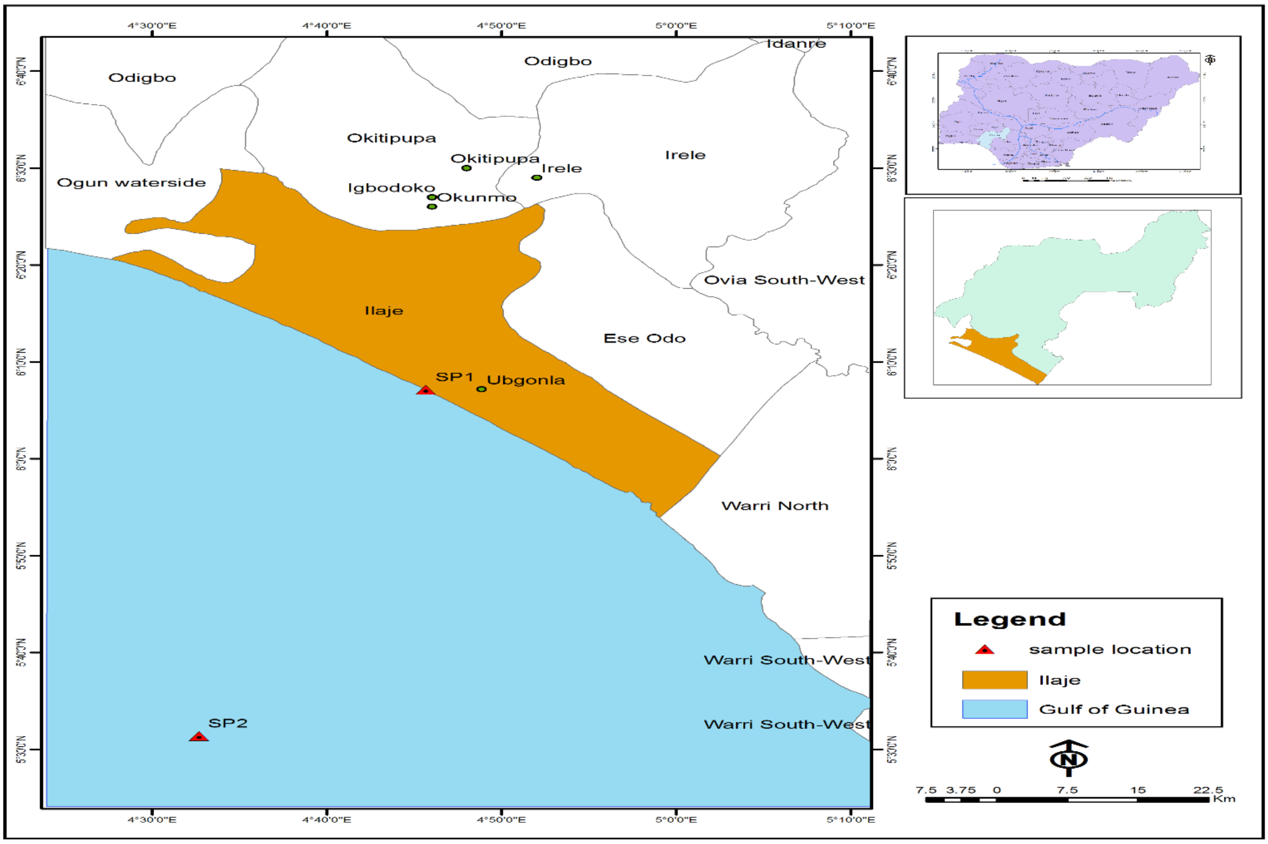


**Figure 1.** Map of Nigeria showing Ondo State, Ilaje LGA and sampling points

According to a survey by Olufayo, et al. [25], fishing is the commonest occupation in the study area, with 32 %, followed by sand dredging with 26.3 %, farming 15.1 %, boat operators 8.8 %, thrift collectors 8.8 %, civil servants 4.5 %, and traders 4.5 %. However, the survey did not consider the unemployed and the section of the population which may be involved in other sources of livelihood.

The water at both locations is used basically for fishing and navigation, which necessitates the determination of bioaccumulation.

### 2.2 Sampling and sample analysis

All glassware and porcelain were soaked in 3 % $HNO_3$, washed 24 hours prior to use, rinsed with distilled water, oven-dried and allowed to cool to room temperature. Water samples were collected with a 500 mL beaker at Ugbonla Sampling Point (SP1) and Oyo Field Sampling Point (SP2) between

10cm to 30cm below the water surface. At SP1 10 samples were collected at different locations along the shoreline at interval of about 100 metres covering a distance of about 1 – 2 km and pooled into a representative sample. According to [26], samples were collected at the operational area of SP2, 500 m upstream and 500 m downstream, then pooled to make a representative sample. The water samples were acidified by addition of conc. $HNO_3$.

All samples were stored in icebox, transported to the laboratory, stored in a refrigerator and analysed within 3 days. Exactly 100 mL of acidified sample was measured in a 250 mL conical flask and 5 mL of conc. $HNO_3$ was added and covered with watch glass. The content was heated on a hot plate at 60 °C for 30 minutes and then cooled to 45 °C. The flask and watch glass were rinsed with distilled water and filtered into a clean conical flask, then the filter was rinsed with 3 mL distilled water. The filtrate was transferred into a 100 mL volumetric flask, and the conical flask rinsed with 5 mL distilled water into the volumetric flask. The sample was made up to 100 mL mark with distilled water, agitated to mix thoroughly and analysed using FS240AA Agilent Atomic Absorption Spectrometer.

At SP1, four mature samples of *Tilapia brevimanus* with an average length of 19 cm and average weight 92.4 g were bought from fishermen by the coast. They were stored in icebox and transported to the laboratory to retain their freshness. At SP2 two mature *Euthynnus alleteratus* with an average length of 34.5 cm and average weight of 468.2 g were caught with hook and line, stored in icebox filled with ice and transported onshore by helicopter. The fish were measured with measuring ruler to determine length, and weighed with a calibrated scale to determine weight. The gills, intestines and muscle were dissected and each organ was oven-dried to a constant weight at 105 °C. Each dried part was crushed with mortar and pestle to fine sizes, then stored in labelled plastic containers in a desiccator until digestion. Approximately 1 g of the crushed sample was weighed into a round bottom flask and 10 mL conc. $HNO_3$ and 2 mL $HClO_4$ were added. After digestion at 60 – 65 °C, 50 mL distilled water was added and filtered. More distilled water was used to rinse the filter paper to make up to 100 mL. The digest was stored in pre-cleaned polypropylene bottles until analysis using FS240AA Agilent Atomic Absorption Spectrometer. The standard was run for the working curves, followed by the blank to overcome instrument drift. Heavy metal analysis was carried out using FS240AA Agilent Atomic Absorption Spectrometer. The sample was thoroughly agitated and 100 mL transferred into a glass beaker of 250 mL. The sample was aspirated into oxidising air/acetylene, nitrous oxide/acetylene flame. The metal concentration was read in mg/L from the extracted standard calibration curve. The analyses were conducted in triplicate for each element and the average values were calculated.

## 3. RESULTS AND DISCUSSION

Mean concentrations of heavy metals (Pb, Cd, As, Cr and Hg) in SP1 and SP2 are presented in Table 1 side-by-side with NESREA limits. In Table 2 concentrations of heavy metal in organs (gills, intestine and muscle) of *Tilapia brevimanus* from SP1 are presented. Similarly, concentrations of heavy metals in the organs (gills, intestine and muscle) of *Euthynnus alleteratus* (SP2) are presented in Table 3. Concentration of heavy metals in SP1 and SP2 are presented side-by-side with concentrations in water and the bioaccumulation factor (BAF) in Table 4. Trend of Heavy Metals in Water and Fish Species is presented in Table 5. Trend of Heavy Metals in Fish Organs from SP1 and SP2 is presented in Table 6. Trend of organ accumulation of heavy metal in SP1is presented in Table 7 and 8. Graphical presentation of the results is on Tables 2, 3 and 4.

**Table 1.** Mean concentration of heavy metals in SP1 water compared with NESREA limits

| Heavy metal | SP1 water (µg/L) | SP2 water (µg/L) | NESREA limits (µg/L) |
|---|---|---|---|
| Pb | 0.18 | 0.24 | 10 |
| Cd | 70 | 0.48 | 5 |
| As | 12 | 0.32 | 50 |
| Cr | 18 | 0.72 | * |
| Hg | 219 | 0.54 | 1 |

**Note:** * $Cr^{+6}$ = 1 µg/L, $Cr^{+3}$ = 500 µg/L

**Table 2.** Comparative concentration of heavy metals in organs of *Tilapia brevimanus* (SP1)

| Heavy metal | Concentration (µg/L) | | |
|---|---|---|---|
| | Muscle | Gills | Intestine |
| Pb | 0.12 | 0.31 | 217 |
| Cd | 103 | 114 | 166 |
| As | 67 | 72 | 86 |
| Cr | 60 | 121 | 81 |
| Hg | 227 | 1264 | 282 |

**Table 3.** Comparative concentration of heavy metals in organs of *Euthynnus alleteratus* (SP2)

| Heavy metal | Concentration (µg/L) | | |
|---|---|---|---|
| | Muscle | Gills | Intestine |
| Pb | 0.22 | 0.12 | 31 |
| Cd | 62 | 67 | 118 |
| As | 93 | 56 | 85 |
| Cr | 101 | 111 | 119 |
| Hg | 0.21 | 157 | 0.16 |

**Table 4.** Bioaccumulation of heavy metals

| Heavy wetal (µg/L) | SP1 water | BAF | SP2 water | BAF |
|---|---|---|---|---|
| Pb | 0.18 | 1207.94 | 0.24 | 130.58 |
| Cd | 70 | 5.47 | 0.48 | 514.58 |
| As | 12 | 18.75 | 0.32 | 731.25 |
| Cr | 18 | 14.56 | 0.72 | 459.72 |
| Hg | 219 | 8.10 | 0.54 | 291.43 |

This study revealed higher concentrations of heavy metal at SP1, which is by the shoreline, than SP2, about 70 km offshore. Pb had the least concentration at both locations while Hg was highest at SP1 and Cr highest in SP2 (Table 1). The nearshore water is predisposed to contaminant from erosion of coastal rivers that empty into the sea and other anthropogenic activities around the coastal area, while the offshore receives contaminants from dry and wet precipitation, ocean currents [27]. In this study area (Oyo Oil Field) there is also a likelihood of point source contamination from oil exploration. According to [28], dilution of contaminants increases seaward from the coast because of the depth of the water. However, this can only be true in the case of SP1 and SP2 if the two areas were receiving the same quantity of contaminants. On the other, heavy metal contamination from oil exploitation may be lower than contamination from natural processes and anthropogenic activities at the coastal area. Concentrations at both points were within NESREA limits except for Cd and Hg at SP1. Results of previous studies in Ugbonla and around Ilaje axis revealed higher concentrations of some heavy metals than in this study. [29], working on water quality of Oil Producing Areas of Ilaje, Nigeria, reported Pb 160 μg/L, Cd 20 μg/L and AS 1530 μg/L at Ugbonla. [3] reported Pb 570 – 790 μg/L, Cd 80 – 210 μg/L, Cr 310 – 340 μg/L for coastal waters of Ondo State, Nigeria. [30] reported Pb 2900 – 3400 μg/L, Cd 3700 – 3800 μg/L, Cr 4500 – 6900 μg/L. [31] reported Pb 1480 – 2290 μg/L, Cd 350 – 480 μg/L, As 20 – 40 μg/L and Cr 390 – 560 μg/L.

**Table 5.** Trend of heavy metals in water and fish species

| Water | Trend of heavy metals |
|---|---|
| SP1Water | Hg > Cd > Cr > As > Pb |
| SP1 Fish | Hg > Cd > Cr > As > Pb |
| SP2 Water | Cr > Hg > Cd > As > Pb |
| SP2 Fish | Cr > Cd > As > Hg > Pb |

**Table 6.** Trend of organ accumulation of heavy metal in SP1

| Heavy metal | Trend of accumulation in organs |
|---|---|
| Pb | Intestine > Gills > Muscle |
| Cd | Intestine > Gills > Muscle |
| As | Intestine > Gills > Muscle |
| Cr | Gills > Intestine > muscle |
| Hg | Gills > Intestine > muscle |

**Table 7.** Trend of organ accumulation of heavy metal in SP2

| Heavy Metal | Trend of accumulation in organs |
|---|---|
| Pb | Intestine > Muscle > Gills |
| Cd | Intestine > Gills > Muscle |
| As | Muscle > Intestine > Gills |
| Cr | Intestine > Gills > Muscle |
| Hg | Gills > Muscle > Intestine |

**Table 8.** Trend of heavy metals in fish organs from SP1 and SP2

| Fish organ | Trend of heavy metals | General Trend |
|---|---|---|
| SP1 Muscle | Hg > Cd > As > Cr > Pb | |
| SP1 Gill | Hg > Cr > Cd > As > Pb | Hg>Cd>AS>Cr>Pb |
| SP1 Intestine | Hg > Pb > Cd > As > C | |
| SP2 Muscle | I Cr > As > Cd > Pb > Hg | |
| SP2 Gill | Hg > Cr > Cd > As > Pb | Cr>Cd>AS>Hg>Pb |
| SP2 Intestine | Cr > Cd> As > Pb > Hg | |

Different organs of *Tilapia brevimanus* and *Euthynnus alleteratus species* exhibited varying levels of heavy metals bioaccumulation (Tables 2, 3 and 4). Hg (1264 μg/kg) was the highest concentration of heavy metal in the gills of *Tilapia brevimanus* while Pb (0.12 μg/kg) was the least, in the muscle. In SP1 gills had 54.39 % of total heavy metal load, intestine 29.09 % and muscle 15.98 %. Heavy metals distribution in SP1 was as follows: Pb (intestine 99.80 %, gills 0.14 %, muscle 0.06 %), Cd (intestine 43.34 %, 29.77 %, muscle 26.89 %), AS (intestine 38.22 %, gills 32 %, muscle 29.78 %), Cr (gills 46.18 %, intestine 30.92 %, muscle 22.90 %), Hg (gills 71.29 %, intestine 15.91 %, muscle 12.80 %). In SP2 Cr was the most concentrated heavy metal in *Euthynnus alleteratus*, and the intestine of the fish had the highest concentration, while Pb was the least concentrated, with gills showing the least concentration (Figure 4). Gills had 39.08 %, intestine 35.29 % and muscle 25.63 % of the heavy metal load.

Heavy metals distribution in SP2 was as follows: Pb (intestine 98.92 %, muscle 0.70 %, gills 0.38 %), Cd (intestine 47.77 %, gills 27.13 %, 25.10 %), As (muscle 39.74 %, intestine 36.33 %, gills 23.93), Cr (intestine 35.95 %, gills 33.54%, muscle 30.51 %), Hg (gills 99.76 %, muscle 0.14 %, intestine 0.10 %). At both locations Pb and Cd were concentrated in the intestine while Hg was most in the gills.

Concentration of Hg in the gills and Pb in the intestine of both fishes suggests feeding habit may be a factor in the bioaccumulation. Hg, which turned out to have the highest concentrations in both water and fish at the SP1, was not determined in the previous studies of Ondo coastal waters reviewed in the course of this research. This may be due to the difficulty of getting an AAS machine capable of determining Hg conc. [30] reported heavy metal presence in the organs of *Oreochromis niloticus, Synodonthis* sp., and *Clarias gariepinus,* so did [3] in the organs of *Pentanemus quinquarius, Pseudoltolithus senegalensis, Trichirus lepturus, Plectorhynchus meditarraneus and Seudotolithus typus,* indicating a history of accumulation of heavy metals in the body tissues of biota of the coastal waters of Ondo State. However, samples for these studies were drawn only at the shoreline. In this study samples were also drawn at about 70 km offshore where water depth is about 400 meters. This gives an idea of comparative concentration of heavy metals in water and biota at the shoreline and 70 km offshore.

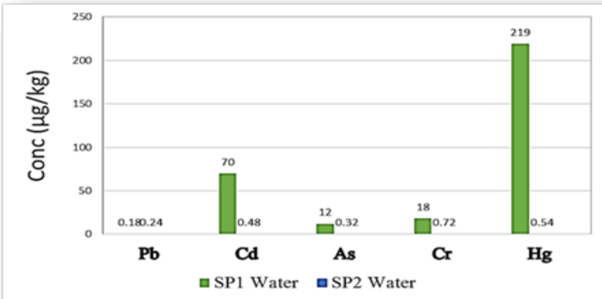


**Figure 2.** Comparative Concentration of Heavy Metals in SP1 Water and SP2 Water

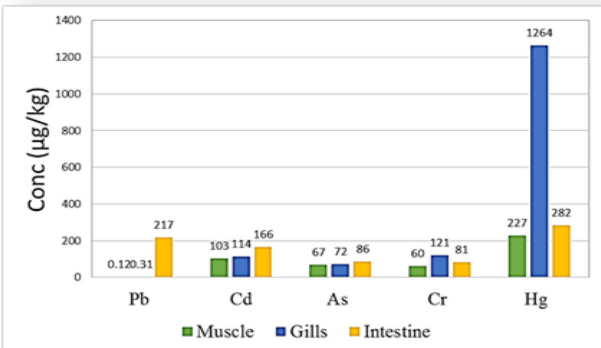


**Figure 3.** Comparative Concentration of heavy metals in muscle, gills and intestine of *Tilapia brevimanus* (SP1)

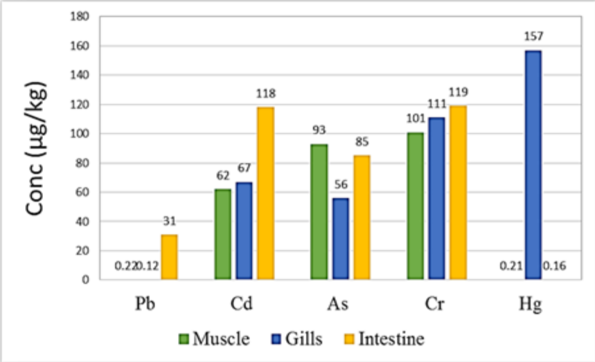


**Figure 4.** Comparative Concentration of heavy metals in muscle, gills and intestine of *Euthynnus alleteratus* (SP2)

## 4. CONCLUSION AND RECOMMENDATIONS

The concentration of heavy metals at the shoreline was higher than at Oyo Field, about 70 km from the shoreline, with corresponding higher concentrations in *Tilapia brevimanus* than in *Euthynnus alleteratus*. This suggests that the pollution may be from anthropogenic activities onshore.

Higher concentration of heavy metals in SP1 water may be a contributory factor to higher concentration in *Tilapia brevimanus*. Feeding habit may be another factor. Concentration of heavy metals in the fish tissues showed that there was bioaccumulation of the heavy metals. In both locations there was higher concentration of heavy metals in the gills and intestines than in the muscles of the fishes; this may be attributed to the difference in physiological roles in fish metabolism played by the three organs. High concentration of Hg in the gills of *Tilapia brevimanus* may be an indication of active contamination along the coastal. The novelty of this study is in two findings: heavy metal levels were lower deeper offshore (about 70 km from the shoreline) than at the shoreline, and Hg, which had never been determined was the most concentrated at the shoreline.

Further studies are needed to determine the wider level of heavy metal contamination. These should include sediments and other relevant biota, covering all the seasons, to ascertain a more holistic pollution status of the water, and give a better basis for comparison of heavy metal load of the two locations considered in this study.

This study therefore concludes that heavy metal contamination at the shoreline is higher than 70 kilometres offshore, and *Tilapia brevimanus* and *Euthynnus alleteratus* are good bio-indicators for monitoring heavy metal contamination.


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## NOMENCLATURE

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| ATSDR | Agency for Toxic Substances and Disease Registry |
| BAF | Bioaccumulation Factor |
| DPR | Department of Petroleum Resources |
| EGASPIN | Environmental Guidelines and Standards for the Petroleum Industry in Nigeria |
| $LD_{50}$ | Lethal Dose |
| NBS | National Bureau of Statistics |
| NESREA | National Environmental Standards and Regulation Enforcement Agency |
| NOAA | National Oceanic and Atmospheric Administration |
| NPC | National Population Commission |
| SP1 | Sampling Point 1 |
| SP2 | Sampling Point 2 |

## Greek symbol

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|---|---|
| μ | Microgram, μ/L |