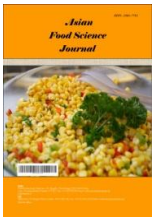

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Assessment of the Bioaccumulation of TPH and Heavy Metals in Tissue of Selected Fishes' Species Caught from Stream near a Petroleum Processing Plant

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Authors' contributions

This work was carried out in collaboration among both authors. Both authors read and approved the final manuscript.

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ABSTRACT

This article investigated the bioaccumulation of total petroleum hydrocarbon (TPH) and selected heavy metals in the tissue of selected fishes' species from a stream near a petroleum processing plant in Rivers State, Nigeria. The concentration of TPH and Ni, Mn, Fe, Zn, Pb, Cr and Co in the fish species' tissue were measured using standard methods. The TPH concentrations in the fish species were generally high (1.97 to 52.84 mg/kg), greater than the European Union threshold limit of 2µg/kg. *Gobius* sp and *Periophthalmus* sp recorded the highest TPH content: 52.84 mg/kg and 28.07mg/kg levelsrespectively. Tilapia sp and *Sardinella* sp recorded the least and highest concentration of Manganese (0.61 mg/kg and 5.90 mg/kg respectively). The levels of Fe content in the fish tissue were higher than all other metals: 8.27 mg/kg in *Gobius*sp to 129.7 mg/kg in *Sardinella* sp. Zn: 4.39 mg/kg in Mugilidae to 11.84 mg/kg in *Gobius* sp while Co:<0.02 mg/kg in *Tilapia* sp and 1.51mg/kg in *Calinectis* sp. The concentrations of Cd:0.24 mg/kg in *Pomadasys* sp to 0.56 mg/kg *in Gobius* sp. These values are relatively high because of the metal's high toxic

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response factor of 30. The levels of Pb:<0.03 mg/kg in *Tilapia* sp and Carangidae to 2.69 mg/kg in *Calinectis* sp. *Gobius* sp recorded the highest level (3.36 mg/kg) of Ni. The concentrations of Co, Cd, Pb, and Ni in consumable fish are of great concern because of their high toxic response factors. These results implied that the petroleum processing activities (legal and illegal) are negatively impacting aquatic habitats/ food chains. This calls for caution in the consumption of the fish from this area, the audit of the legal petroleum processing facilities and stoppage of the illegal operations.

Keywords: Bioaccumulation; TPH; heavy metals; fish tissue; petroleum processing and toxic response factors.

1. INTRODUCTION

The petroleum industries' (upstream, midstream, and downstream) operations have been reported to generate a lot of wastes including large quantities of liquid waste. These liquid wastes are generally from the high volume of water employed at the various stages of petroleum processing especially the refining stage. It is widely reported that petroleum production processes generate wastewater higher than the volume of crude oil being produced and this effluent contains hydrocarbons (organics), phenols and dissolved minerals as well as some heavy metals. The organics include oil and grease, total hydrocarbon content (THC) and total petroleum hydrocarbon (TPH) as well as benzene, toluene, ethylbenzene, and xylene (BTEX). Oil and grease for instance, the aggregate of the hydrocarbon content of sampled water, soil, or sediment prevents oxygen absorption, leading to the decline of dissolved oxygen levels in water and this may inhibit aquatic flora and fauna. It also causes direct damage to plants and animals [1]. These contaminants could be toxic or carcinogenic especially when ingested [2] and [3]. Generally, contaminants from petroleum processing are harmful to both flora and fauna as well as other components of the environment especially if not treated or inadequately treated. These pollutants are mostly associated with varieties of human and plant diseases such as organ malfunctions, cancer indices and springboard for infectious diseases. Heavy metals and TPH is among the contaminants released into the soils and water bodies from petroleum processing which impacts soil and water quality leading to bioaccumulation in aquatic animals and plants thereby interfering with the food chain and water quality. The total petroleum hydrocarbon (TPH) of liquid is an indicator of the extent or otherwise of hydrocarbon (from petroleum source) contamination of the medium. However, other sources like municipal/industrial waste also contribute to the total hydrocarbons pollution of the environment. While the TPH is known to certainly come from the crude, the heavy metals are suspected to have emanated from various sources including the reservoir rocks [4] metallic parts of the drilling equipment and some drilling fluids additives. It is generally observed that the procreation capacities of aquatic organisms are retarded by their exposure to heavy metals which affects their relative abundance or existence in polluted water bodies [5]. The effluents from the processing operations are channelled to available and nearby surface water as disposal route. Olaji et al. [6] and Al-Hejaje et al. [7] noted with one accord that the riverine areas of oil-rich whose main livelihood are fishing are adversely affected by petroleum production activities due to either spillage or discharge of produced/effluents into the water bodies. However, the quality of the effluent can hardly be ascertained as regulatory agencies may not be technologically equipped enough to carry out in situ tests on the effluent before release to the water bodies. Enuneku et al. [8] noted that all fish takes in petroleum hydrocarbons as food through their presence in the habitat. This contaminant is readily absorbed by aquatic fauna due to lipid solubility. The study traced the presence of TPH in water vis a vis fish to either accidental discharge during oil exploration operations or from industrial by products. TPH contamination of the environment could also be attributed to sabotage, bunkering, or operations of illegal modular refinery popularly called "kpo fire" in the Niger Delta area especially in the Creeks. Umeh [9] and Nkpaa et al. [10] had previously reported the presence and contamination of petroleum hydrocarbon fractions in the area. Generally, consumption of TPH infested fish poses a high health risk to man [11] and [12]. This study therefore focuses on the assessment of the level of TPH and selected heavy metals in fish tissues caught from the stream near a petroleum refining facility in Niger Delta, Nigeria whose effluents are channelled to it and the possible health risks

associated with the consumption of such polluted fish.

2. MATERIALS AND METHOD

2.1 Fisheries Survey and Sampling

Fish sampling was carried out along the ocean/river waterfront around Okrika area of Rivers State, Niger Delta Nigeria. The survey involved inspections/visits of the nearest fishing camp and purchase of catches from the area. Fish samples were collected and identified with the assistance of local fishermen for the common name. Samples were also preserved in a cool box with ice before transfer to the laboratory for analysis.

2.2 TPH Analysis

Samples of the various fish species stored previously for preservation were removed from the ice pack, thawed and washed under running water to free them of accompanying debris. The cleaned fish samples were dissected with a kitchen knife to obtain the component of interest (the muscles). The Organics were extracted using 10g of wet fish in an ultrasonic bath (EPA 3550B) with 20ml of Dichloromethane accompanied with a chemical drying using anhydrous Sodium Sulphate in the extraction bottle. The TPH was determined on an extract using GC/FID in accordance by EPA 8015C.

2.3 Heavy Metals Determination

The samples were air-dried to stop all microbial activities and passed through a 2 mm sieve in readiness for digestion. 20 grams of each species of fish was digested by wet oxidation using an acid mixture of concentrated HNO_3, $HClO_4$, and H_2SO_4 in the ratio 2:1:2 [13]. The concentrations of the metals in the fish digestate were measured using PerkinElmer Atomic Absorption Spectrophotometer (AAS) Model, Analyst 200. This method involved direct aspiration of the sample into an air/acetylene flame. A hollow cathode lamp at a specific wavelength peculiar only to the metal under investigation generated a beam of chromatic light characteristic of the metal under investigation which was absorbed on passage through the flame. The minimum acceptable absorbance from which metal concentrations were calculated as 0.001. Before each metal determination, the AAS was calibrated using prepared serial calibration standards from ACCU STANDARD used as an external calibration standard for quantification of the result. The analytical process was also checked after 10 runs with a QC standard different from the calibration standard to verify equipment and calibration status.

3. RESULTS AND DISCUSSION

3.1 Fish Tissue Analysis

The concentration of total petroleum hydrocarbon and selected heavy metals (Ni, Mn, Fe, Zn, Pb, Cr, and Co) in the tissue (muscle) of selected fish species examined are tabulated in Table 1 below and discussed under the subheadings: TPH and Heavy metals.

Table 1. Concentration of TPH and heavy metals in tissues of fish

Fish Species	Heavy Metals (mg/kg)									TPH mg/kg
	Co	Mn	Cr	Zn	Cd	Cu	Pb	Ni	Fe	
Calinectissp	1.51	4.90	<0.05	6.77	0.33	1.62	2.69	3.25	59.10	5.24
Periopthalmussp	0.13	2.44	<0.05	9.71	<0.01	0.18	0.10	0.42	24.76	28.07
Carangidae	<0.05	2.44	<0.05	9.14	<0.01	0.17	<0.03	0.29	24.76	21.75
Gobiussp	1.47	0.90	<0.05	11.84	0.56	0.25	2.21	3.36	8.27	52.84
Pomadasyssp	1.27	0.96	<0.05	6.81	0.24	0.31	1.97	2.88	15.12	4.27
Dentex sp	1.33	0.96	<0.05	5.15	0.32	0.14	2.12	2.59	17.54	13.15
Lutjanus sp	1.37	1.07	<0.05	8.05	0.25	0.24	1.86	2.90	22.10	8.76
Tilapia sp	<0.02	0.61	<0.05	9.66	<0.01	0.67	<0.03	0.12	15.13	4.37
Sardinella sp – Clupeidae (bonga)	0.04	5.90	<0.05	6.82	<0.01	0.40	0.35	0.48	129.7	1.97
Sphyrinidae	1.34	1.18	<0.05	4.72	0.07	0.11	2.42	2.89	8.88	5.24
Sphyraenasp	1.02	0.68	<0.05	6.76	<0.01	0.49	1.72	2.37	8.71	16.54
Mugilidae	0.83	1.22	<0.05	4.39	<0.01	0.52	1.04	1.92	31.86	16.58

3.2 TPH

The total petroleum hydrocarbon (TPH) concentrations recorded in the species of fish tested were generally high. The values ranged from 1.97 to 52.84 mg/kg and were all greater than the European Union threshold limit of 2µg/kg. The values are higher than those reported by Olaji et al. [6]. *Gobius* sp and *Periophthalmus* sp recorded the highest TPH content with values of 52.84 mg/kg and 28.07 mg/kg respectively. These organisms have benthic lifestyles and are prone to such chemicals in their environment. The least concentration of TPH (1.97 mg/kg) was observed in the Clupeidae sp which has pelagic habits. This compound is carcinogenic. Olaji et al. [6] noted that the TPH concentration in fish is linked to diverse factors including route, exposure intervals and lipid content of tissues which is dependent on the species. Hence, these high levels of TPH may not be holistically linked to the activities of the regulated petroleum processing operations but majorly on other factors like discharges from illegal refining operations ('kpo fire'), accidental discharges, pipe failure, etc.

3.3 Heavy Metals

It has been generally observed that the procreation capacities of aquatic organisms are retarded by their exposure to heavy metals which affects their relative abundance or existence in polluted water bodies [5]. The concentration of Mn varied from 0.61 mg/kg in Tilapia sp to 5.90 mg/kg in *Sardinella* sp – Clupeidae (Bonga). The results also showed that Mn concentrations (2.44 mg/kg) were thesame for *Periopthalmus* sp and Carangidae sp. Mn levels were also observed to have equal values (0.96mg/kg) in Pomadasys sp and *Dentex* sp. Chromium concentrations were generally less than 0.05 mg/kg in all the species of fish examined. This level of Cr bioaccumulation is within an acceptable limit because its low toxic response factor is 2. The levels of Fe content in the tissue of the fish species investigated were generally higher than all other metals with values ranging from 8.27 mg/kg in *Gobius* sp to 129.7 mg/kg in *Sardinella* sp – Clupeidae (Bonga). *Calinectis* sp also recorded high iron content 59.10 mg/kg. The same trend in Mn concentration in *Periopthalmus* sp and Carangidae sp were also observed in Fe contents with values 24.76 mg/kg in both species. The levels of Zn in the fish tissue differed from 4.39 mg/kg in Mugilidae to 11.84 mg/kg in *Gobius* sp. This Zn concentration may not be ranked dangerous because of its low toxic response factor of unity (1). The difference in the accumulation of the metals in all the fish species studied was not far apart, hence, this could be linked to the low presence of the metal in the water habitat. The levels of Co in the fish species examined were between <0.02 mg/kg in *Tilapia* sp and 1.51mg/kg in *Calinectis* sp. Gobius sp, *Pomadasys* sp, and *Dentex* sp recorded 1.47 mg/kg, 1.27 mg/kg and 1.33 mg/kg respectively. The concentrations of Co in*Lutjanus* sp (1.37 mg/kg) and Sphyrinidae (1.34 mg/kg) alongside those of *Calinectis* sp, Gobius sp, *Pomadasys* sp, and *Dentex* sp are worrisome because Co is a highly toxic metal with atoxic response factor of 5.

Cadmium has been reported to be one of the non-essential but most toxic heavy metals with atoxic response factor of 30 [5,14,15]. The concentrations of Cd were 0.33 mg/kg in *Calinectis* sp, 0.56 mg/kg *in Gobius* sp, 0.24 mg/kg in *Pomadasys* sp, *0.32* mg/kg *in Dentex*sp *and* 0.25 mg/kg *in Lutjanus* sp. These values are relatively high because of the metal's high toxic response factor of 30. The Cd content of *Periopthalmus* sp, Carangidae, and *Tilapia* sp as well as *Sardinella* sp – Clupeidae (bonga), *Sphyraena* sp and Mugilidae were below 0.01mg/kg. Cd contamination in man causes damage to the kidney alongside retarded procreation capabilities [16]. The lowest concentration of Pb was <0.03 mg/kg and was found in *Tilapia* sp and Carangidae while the highest concentration (2.69 mg/kg) was recorded in *Calinectis* sp. Pb contents in fish species studied were in the range Carangidae = *Tilapia*sp< *Periopthalmus*sp< *Sardinella*sp – Clupeidae (Bonga) < Mugilidae < *Sphyraena*sp < *Lutjanus*sp < *Pomadasys*sp < *Dentex*sp< *Gobius*sp< Sphyrinidae<*Calinectis* sp. The various concentrations were generally high for consumable fish because of the high toxic response factor of 5 for lead metal. Juberg et al. [17] and Afshan et al. [18] highlighted the effects of lead contamination to include damage to vital organs like the liver, kidneys, and brain. It is also reported to cause reproductive disorders and anemia. Ni concentrations with a toxic response factor of 6 were generally higher than those of Pb in all the fish species investigated. The highest value (3.36mg/kg) was recorded by *Gobius*sp followed by Calinectissp (3.25mg/kg). Ni contents in *Pomadasys*sp (2.88 mg/kg) *Dentex*sp (2.58 mg/kg) and Lutjanus sp (2.90mg/kg) as well as Sphyrinidae (2.89mg/kg), *Sphyraena*sp (2.37mg/kg) and Mugilidae (1.92mg/kg) were

generally high because of the value of the toxic response factor. The copper contents of the fish species studied ranged from 0.11mg/kg in Sphyrinidae to 1.62 mg/kg in *Calinectis* sp. The values were generally low compared to Ni and Pb with similar toxic response factors.

4. CONCLUSION

Twelve predominant fish species obtained from surface water around a petroleum processing facility in Rivers State, Nigeria and tested for bioaccumulation of total petroleum hydrocarbon (TPH) and selected heavy metals (Ni, Mn, Fe, Zn, Pb, Cr and Co) showed that the TPH levels in all the fish species were generally high and greater than the European Union threshold limit of 2µg/kg. This could be linked to the effluents from the processing plants discharged into the water body and crude from the illegal refining operations as well as bunkering activities and accidental discharges resulting from pipe rupture. The concentration of the heavy metals in the fish samples was relatively high. The concentration of Cd in the fish samples is prone to higher risk because of the metal's high toxic response factor of 30.The Zn concentrations may not be ranked dangerous yet because of their low toxic response factor of unity (1) but those of Co, Cd, Pb, and Ni in consumable fish are of great concern because of their high toxic response factors. The petroleum processing activities (legal and illegal) have negatively impacted these aquatic habitats in particular and thefood chain in general. This calls for caution in consumption of the fish from this area, audit of the legal petroleum processing facilities and stoppage of all illegal operations.

DISCLAIMER

The products used for this research are commonly and predominantly use products in our area of research and country. There is absolutely no conflict of interest between the authors and producers of the products because we do not intend to use these products as an avenue for any litigation but for the advancement of knowledge. Also, the research was not funded by the producing company rather it was funded by personal efforts of the authors.

CONSENT

As per international standard or university standard, participant's written consent has been collected and preserved by the author(s).

COMPETING INTERESTS

Authors have declared that no competing interests exist.

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