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(RESEARCH ARTICLE)

# Polyaromatic hydrocarbon profile and health risk assessment of popularly consumed species of fish in Remo Zone, Ogun State, South-West Nigeria

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## Abstract

Polyaromatic hydrocarbon (PAH) compounds are usually introduced into foods through processing methods like smoking, grilling, etc and they have been identified as potential carcinogens. This study was aimed assessing the concentrations of PAHs and evaluating the health risk associated with consumption of 3 main fish species popularly consumed in the study area. Health risk factors like daily dietary intake (DDI), carcinogenic potencies of individual PAHs (B(A)Pteq) and the excess cancer risk (ECR) induced by dietary exposure of smoked fish consumers were examined for 16 PAHs considered as priority pollutants.. The three fish species analyzed in this study: Herring (*Clupea harengus*), Blue Whiting (*Micromesistius poutassou*) and Mackerel (*Scomber scombrus*) samples were extracted by liquid extraction and the concentrations of 22 selected PAHs were analyzed using GC-MS. The cumulative concentrations of PAH22 and PAH16 in the three species of fish are of the order Herring > Blue wilting > Mackerels. The results from the GC-MS analysis showed a significant difference ($p<0.05$) in the PAH concentration detected in the fish samples collected from the five study locations. The DDI for PAH16 in smoked Herring was found to be between 0 and 0.1277 µg/day, 0 and 0.007124 µg/day for mackerel and 0 and 0.07946 µg/day for blue whiting. Most of the ECR values obtained in this work were higher than the $10^{-5}$ guideline and this calls for intense monitoring.

**Keywords:** Polyaromatic hydrocarbons; GC-MS; Herring; Blue whiting; Mackerel; Health risk

## 1. Introduction

Fish has been a major component of human diet as a source of essential amino acids. As a much-cherished delicacy, fish enjoys wide acceptance that cuts across socio-economic, age, religious and educational barriers (Adepoju *et. al.,* 2022). However, the consumption of fish has been a dietary route for many contaminants, pollutants and toxins into human body (Wangboje and Besiru, 2023; Liu *et. al.,* 2018; Feldhusen, 2020; Djedjibegovic *et. ai.,* 2020). One major contaminant group commonly ingested with fish is the polyaromatic hydrocarbon. Polyaromatic hydrocarbon (PAH) compounds belong to a varied class of organic compounds with usually three or four benzene rings fused together containing carbon and hydrogen only and having properties varying based on ring structure and/or configuration. Oranusi *et al*, (2018) defined PAHs as a large group of chemically inert, hydrophobic compounds consisting of three or more condensed aromatic rings soluble in organic solvents which are ubiquitous in the environment as a result of incomplete combustion of organic materials during industrial processing and various human activities., PAHs are formed mainly as a result of pyrolytic processes, especially the incomplete combustion of organic materials during industrial and other human activities, such as processing of coal and crude oil, heating, burning of refuse, cooking and tobacco smoking, as well as in natural processes such as carbonization (Sojinu *et. al.,* 2019).. The presence of PAHs in food is usually a consequence

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of the nature of these compounds in the environment, their formation during cooking processes or as a result of the manufacturing processes

More than 100 PAHs have been characterized, 16 of which were classified by United States Environmental Protection Agency (USEPA) and the European Food Safety Authourity (EFSA) as priority pollutants because of their toxicity (USEPA, 1993; EFSA, 2008). The chemical structure of these priority PAHs are as shown in Figure 1. Due to their mutagenic and carcinogenic nature, both European Union and US Environmental Protection Agency (US EPA) have pointed out Polyaromatic Hydrocarbons (PAHs) as priority pollutants (Ramalhosa, 2019). Several studies have implicated PAHs in the incidences of reduced lung function, worsening asthma, and increasing cases of obstructive lung diseases, and dietary sources have been identified as one of the predominant avenue of human exposure to them, though not a primary source.

At the fore-front of all avenues of human exposure to PAHs is smoking – either smoking of cigarette or smoking of food. Unfortunately, smoking/grilling of meat and fish in open air till date is embraced as a popular method of food processing and preservation, especially in African countries. The amount of PAHs generated during smoking however, depends on several parameters such as temperature, duration of the treatment, distance from the source of heating, oxygen accessibility, fat content, and type of combustible used (Alonge, 1998; Visciano *et. al.,* 2006).

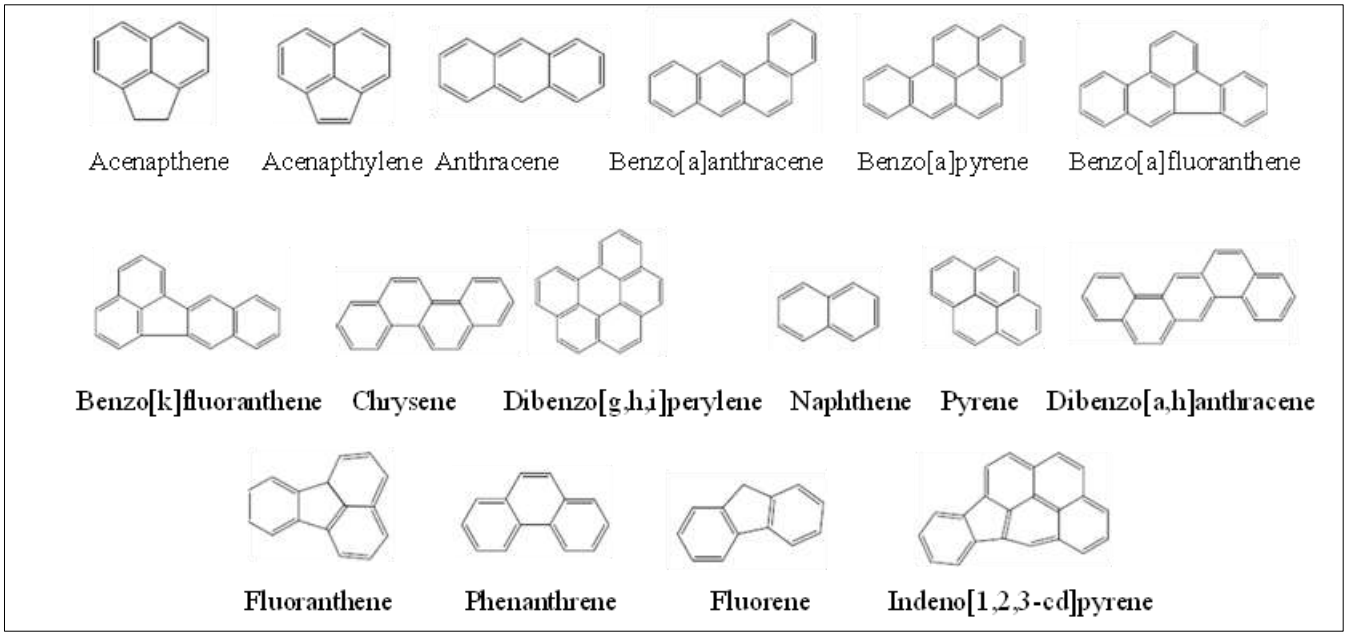



**Figure 1** Chemical Structures of Priority PAHs (Pule *et. al., 2007).*

**Table 1** USEPA Priority PAHs and their Carcinogenicity rating

| PAH | Molecular Formula | Carcinogenic rating |
|---|---|---|
| Naphtalene | $C_{10}H_8$ | * |
| Phenanthrene | $C_{14}H_{10}$ | * |
| Anthracene | $C_{14}H_{10}$ | * |
| Fluoranthene | $C_{16}H_{10}$ | * |
| Pyrene | $C_{16}H_{10}$ | * |
| Chrysene | $C_{18}H_{12}$ | *** |
| Benzo(a)anthracene | $C_{18}H_{12}$ | *** |
| Benzo(b)fluoranthene | $C_{18}H_{12}$ | *** |
| Benzo(k)fluoranthene | $C_{20}H_{12}$ | ** |

| | | |
|---|---|---|
| Fluorene | $C_{13}H_{10}$ | * |
| Benzo(a)pyrene | $C_{20}H_{12}$ | *** |
| Acenaphtene | $C_{12}H_{10}$ | * |
| Benzo(g, h, i)perylene | $C_{20}H_{12}$ | ** |
| Dibenzo(a, h)anthracenes | $C_{22}H_{14}$ | ** |
| Indeno(c, d)pyrylene | $C_{22}H_{12}$ | ** |
| Acenaphtylene | $C_{12}H_8$ | * |

* Non-Carcinogenic PAHs. ** Carcinogenic PAHs. *** Carcinogenic PAH and PAH usually sed to derive the carcinogenic Index (Tongo et al., 2017).

The three fish species analyzed in this study: Herring (*Clupea harengus harengus)*, Blue whiting (*Micromesistius poutassou)* and Mackerel (*Scomber scombrus)* are popularly consumed in Nigeria and especially by residents of the study area. Blue whiting alongside the other two can be classified as pelagic species (Gatt, 2023). EU export data indicated that Nigeria was the highest importer of blue whiting, mackerel and herring in 2022 with the latter's import being almost 70,000 tonnes (EUMOFA, 2023). Herring is locally called '*shawa',* mackerel *'alaran' and* blue whiting *'panla egun' among t*he majorly Yoruba people of the study area. The sampling points in the study area are shown in table2.

**Table 2** Sampling Points within the Study Area and Their Coordinates

| **S/N** | **Town** | |
|---|---|---|
| 1 | Isara | 6° 59' 15"N, 3° 40' 40"E |
| 2. | Ipara | 7° 00' 12.2"N, 3° 40' 04.8"E |
| 3. | Iperu | 6° 55' 00"N, 3° 39' 54"E |
| 4 | Ilishan | $6^0$ 53' 4''N, $3^0$ 42' 45"E |
| 5 | Sagamu | 6° 50' 04"N, 3° 37' 52"E |

## 2. Materials and methods

### 2.1. Sampling

A total of 15 samples were employed in this study. Three different species of fishes, namely; herring, blue whiting and Mackerel commonly consumed in Ogun states were purchased from Isara market, Ipara market, Akesan market, Iperu, Ilishan market and Awolowo market, Sagamu Ogun state respectively. Herring, Blue whiting and Mackerel were labeled A, B and C respectively. Each of the location source of the sample was labeled as follows; 1 for Isara, 2 for Ipara, 3 for Iperu, 4 represent Ilishan and 5 for Sagamu. Each sample was wrapped in aluminum foil and transported to the laboratory in cold coolers.

### 2.2. Sample Extraction

The fish samples were crushed and pounded into fine form using mortar and pestle. 10 g of the sample was measured into the 250 ml conical flask, 25 ml of HPLC grade Dichloromethane (DCM) was added and the aliquot subject to ultrasonication for 20mins. The clear portion was decanted into a clean 100 ml beaker under the fume cupboard. Another 25ml DCM was added to the residue in the conical flask and sonicated for another 20mins. The clear portion was decanted into the initial 100ml beaker. The sample extract was allowed to concentrate to about 5ml under liquid concentrator.

### 2.3. Sample Clean-up technique

Analytical column was packed with cotton wool containing anhydrous sodium sulphate and silica gel that has been dried for 2 hours at 105 $^0$C. A mixture of the silica gel and anhydrous sodium sulphate (1g each) was placed on the cotton wool inside the column. The column was conditioned by using a mixture of 2.5ml of n-hexane and DCM. The concentrated sample above was allowed to pass through the column and later eluted with 2.5ml of mixture of acetone and DCM. The

eluted sample was evaporated to dryness under nitrogen concentrator. The evaporated extract was reconstituted with 2ml DCM and later injected into the GCMS.

### 2.4. Chromatographic Parameters

Agilent 8860A gas chromatograph coupled to 5977C inert mass spectrometer (with triple axis detector) with electron-impact source (Agilent Technologies) was used in this study. The stationary phase of separation of the compounds was HP-5 capillary column coated with 5% Phenyl Methyl Siloxane (30m length x 0.25mm diameter x 0.25µm film thickness) The carrier gas was Helium used at constant flow of 1.2 mL/min at an initial nominal pressure of 026 psi and average velocity of 40.00 cm/sec.

1µL of the samples were injected in splitless mode at an injection temperature of 250 °C. Purge flow to spilt vent was 30.0 mL/min at 0.35 min with a total flow of 31.24 mL/min; gas saver mode was switched off. Oven was initially programmed at 50 °C (2 min) then ramped at 10 °C/min to 300 °C (5 min). Run time was 32 min with a 3 min solvent delay.

### 2.5. Health Risk Assessment

This study conducted a risk assessment of the collected fish samples by estimating metrics like daily dietary intake (DDI), carcinogenic potencies of individual PAHs ($B(A)P_{teq}$) and the excess cancer risk (ECR). Methods similar to those of Tongo *et al.* (2017) were adopted in this study's PAHs risk assessment. The result of all three parameters assessed are summarized in Tables 4, 5 and 6, for Herring, Mackerel and Blue whiting respectively.

### 2.6. Dietary Daily Intake (DDI) of PAHs from the Three Fish Samples

Dietary Daily Intake (DDI) of PAHs in the smoked fish samples collected from the five study locations was estimated using Eq. (1). The daily ingestion of PAHs through locally processed smoked fish was obtained by multiplying the concentration of individual PAH with the fish ingestion rate (IFR).

$$DDI = C_i \times IFR \quad (1)$$

The adult weight of smoked fish consumers was taken as 70 kg as also used by Tongo *et al.* (2017). Also, an average fish ingestion rate (IFR) of 0.0548 kg/capita/day, as estimated by FAO (2014), was used in the calculation of the DDI.

### 2.7. Carcinogenic Potencies of PAHs

The carcinogenic potency of PAHs is a measure of the carcinogenic risk that PAHs compounds may pose to persons ingesting them. It is usually expressed as the equivalence of the toxicity of Benzo [a] pyrene which has been accepted as a marker for the occurrence and effect of carcinogenic PAHs in smoked foods as specified in the EU Commission Regulation (EU Commission, 2014). Toxicity equivalence factors ($TEF_i$) estimated by Nisbet and Lagoy (1992) were used in the calculation of carcinogenic potencies in this study (Equation. 2).

$$\text{Carcinogenic potencies of individual PAHs} = (B(A)Pteq) = Ci \times TEFi \quad (2)$$

### 2.8. Excess Cancer Risk (ECR) of PAHs

This study also assessed the excess cancer risk (ECR) potentially induced by dietary exposure of smoked fish consumers in the study area to PAHs. ECR was calculated using equation (3) (Tongo *et al,* 2017)

.

$$\text{Excess Cancer Risk (ECR)} = \frac{Q\ X\ B\ (A)\ Pteq\ X\ IFR\ X\ ED}{(BW\ X\ ATn)} \quad (3)$$

## 3. Results and discussion

The concentrations of the respective PAHs in each fish specie were as contained in Table 3. The concentrations of the 22 PAHs summed up to ∑PAH22 (Figure 2 and Table 3). ∑PAH22 ranged from 23.769 µg/kg found in herring sample from Ipara market to 1.48 µg/kg found in the sample from Ilishan market. Mackerel (*Scomber scombrus*) had the highest ∑PAH22 in the sample from Ipara market with 1.80 µg/kg and lowest concentration from Ilishan market with 1.556 µg/kg.. Blue Whiting (*Micromesistins poutasou*) had the highest concentration of ∑PAH22 in the sample from Ilishan

market (11.68 µg/kg) and the lowest from Iperu market (1.94 µg/kg). Herring had a mean PAH22 concentration of 7.73 µg/kg and a mean of 1.66 µg/kg was observed for PAH22 in Mackerel. Mean PAH22 concentration of 4.16 was found in blue whiting across the five markets. Unlike the other two fish samples from Ilishan, herring had a high concentration of ∑PAH22 suggesting that different smoking methods or materials were employed for the Blue whiting sample from the location. Some of these values are higher than the 10 µg/Kg$^{-1}$ maximum limits set by the European Union for total PAHs (Ogundiran *et. al.,* 2024). The results from the GC-MS analysis showed a significant difference ($p<0.05$) in the PAHs concentration detected in the smoked fish samples collected from the five study locations.

The result obtained in this study were similar to those obtained by Adesina *et. al.* (2021).who obtained PAHs concentration levels ranging between 0.0001 and 0.996 µg/kg in *Clupea herengus* (herring) and hake fish samples analyzed. However, our results were below the 3.585 mg/kg of PAHs found in *Scomber scombrus* obtained in Benin, Nigeria (Tongo *et al,* 2017).

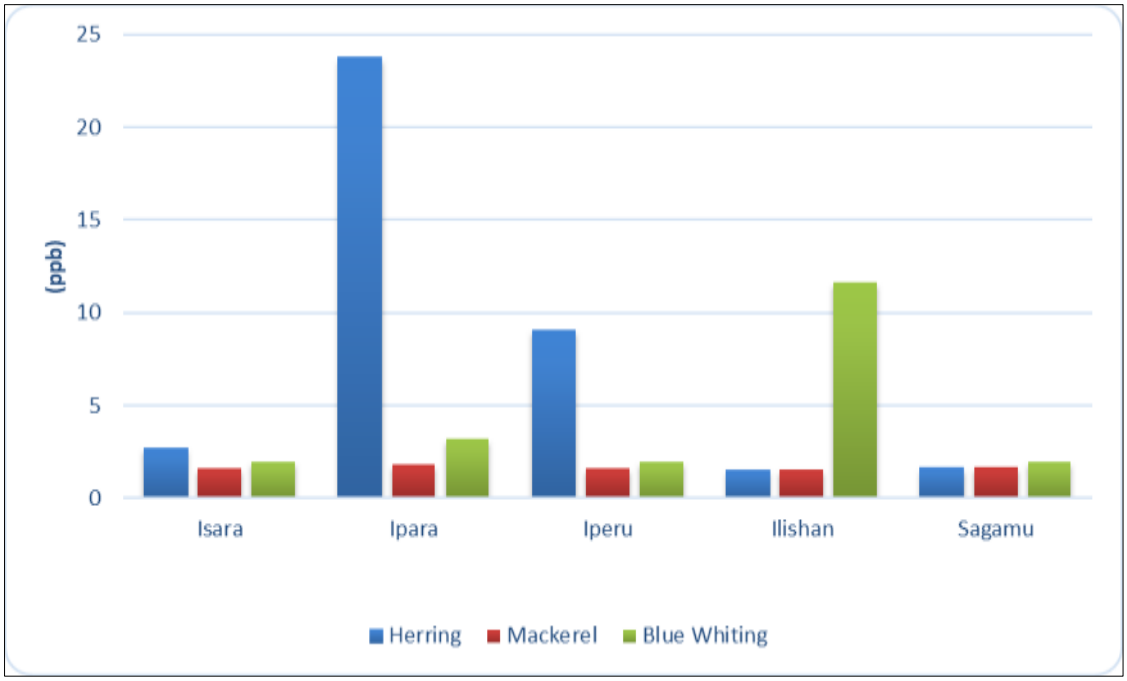
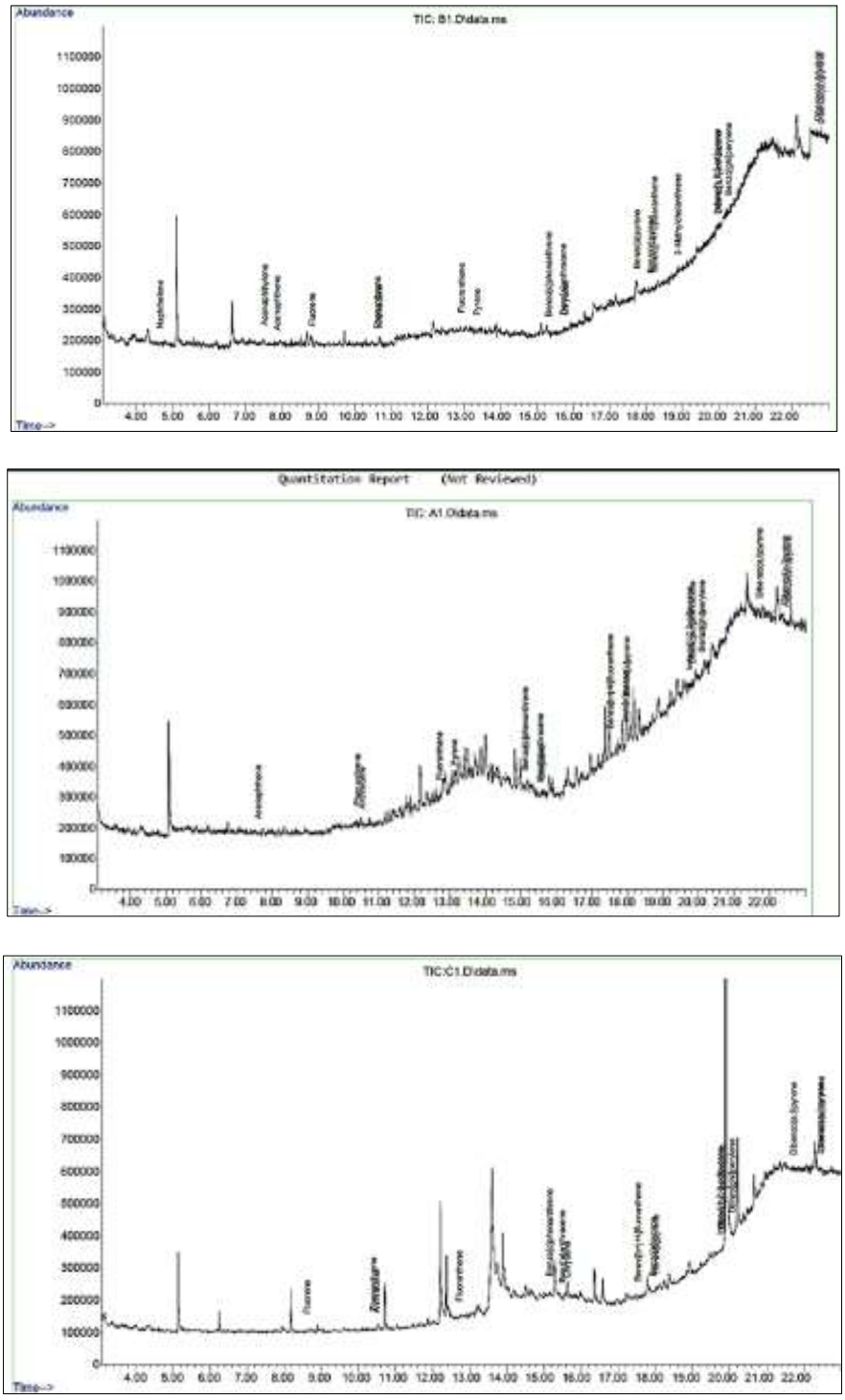



**Figure 2** ∑ PAH22 in Herring (*C. harengus),* Mackerel *(S. scombrus)* and Blue Whiting (*M. poutassou)* samples.

The concentrations of the 16 PAHs highlighted as priority pollutants (USEPA, 1993) were also assessed in this study. Their sum total ∑PAH16 in each sample from each market was as contained in Table 3 and Figure 4. The four highest ∑PAH16 concentrations were found in herring from Ipara (8.82 µg/kg), blue whiting sample from Ilishan (6.15 µg/kg), herring from Iperu (5.67 µg/kg) and Blue whiting from Ipara (2.24 µg/kg). the member of the ∑PAH16 with the highest concentration in all the samples from all market was Indeno (1,2,3-cd) pyrene (1.61 µg/kg). Indeno (1,2,3-cd) pyrene and other seven PAHs Benzo [a] pyrene, benzo [a] anthracene, chrysene, benzo [k] fluoranthene, benzo [b] fluoranthene, dibenzo [a,h] anthracene and benz [g,h,i] pyrene referred to as PAH8 have been reported in an *in-vivo* experiment on animals to have a mutagenic/genotoxic effect in somatic cells (EFSA, 2008).




A = GC-MS Chromatogram of PAHs in Mackerel Sample from Isara. B = GC-MS Chromatogram of PAHs in Herring Sample from Isara., C = GC-MS Chromatogram of PAHs in Blue Whiting Sample from Isara.

**Figure 3** Chromatograms of Fish Sample Analysis**.**

**Table 3** Concentration of PAHs Detected in the Fish samples

| **PAH** | **Herring (ppb)** | | | | | **Mackerel (ppb)** | | | | | **Blue Whiting (ppb)** | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | Isara | Ipara | Iperu | Ilishan | Sagamu | Isara | Ipara | Iperu | Ilishan | Sagamu | Isara | Ipara | Iperu | Ilishan | Sagamu |
| NAPT | - | $0.279^{a}$ | $0.221^{c}$ | $0.088^{e}$ | $0.079^{f}$ | $0.092^{e}$ | $0.10^{d}$ | $0.081^{f}$ | $0.091^{e}$ | $0.07^{g}$ | - | - | - | $0.231^{b}$ | - |
| ACTY | - | $0.206^{d}$ | $0.299^{b}$ | $0.062^{ef}$ | $0.052^{gh}$ | $0.069^{e}$ | $0.061^{efg}$ | $0.059^{efgh}$ | $0.058^{fgh}$ | $0.051^{h}$ | - | $1.450^{a}$ | - | $0.290^{c}$ | - |
| ACTE | $0.012^{f}$ | $0.319^{b}$ | $0.249^{c}$ | $0,049^{e}$ | $0.051^{e}$ | $0.052^{e}$ | $0.060^{d}$ | $0.061^{d}$ | $0.050^{e}$ | $0.060^{d}$ | - | - | - | $0.405^{a}$ | - |
| FLUO | - | $0.612^{a}$ | $0.231^{c}$ | $0.091^{f}$ | $0.089^{f}$ | $0.089^{f}$ | $0.091^{f}$ | $0.091^{f}$ | $0.103^{e}$ | $0.110^{d}$ | 0.00 | - | - | $0.339^{b}$ | - |
| PHEN | $0.012^{f}$ | $0.159^{a}$ | $0.089^{c}$ | $0.051^{e}$ | $0.052^{e}$ | $0.051^{e}$ | $0.050^{e}$ | $0.059^{d}$ | $0.049^{e}$ | $0.061^{d}$ | 0.00 | $0.010^{f}$ | 0.00 | $0.140^{b}$ | $0.011^{f}$ |
| ANTR | $0.021^{g}$ | $0.271^{a}$ | $0.231^{c}$ | $0.039^{e}$ | $0.031^{f}$ | $0.049^{d}$ | $0.031^{f}$ | $0.030^{f}$ | $0.040^{e}$ | $0.029^{f}$ | $0.020^{g}$ | $0.020^{g}$ | $0.021^{g}$ | $0.249^{b}$ | $0.021^{g}$ |
| FLRT | $0.031^{f}$ | $0.331^{a}$ | $0.239^{b}$ | $0.059^{e}$ | $0.059^{e}$ | $0.069^{d}$ | $0.061^{e}$ | $0.061^{e}$ | $0.060^{e}$ | $0.060^{e}$ | $0.022^{g}$ | $0.020^{g}$ | $0.020^{g}$ | $0.110^{c}$ | $0.021^{g}$ |
| PYRE | 0.00 | $0.259^{b}$ | $0.219^{c}$ | $0.039^{d}$ | $0.042^{d}$ | $0.041^{d}$ | $0.040^{d}$ | $0.030^{e}$ | ND | $0.041^{d}$ | - | - | - | $0.511^{a}$ | - |
| BcPT | $0.061^{d}$ | $0.488^{a}$ | $0.307^{b}$ | $0.041^{efg}$ | $0.039^{g}$ | $0.041^{efg}$ | $0.041^{efg}$ | $0.041^{efg}$ | $0.040^{fg}$ | $0.051^{def}$ | $0.051^{fg}$ | $0.050^{def}$ | $0.051^{de}$ | $0.249^{c}$ | $0.050^{ef}$ |
| BaATR | $0.052^{g}$ | $0.410^{b}$ | $0.249^{c}$ | $0.089^{f}$ | $0.111^{d}$ | $0.091^{f}$ | $0.089^{f}$ | $0.089^{f}$ | $0.090^{f}$ | $0.101^{e}$ | $0.050^{g}$ | $0.040^{h}$ | $0.049^{g}$ | $0.431^{a}$ | $0.051^{g}$ |
| CHRY | $0.051^{f}$ | $0.629^{a}$ | $0.268^{c}$ | $0.062^{e}$ | $0.069^{d}$ | $0.061^{e}$ | $0.061^{e}$ | $0.062^{e}$ | $0.060^{e}$ | $0.062^{e}$ | $0.050^{f}$ | $0.040^{g}$ | $0.040^{g}$ | $0.521^{b}$ | $0.040^{g}$ |
| BbFN | $0.990^{d}$ | $0.571^{a}$ | $0.351^{b}$ | $0.111^{d}$ | $0.109^{d}$ | $0.110^{d}$ | $0.109^{d}$ | $0.110^{d}$ | $0.110^{d}$ | $0.111^{d}$ | $0.116^{d}$ | $0.102^{d}$ | $0.101^{d}$ | $0.230^{c}$ | $0.105^{d}$ |
| BjFN | $0.106^{d}$ | $0.469^{a}$ | $0.231^{c}$ | $0.109^{d}$ | $0.112^{d}$ | $0.110^{d}$ | $0.111^{d}$ | $0.111^{d}$ | $0.111^{d}$ | $0.110^{d}$ | $0.122^{d}$ | $0.101^{d}$ | $0.100^{d}$ | $0.271^{b}$ | $0.106^{d}$ |
| BkFN | - | $0.471^{c}$ | $0.230^{d}$ | $0.111^{d}$ | $0.109^{d}$ | $0.111^{d}$ | $0.110^{d}$ | $0.110^{d}$ | $0.110^{d}$ | $0.110^{d}$ | $0.101^{d}$ | $0.101^{d}$ | $0.118^{d}$ | $0.270^{b}$ | $0.102^{d}$ |
| DiBaANT | - | - | - | - | - | - | - | - | - | - | - | - | - | - | - |
| BePYR | $0.081^{e}$ | $0.580^{a}$ | $0.341^{c}$ | $0.059^{f}$ | $0.094^{de}$ | $0.101^{de}$ | $0.100^{de}$ | $0.102^{de}$ | $0.102^{de}$ | $0.106^{d}$ | $0.111^{d}$ | $0.080^{ef}$ | $0.080^{e}$ | $0.542^{b}$ | $0.081^{e}$ |
| BaPYR | $0.110^{d}$ | $0.811^{a}$ | $0.328^{b}$ | $0.095^{ef}$ | $0.061^{h}$ | $0.060^{h}$ | $0.062^{h}$ | $0.061^{h}$ | $0.061^{h}$ | $0.061^{h}$ | $0.081^{g}$ | $0.090^{fg}$ | $0.105^{de}$ | $0.280^{c}$ | $0.102^{def}$ |
| 3MCOL | $0.180^{g}$ | $10.470^{a}$ | $1.591^{c}$ | $0.069^{j}$ | $0.181^{g}$ | $0.159^{h}$ | $0.340^{d}$ | $0.139^{i}$ | $0.250^{e}$ | $0.221^{f}$ | - | - | - | $3.022^{b}$ | - |
| Ind[1,2,3]PYR | - | $1.612^{a}$ | $1.069^{b}$ | $0.071^{e}$ | $0.069^{e}$ | $0.070^{e}$ | $0.071^{e}$ | $0.069^{e}$ | $0.070^{e}$ | $0.070^{d}$ | $0.106^{d}$ | $0.105^{d}$ | $0.101^{e}$ | $0.870^{c}$ | $0.110^{d}$ |
| DiB[a.h}ANT | $0.091^{h}$ | $1.160^{a}$ | $1.071^{b}$ | $0.120^{g}$ | $0.121^{g}$ | $0.120^{g}$ | $0.120^{g}$ | $0.130^{f}$ | ND | $0.130^{f}$ | $0.181^{d}$ | $0.171^{e}$ | $0.171^{e}$ | $0.871^{c}$ | $0.180^{d}$ |
| B[g,h,i]PE | $0.241^{bc}$ | $0.719^{a}$ | $0.328^{c}$ | $0.059^{c}$ | $0.060^{c}$ | $0.060^{c}$ | $0.061^{c}$ | $0.061^{c}$ | $0.070^{c}$ | $0.061^{c}$ | $0.090^{c}$ | $0.090^{c}$ | $0.090^{c}$ | $0.402^{b}$ | $0.091^{c}$ |
| DiB[a,l]PY | $0.350^{c}$ | $0.613^{a}$ | $0.242^{d}$ | $0.031^{h}$ | $0.022^{i}$ | $0.022^{i}$ | $0.020^{i}$ | $0.021^{i}$ | $0.021^{i}$ | $0.021^{i}$ | $0.221^{f}$ | $0.090^{g}$ | $0.220^{f}$ | $0.450^{b}$ | $0.230^{e}$ |
| DiB[a,i]PY | $0.321^{e}$ | $2.330^{a}$ | $0.679^{c}$ | $0.019^{g}$ | $0.031^{f}$ | $0.011^{h}$ | $0.010^{h}$ | $0.020^{g}$ | $0.010^{h}$ | $0.011^{h}$ | $0.350^{d}$ | $0.351^{d}$ | $0.351^{d}$ | $0.994^{b}$ | $0.351^{d}$ |

| | | | | | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| DiB[a,h]PY | - | - | - | - | - | - | - | - | - | - | 0.320[a] | 0.320[a] | 0.321[a] | - | 0.320[a] |
| ∑PAH22 | 2.71 | 23.77 | 9.06 | 1.48 | 1.64 | 1.64 | 1.80 | 1.60 | 1.56 | 1.71 | 1.99 | 3.23 | 1.94 | 11.68 | 1.97 |
| ∑ PAH16 | 1.61 | 8.82 | 5.67 | 1.15 | 1.16 | 1.2 | 1.18 | 1.16 | 1.02 | 1.19 | 0.82 | 2.24 | 0.82 | 6.15 | 0.83 |

NAPT = naphthalene, ACTY = acenaphthylene, ACTE = acenaphthene, FLUO = fluorene, PHEN = phenanthrene, PYRE = pyrene, BcPT = benzo [c] phenanthrene, BaATR = benzo [a] anthracene, CHRY = chrysene, BaPYR = benzo [a] pyrene, BePYR = benzo [e] pyrene Ind[1,2,3,]PYR = indeno [1, 2, 3-cd] pyrene, Anthracene, = ANTR , FLRT = Fluoranthene, BbFN = benzo [b] fluoranthene, BjFN = benzo [j] fluoranthene, BkFN = benzo [k] fluoranthene, DiBaANT = Dimethylbenz[a]anthrancene, 3MCOL = 3-MethylCholanthrene, DiB[a,h]ANT = Dibenz[a,h]anthrancene, B[g,h,i]PE = Benzo[g,h,i]perylene, DiB[a,l]PY = Dibenz[a,l]pyrene, DiB[a,i]PY = Dibenz[a,i]pyrene, DiB[a,h]PY = Dibenz[a,h]pyrene

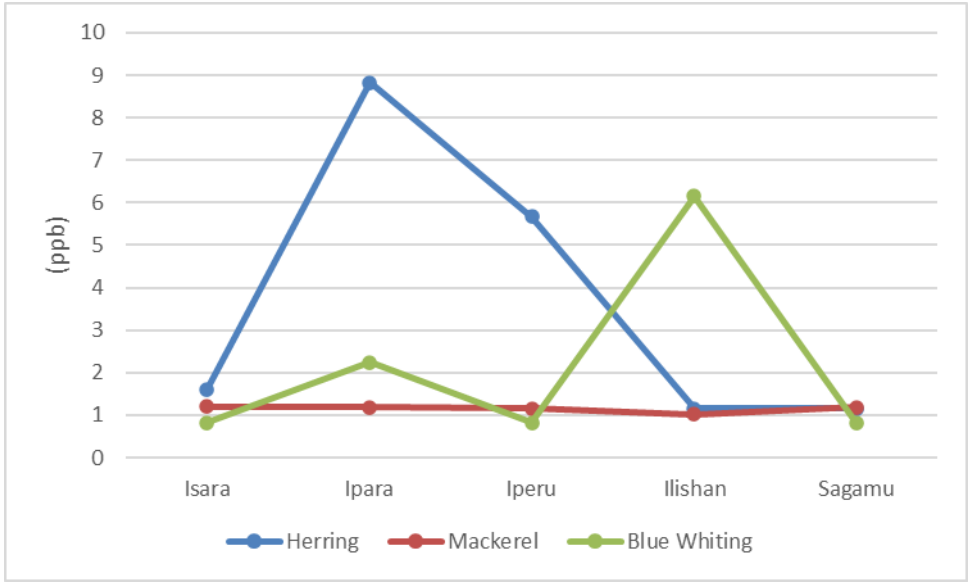



**Figure 4** Total PAH16 Concentrations (µg/kg) in Fish Samples from the Study Markets

### 3.1. Health Risk of PAHs

#### *3.1.1. Health Risk of PAHs from Herring (Clupea harengus)*

Smoked fish is cheaper and readily available in Nigeria and consequently gets consumed more than those processed via other means. This availability makes smoked fish have a relatively higher daily dietary consumption among locals in the study area. Thus, an assessment of the toxicity risk of consuming the smoked fish species was determined by estimating the daily dietary intake of each sample. As summarized in Table 4, an adult (70 kg-bw) DDI for smoked Herring was found to be between 0 and 0.1277 µg/day. The highest DDI, B(A)PTEQ and ECR ∑PAH22 for herring was for the sample obtained from Ipara having 1.302 µg/day, 6.934 and 0.106 respectively. The DDI indicate that consumers of the smoked herring form the location are more exposed to the risk of toxicity from PAHs via the fish. The B(A)PTEQs obtained in this study were much higher than the Maximum Acceptable Risk level of $10^{-5}$. This reveals the high potency of the PAHs in the sample to pose risks to the consumer of the fish samples studied in this work The ECR expresses the potential risk caused by dietary exposure to PAHs for an adult weighing 70kg. ECR is usually estimated from lifetime exposure to PAH through a particular dietary route and an acceptable guideline of $10^{-6}$ has been set by set by USEPA (2001). A lifetime cancer risk of one in a million (ECR = $10^{-6}$) is deemed acceptable while an lifetime cancer risk of one in ten thousand or greater (ECR = $10^{-4}$), is considered serious (Tongo *et al.,* 2017). The values of ECR obtained from this study are higher than the guideline and indeed call for serious monitoring.

#### *3.1.2. Health Risk of PAHs from Mackerel (Scomber scombrus)*

Table 5 contains the DDI, B(A)PTEQ and ECR from the consumption of Mackerel from the study area for an adult (70 kg-bw), DDI for smoked Mackerel was found to be between 0 and 0.007124 µg/day. The same values of DDI, B(A)PTEQ and ECR for Mackerel were found in the samples obtained from Iperu and Sagamu at 0.007124 µg/day, 0.65 and 0.009937 respectively. The DDI indicate that consumers of the smoked Mackerel from Iperu and Sagamu have same extent of exposure to the risk of toxicity from PAHs via the Mackerel consumption. The B(A)PTEQs obtained in this study were much higher than the Maximum Acceptable Risk level of $10^{-5}$. This reveals the high potency of the PAHs in the sample to pose risks to the consumer of the fish samples studied in this work. 9 of the B(A)PTEQ values obtained in this study were far higher than Maximum Risk Levels with DiBenzo[a, h] Anthranccene having B(A)PTEQ of 0.6. ECR is usually estimated from lifetime exposure to PAH through a particular dietary route and an acceptable guideline of $10^{-6}$ has been set by USEPA (2001). The values of ECR obtained for mackerel in this study are moderately higher than the guideline. The ∑PAH16 values for mackerel shows a high risk of potencies of PAHs and high lifetime toxicity from the consumption of mackerel from the study area.

#### *3.1.3. Health Risk of PAHs from Blue Whiting (Micromesistius poutasou)*

The DDI, B(A)PTEQ and ECR of the 16 priority PAHs in adult human (70 kg-bw) consumers of Blue Whiting in the study area are contained in Table 6.. DDI for smoked Blue Whiting was between 0 and 0.07946 µg/day. The highest DDI, B(A)PTEQ and ECR ∑PAH16 for Blue Whiting was for the sample obtained from Ilishan having 0.638968 µg/day, 4.82372 and 0.073741 respectively. The DDI indicate that consumers of the smoked Blue Whiting from this location are more exposed to the risk of toxicity from PAHs via the fish. The B(A)PTEQs obtained in this study were much higher than the Maximum Acceptable Risk level of $10^{-5}$. This reveals the high potency of the PAHs in the sample to pose risks to the consumer of the fish samples studied in this work The values of ECR obtained for Blue whiting in this study are higher than the guideline and also require intense monitoring.

Overall, the cumulative concentrations of ∑PAH22 and ∑PAH16in the three species of fish are of the order Herring > Blue whiting > Mackerel. However, for ∑PAH16, the trend DDI was Blue whiting > Herring > Mackerel, this order probably was due to the relative difference in cost of the three species of fish which is in the order Mackerel > Herring > Blue whiting. Thus, the affordability would play an important role in dietary consumption of the individual specie. This also would impact the PAH intake from the respective fish specie. The same pattern was observed for the other risk assessment parameters DDI, B(A)QTEQ and ECR. This pattern suggests that the exposure to PAH toxicity risk in the study area is relative to the consumption rate of the fish which may be consequent to the the cost of the fish.

**Table 4** DDI, B(A)P and ECR for Herring (*Clupea harengus*)

| **PAH** | **Isara** | | | **Ipara** | | | **Iperu** | | | **Ilishan** | | | **Sagamu** | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | DDI | B(A)P | ECR | DDI | B(A)P | ECR | DDI | B(A)P | ECR | DDI | B(A)P | ECR | DDI | B(A)P | ECR |
| NAPT | | 0 | 0 | 0.015344 | 0.00030 | 4.28043E-06 | 0.012056 | 0.00022 | 3.36319E-06 | 0.004932 | 0.00009 | 1.37585E-06 | 0.004932 | 0.00009 | 1.37585E-06 |
| ACTY | | 0 | 0 | 0.01096 | 0.0002 | 3.05745E-06 | 0.01644 | 0.0003 | 4.58617E-06 | 0.003288 | 0.00006 | 9.17235E-07 | 0.00274 | 0.00005 | 7.64362E-07 |
| ACTE | 0.000548 | 0.00001 | 1.52872E-07 | 0.017536 | 0.0003 | 4.89192E-06 | 0.0137 | 0.00025 | 3.82181E-06 | 0.0548 | 0.001 | 1.52872E-05 | 0.00274 | 0.00005 | 7.64362E-07 |
| FLUO | | 0 | 0 | 0.033428 | 0.0006 | 9.32522E-06 | 0.012604 | 0.00023 | 3.51607E-06 | 0.004932 | 0.00009 | 1.37585E-06 | 0.004932 | 0.00009 | 1.37585E-06 |
| PHEN | 0.000548 | 0.00001 | 1.52872E-07 | 0.008768 | 0.0002 | 2.44596E-06 | 0.004932 | 0.00009 | 1.37585E-06 | 0.00274 | 0.00005 | 7.64362E-07 | 0.00274 | 0.00005 | 7.64362E-07 |
| ANTR | 0.001096 | 0.0002 | 3.05745E-06 | 0.014796 | 0.0027 | 4.12756E-05 | 0.012604 | 0.0023 | 3.51607E-05 | 0.002192 | 0.0004 | 6.1149E-06 | 0.001644 | 0.0003 | 4.58617E-06 |
| FLRT | 0.001644 | 0.00003 | 4.58617E-07 | 0.018084 | 0.0003 | 5.04479E-06 | 0.013152 | 0.00024 | 3.66894E-06 | 0.003288 | 0.00006 | 9.17235E-07 | 0.003288 | 0.00006 | 9.17235E-07 |
| PYRE | 0 | 0 | 0 | 0.014248 | 0.0003 | 3.97468E-06 | 0.012056 | 0.00022 | 3.36319E-06 | 0.002192 | 0.00004 | 6.1149E-07 | 0.002192 | 0.00004 | 6.1149E-07 |
| BaATR | 0.00274 | 0.005 | 7.64362E-05 | 0.022468 | 0.0041 | 0.000626777 | 0.0137 | 0.025 | 0.000382181 | 0.004932 | 0.009 | 0.000137585 | 0.006028 | 0.011 | 0.00016816 |
| CHRY | 0.00274 | 0.0005 | 7.64362E-06 | 0.034524 | 0.0063 | 9.63096E-05 | 0.014796 | 0.0027 | 4.12756E-05 | 0.003288 | 0.0006 | 9.17235E-06 | 0.003836 | 0.0007 | 1.07011E-05 |
| BbFN | 0.00548 | 0.01 | 0.000152872 | 0.031236 | 0.0057 | 0.000871373 | 0.01918 | 0.035 | 0.000535054 | 0.006028 | 0.011 | 0.00016816 | 0.006028 | 0.011 | 0.00016816 |
| BkFN | 0 | 0 | 0 | 0.025756 | 0.0047 | 0.0007185 | 0.012604 | 0.023 | 0.000351607 | 0.006028 | 0.011 | 0.00016816 | 0.006028 | 0.011 | 0.00016816 |
| BaPYR | 0.006028 | 0.11 | 0.001681597 | 0.044388 | 0.8100 | 0.012382667 | 0.018084 | 0.33 | 0.00504479 | 0.00548 | 0.1 | 0.001528724 | 0.003288 | 0.06 | 0.000917235 |

| | | | | | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Ind[1,2,3]PYR | 0 | 0 | 0 | 0.088228 | 0.1610 | 0.002461246 | 0.058636 | 0.107 | 0.001635735 | 0.003836 | 0.007 | 0.000107011 | 0.003836 | 0.007 | 0.000107011 |
| DiB[a.h}ANT | 0.004932 | 0.45 | 0.006879259 | 0.063568 | 5.8000 | 0.088666009 | 0.058636 | 5.35 | 0.081786749 | 0.006576 | 0.6 | 0.009172346 | 0.006576 | 0.6 | 0.009172346 |
| B[g,h,i]PE | 0.013152 | 0.0024 | 3.66894E-05 | 0.039456 | 0.0072 | 0.000110068 | 0.018084 | 0.0033 | 5.04479E-05 | 0.003288 | 0.0006 | 9.17235E-06 | 0.003288 | 0.0006 | 9.17235E-06 |
| ∑PAH16 | 0.09864 | 0.57815 | 0.008838 | 1.302048 | 6.9344 | 0.106007 | 0.496488 | 5.87985 | 0.089887 | 0.135904 | 0.74108 | 0.011328 | 0.09042 | 0.70212 | 0.010732 |

**Table 5** DDI, B(A)P and ECR for *Mackerel (Scomber scombrus*)

| **PAH** | **Isara** | | | **Ipara** | | | **Iperu** | | | **Ilishan** | | | **Sagamu** | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | DDI | B(A)P | ECR | DDI | B(A)P | ECR | DDI | B(A)P | ECR | DDI | B(A)P | ECR | DDI | B(A)P | ECR |
| NAPT | 0.004932 | 0.00009 | 1.37585E-06 | 0.00548 | 0.0001 | 1.52872E-06 | 0.004384 | 0.00008 | 1.22298E-06 | 0.004932 | 0.00009 | 1.37585E-06 | 0.003836 | 0.00007 | 1.07011E-06 |
| ACTY | 0.003836 | 0.00007 | 1.07011E-06 | 0.003288 | 0.00006 | 9.17235E-07 | 0.003288 | 0.00006 | 9.17235E-07 | 0.003288 | 0.00006 | 9.17235E-07 | 0.00274 | 0.00005 | 7.64362E-07 |
| ACTE | 0.00274 | 0.00005 | 7.64362E-07 | 0.003288 | 0.00006 | 9.17235E-07 | 0.003288 | 0.00006 | 9.17235E-07 | 0.00274 | 0.00005 | 7.64362E-07 | 0.003288 | 0.00006 | 9.17235E-07 |
| FLUO | 0.004932 | 0.00009 | 1.37585E-06 | 0.004932 | 0.00009 | 1.37585E-06 | 0.004932 | 0.00009 | 1.37585E-06 | 0.00548 | 0.0001 | 1.52872E-06 | 0.006028 | 0.00011 | 1.6816E-06 |
| PHEN | 0.00274 | 0.00005 | 7.64362E-07 | 0.00274 | 0.00005 | 7.64362E-07 | 0.003288 | 0.00006 | 9.17235E-07 | 0.00274 | 0.00005 | 7.64362E-07 | 0.003288 | 0.00006 | 9.17235E-07 |
| ANTR | 0.00274 | 0.0005 | 7.64362E-06 | 0.001644 | 0.0003 | 4.58617E-06 | 0.001644 | 0.0003 | 4.58617E-06 | 0.002192 | 0.0004 | 6.1149E-06 | 0.001644 | 0.0003 | 4.58617E-06 |
| FLRT | 0.003836 | 0.00007 | 1.07011E-06 | 0.003288 | 0.00006 | 9.17235E-07 | 0.003288 | 0.00006 | 9.17235E-07 | 0.003288 | 0.00006 | 9.17235E-07 | 0.003288 | 0.00006 | 9.17235E-07 |
| PYRE | 0.002192 | 0.00004 | 6.1149E-07 | 0.002192 | 0.00004 | 6.1149E-07 | 0.001644 | 0.00003 | 4.58617E-07 | 0 | 0 | 0 | 0.002192 | 0.00004 | 6.1149E-07 |

| | | | | | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| BaATR | 0.004932 | 0.009 | 0.000137585 | 0.004932 | 0.009 | 0.000137585 | 0.004932 | 0.009 | 0.000137585 | 0.004932 | 0.009 | 0.000137585 | 0.00548 | 0.01 | 0.000152872 |
| CHRY | 0.003288 | 0.0006 | 9.17235E-06 | 0.003288 | 0.0006 | 9.17235E-06 | 0.003288 | 0.0006 | 9.17235E-06 | 0.003288 | 0.0006 | 9.17235E-06 | 0.003288 | 0.0006 | 9.17235E-06 |
| BbFN | 0.006028 | 0.011 | 0.00016816 | 0.006028 | 0.011 | 0.00016816 | 0.006028 | 0.011 | 0.00016816 | 0.006028 | 0.011 | 0.00016816 | 0.006028 | 0.011 | 0.00016816 |
| BkFN | 0.006028 | 0.011 | 0.00016816 | 0.006028 | 0.011 | 0.00016816 | 0.006028 | 0.011 | 0.00016816 | 0.006028 | 0.011 | 0.00016816 | 0.006028 | 0.011 | 0.00016816 |
| BaPYR | 0.003288 | 0.06 | 0.000917235 | 0.003288 | 0.06 | 0.000917235 | 0.003288 | 0.06 | 0.000917235 | 0.003288 | 0.06 | 0.000917235 | 0.003288 | 0.06 | 0.000917235 |
| Ind[1,2,3]PYR | 0.003836 | 0.007 | 0.000107011 | 0.003836 | 0.007 | 0.000107011 | 0.003836 | 0.007 | 0.000107011 | 0.003836 | 0.007 | 0.000107011 | 0.003836 | 0.007 | 0.000107011 |
| DiB[a.h}ANT | 0.006576 | 0.6 | 0.009172346 | 0.006576 | 0.6 | 0.009172346 | 0.007124 | 0.65 | 0.009936708 | 0 | 0 | 0 | 0.007124 | 0.65 | 0.009936708 |
| B[g,h,i]PE | 0.003288 | 0.0006 | 9.17235E-06 | 0.003288 | 0.0006 | 9.17235E-06 | 0.003288 | 0.0006 | 9.17235E-06 | 0.003836 | 0.0007 | 1.07011E-05 | 0.003288 | 0.0006 | 9.17235E-06 |
| ∑PAH16 | 0.089324 | 0.70016 | 0.010703517 | 0.098092 | 0.69996 | 0.01070046 | 0.087132 | 0.74994 | 0.011464516 | 0.08494 | 0.10011 | 0.001530407 | 0.092612 | 0.75095 | 0.011479956 |

**Table 6** DDI, B(A)P and ECR for Blue Whiting (*Micromesistius poutasou*)

| **PAH** | **Isara** | | | **Ipara** | | | **Iperu** | | | **Ilishan** | | | **Sagamu** | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | DDI | B(A)P | ECR | DDI | B(A)P | ECR | DDI | B(A)P | ECR | DDI | B(A)P | ECR | DDI | B(A)P | ECR |
| NAPT | | 0 | 0 | | 0 | 0 | | 0 | 0 | 0.012604 | 0.00023 | 3.51607E-06 | | 0 | 0 |
| ACTY | 0 | 0 | 0 | 0.07946 | 0.00145 | 2.21665E-05 | 0 | 0 | 0 | 0.015892 | 0.00029 | 4.4333E-06 | 0 | 0 | 0 |
| ACTE | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0.02192 | 0.0004 | 6.1149E-06 | 0 | 0 | 0 |

| | | | | | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| FLUO | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0.018632 | 0.00034 | 5.19766E-06 | 0 | 0 | 0 |
| PHEN | 0 | 0 | 0 | 0.000548 | 0.00001 | 1.52872E-07 | 0 | 0 | 0 | 0.007672 | 0.00014 | 2.14021E-06 | 0.000548 | 0.00001 | 1.52872E-07 |
| ANTR | 0.001096 | 0.0002 | 3.05745E-06 | 0.001096 | 0.0002 | 3.05745E-06 | 0.001096 | 0.0002 | 3.05745E-06 | 0.0137 | 0.0025 | 3.82181E-05 | 0.001096 | 0.0002 | 3.05745E-06 |
| FLRT | 0.001096 | 0.00002 | 3.05745E-07 | 0.001096 | 0.00002 | 3.05745E-07 | 0.001096 | 0.00002 | 3.05745E-07 | 0.006028 | 0.00011 | 1.6816E-06 | 0.001096 | 0.00002 | 3.05745E-07 |
| PYRE | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0.027948 | 0.00051 | 7.79649E-06 | 0 | 0 | 0 |
| BcPT | 0.00274 | | 0 | 0.00274 | | 0 | 0.00274 | | 0 | 0.0137 | | 0 | 0.00274 | | 0 |
| BaATR | 0.00274 | 0.005 | 7.64362E-05 | 0.002192 | 0.004 | 6.1149E-05 | 0.00274 | 0.005 | 7.64362E-05 | 0.023564 | 0.043 | 0.000657351 | 0.00274 | 0.005 | 7.64362E-05 |
| CHRY | 0.00274 | 0.0005 | 7.64362E-06 | 0.002192 | 0.0004 | 6.1149E-06 | 0.002192 | 0.0004 | 6.1149E-06 | 0.028496 | 0.0052 | 7.94937E-05 | 0.002192 | 0.0004 | 6.1149E-06 |
| BbFN | 0.00548 | 0.01 | 0.000152872 | 0.00548 | 0.01 | 0.000152872 | 0.00548 | 0.01 | 0.000152872 | 0.012604 | 0.023 | 0.000351607 | 0.00548 | 0.01 | 0.000152872 |
| BjFN | 0.00548 | | 0 | 0.00548 | | 0 | 0.00548 | | 0 | 0.014796 | | 0 | 0.00548 | | 0 |
| BkFN | 0.00548 | 0.01 | 0.000152872 | 0.00548 | 0.01 | 0.000152872 | 0.00548 | 0.01 | 0.000152872 | 0.014796 | 0.027 | 0.000412756 | 0.00548 | 0.01 | 0.000152872 |
| DiBaANT | 0 | | 0 | 0 | | 0 | 0 | | 0 | 0 | | 0 | 0 | | 0 |
| BePYR | 0.00548 | | 0 | 0.004384 | | 0 | 0.004384 | | 0 | 0.029592 | | 0 | 0.004384 | | 0 |
| BaPYR | 0.004384 | 0.08 | 0.001222979 | 0.004932 | 0.09 | 0.001375852 | 0.00548 | 0.1 | 0.001528724 | 0.015344 | 0.28 | 0.004280428 | 0.00548 | 0.1 | 0.001528724 |
| 3MCOL | 0 | | 0 | 0 | | 0 | 0 | | 0 | 0.165496 | | 0 | 0 | | 0 |
| Ind[1,2,3]PYR | 0.00548 | 0.01 | 0.000152872 | 0.00548 | 0.01 | 0.000152872 | 0.00548 | 0.01 | 0.000152872 | 0.047676 | 0.087 | 0.001329999 | 0.00548 | 0.01 | 0.000152872 |

| | | | | | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| DiB[a.h}ANT | 0.009864 | 0.9 | 0.013758519 | 0.009316 | 0.85 | 0.012994156 | 0.009316 | 0.85 | 0.012994156 | 0.047676 | 4.35 | 0.066499506 | 0.009864 | 0.9 | 0.013758519 |
| B[g,h,i]PE | 0.004932 | 0.0009 | 1.37585E-05 | 0.004932 | 0.0009 | 1.37585E-05 | 0.004932 | 0.0009 | 1.37585E-05 | 0.02192 | 0.004 | 6.1149E-05 | 0.004932 | 0.0009 | 1.37585E-05 |
| DiB[a,l]PY | 0.012056 | | 0 | 0.004932 | | 0 | 0.012056 | | 0 | 0.02466 | | 0 | 0.012604 | | 0 |
| DiB[a,i]P | 0.01918 | | 0 | 0.01918 | | 0 | 0.01918 | | 0 | 0.054252 | | 0 | 0.01918 | | 0 |
| DiB[a,h]PY | 0.017536 | | 0 | 0.017536 | | 0 | 0.017536 | | 0 | 0 | | 0 | 0.017536 | | 0 |
| ∑PAH16 | 0.105764 | 1.01662 | 0.01554 | 0.176456 | 0.97698 | 0.014935 | 0.104668 | 0.98652 | 0.015081 | 0.638968 | 4.82372 | 0.073741 | 0.106312 | 1.03653 | 0.015846 |

## 4. Conclusion

Priority PAHs were found in all the samples analyzed in this work. While their concentrations of benzo [a]pyrene which is a benchmark PAH relative to which the toxicity of other PAHs are usually calculated was below the 5 µg/kg guideline value, the bioaccumulation potentials of the PAHs detected in this work should be well considered. The high human risks observable potential of the PAHs due to consumption of these fishes also calls for attention. Therefore fish processors in the study area should be educated as to safer processing method that could eliminate or reduce the risk of exposure to PAHs. Alternatively, to preserve organoleptic preferences for the flavour of smoked fish, the use of approved smoke flavourings could also be promoted.

## Compliance with ethical standards

*Disclosure of conflict of interest*

Authors declare that there is No conflict of interest.

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