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Prevalence of per- and polyfluoroalkyl substances (PFASs) in marine seafood from the Gulf of Guinea

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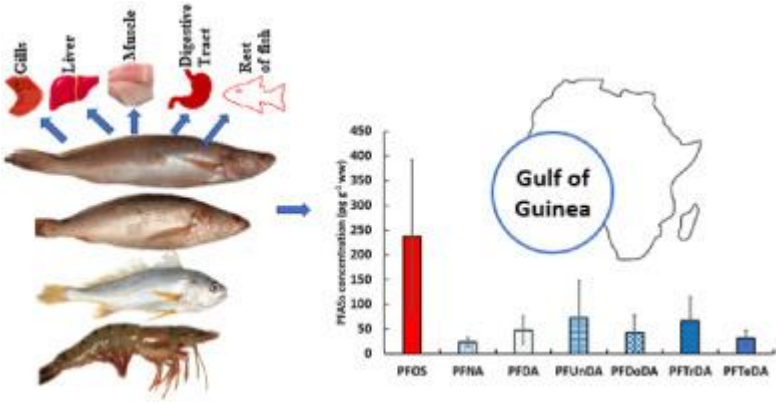
Abstract :

PFASs are ubiquitous in the global environment due to their wide use, persistence and bioaccumulation, and are of concern for human health. This study investigated the levels of PFASs in seafood with a view to provide knowledge on the occurrence of PFASs in marine resources and to evaluate seafood safety and human health risk via dietary exposure to coastal communities in the Gulf of Guinea, where there is currently very little data. The sum of targeted PFASs was between 91 and 1510 pg g-l ww (mean 465 ± 313 pg g-l ww), with PFOS and long-chain PFCAs prevailing. The concentrations of PFASs in the three species of croakers were species- and location-dependent, with habitat and anthropogenic pressure as likely drivers of the differences. Significantly higher contamination levels were found in male croakers. The trophic transfer and biomagnification of PFASs from shrimps to croakers was evidenced for PFOS and long-chain PFCAs (with a significant increase of contaminants from the prey to the predator). The calculated estimated daily intakes (EDIs) and hazard ratio (HR) for PFOS in croakers (whole fish and muscles) and shrimp were lower than the European Food and Safety Agency's recommended level for PFOS (1.8 ng kg−1 day−1) and below the HR safety threshold value of 1. From the results, based on present safety limits, PFOS levels in croakers and shrimps from the Gulf of Guinea do not pose immediate health risks to the human population. This study provides the first insight regarding the distribution of PFASs in seafood from the tropical NE Atlantic region of the Gulf of Guinea and highlights the need for further monitoring across the Gulf.

Graphical abstract



Highlights

► PFOS and long-chain PFCAs are predominant PFASs in marine seafood from the Gulf of Guinea. ► The concentrations of PFASs in croakers were species- and location-dependent. ► Significantly higher contamination levels in male croakers compared to females. ► Biomagnification from shrimps to croakers was evidenced for PFOS and long-chain PFCAs. ► Calculated BMFs decreased with increasing PFCA chain lengths.

Keywords : West Africa, chemical pollution, contaminants of emerging concern, fish, shrimp, risk assessment

1. Introduction

Seafood is a major source of protein and income for millions of people in the Gulf of Guinea (ECOWAS and FAO, 2020). Although the Gulf of Guinea is the smallest shelf area of the four main tropical regions of the world (Briggs, 1974), the region is host to the Canary, Guinea and partly the Benguela current large marine ecosystems and supports some of the most productive fisheries in the world (Polidoro et al., 2016; ECOWAS and FAO, 2020). Among exploited seafood in the sub-region, croakers (Family: Sciaenidae, Genus: *Pseudotolithus*) are widely distributed across the gulf (Longhurst, 1964; Chao and Trewavas, 1990; Sossoukpe et al., 2013) and account for about 40% of the value of landings made by fishing operations (Bayagbona, 1969; Anyanwu, 1983; Etim et al., 1994; Edwards et al., 2001). Nine species of croakers have been described, but the most dominant species include the bobo croaker (*P. elongatus,* Bowdich 1825), cassava croaker (*P. senegalensis,* Valenciennes 1833) and longneck croaker (*P. typus,* Bleeker 1863), (Longhurst, 1964; Anyanwu, 1983; Isangedighi, 2014).

In the last five decades, there have been increasing urbanization, industrial and agricultural activities within the region leading to the input of persistent and emerging contaminants into

the Gulf of Guinea. Despite a growing research effort on contaminant distribution in seafood, there is still a dramatic lack of knowledge in the region. Previous studies have focused on heavy metals (Okuo and Okolo, 2003; Oguguah et al., 2017; Effah et al., 2021), polycyclic aromatic hydrocarbons (Iwegbue et al., 2015; Sogbanmu et al., 2019), organochlorine pesticides (Unyimadu et al., 2018a, b) and polychlorinated biphenyls, but not on PFASs in seafood from the region. PFASs are a large family of synthetic and ever-expanding fluorinated chemicals whose production started in the 1940s (Buck et al., 2011; OECD, 2018). Partial (polyfluorinated) or full replacement (perfluorinated) of hydrogen with fluorine atoms creates highly stable molecules with unique physicochemical properties (Buck et al., 2011). The increasing production of PFASs and their wide use in several ranges of products (Gluge et al., 2020) have led to their environmental release, with ubiquitous presence in the global environment and elevated levels reported in tissues of terrestrial and aquatic animals (Buck et al., 2011; Ahrens and Bundschuh, 2014; Dai and Zeng, 2019). The advancement in analytical tools within the last 20 years and the improvement in elucidating the behaviour, transport and fate of PFASs in environmental matrices and toxicological studies have greatly raised scientific, regulatory, and public health concerns (Ng et al., 2021). These concerns have led to efforts to restrict and ban the production and use of certain long-chain PFASs. Despite the inclusion of selected PFASs in the Stockholm Convention, recent evidence shows there are still widespread and elevated levels of PFASs in water, soil, biota and humans (De Silva et al., 2021; McDonough et al., 2021; Cousins et al., 2022; Groffen et al., 2023), hence the need to urgently address PFASs impact on the global environment by identifying production areas, pinpointing contaminants hotspots and establishing global monitoring programmes to tackle the global burden of PFASs (Ng et al., 2021).

Although multiple studies on the global distribution of PFASs in marine biota have been conducted in Asia, Australia, Europe, North and South America, (Quinete et al., 2009; Buck et

al., 2011; Pan et al., 2018; Fair et al., 2019; Fujii et al., 2019; Taylor et al., 2019; Wang et al., 2020; Munschy et al., 2022), and recently in East, South and North Africa (Verhaert et al., 2017; Ojemaye and Petrik, 2019; Arinaitwe et al., 2020; Fauconier et al., 2020; Munschy et al., 2020a, b; Barhoumi et al., 2022), to our knowledge, no data is available on PFASs distribution in seafood from the Gulf of Guinea. Therefore, there is an urgent need to investigate PFASs levels in seafood to evaluate the implication to the nascent blue economy agenda of the subregion as evidence indicates that fish and other seafood are the main food categories contributing to human exposure (EFSA, 2020). Thus, the aim of this study was to assess the occurrence of PFASs, contaminants of emerging concern, in seafood from the Gulf of Guinea in view of evaluating seafood safety and human health risk. Specific objectives are to determine 1) PFASs levels and profiles in seafood, 2) the geographical distribution in the Gulf of Guinea, 3) the distribution of PFASs in fish tissues, 4) the influence of biological and trophic parameters, 5) the possibility for trophic transfer and 6) human health risk associated with the consumption of PFASs in contaminated seafood. This study represents important baseline information on emerging contaminants in seafood from the Gulf of Guinea.

2. Materials and Methods

2.1. Study Area

Fishery products were collected from coastal communities in the Gulf of Guinea in January 2022 (Figure 1). The Gulf of Guinea consists of 18 countries and adjoining islands covering over 5700 km^2 and it is projected to have a quarter of the world population by 2050 (Morcos, 2021). Four major rivers - Gambia, Volta, Niger and Congo - drain principally into the gulf, carrying input from anthropic activities. Samples were collected from Nigeria and Ghana as representatives of the gulf. Both countries have similar climatic and economic activities. The major economic activities include petroleum production, solid mineral mining, consumer goods

production, agriculture, maritime traffic, and artisanal and trawl fishing. Both countries have an arid region in the north, savannah in the middle and tropical rainforest with rivers draining into the Atlantic Ocean at the southern end. A detailed description of the study area is provided in the supplementary file.

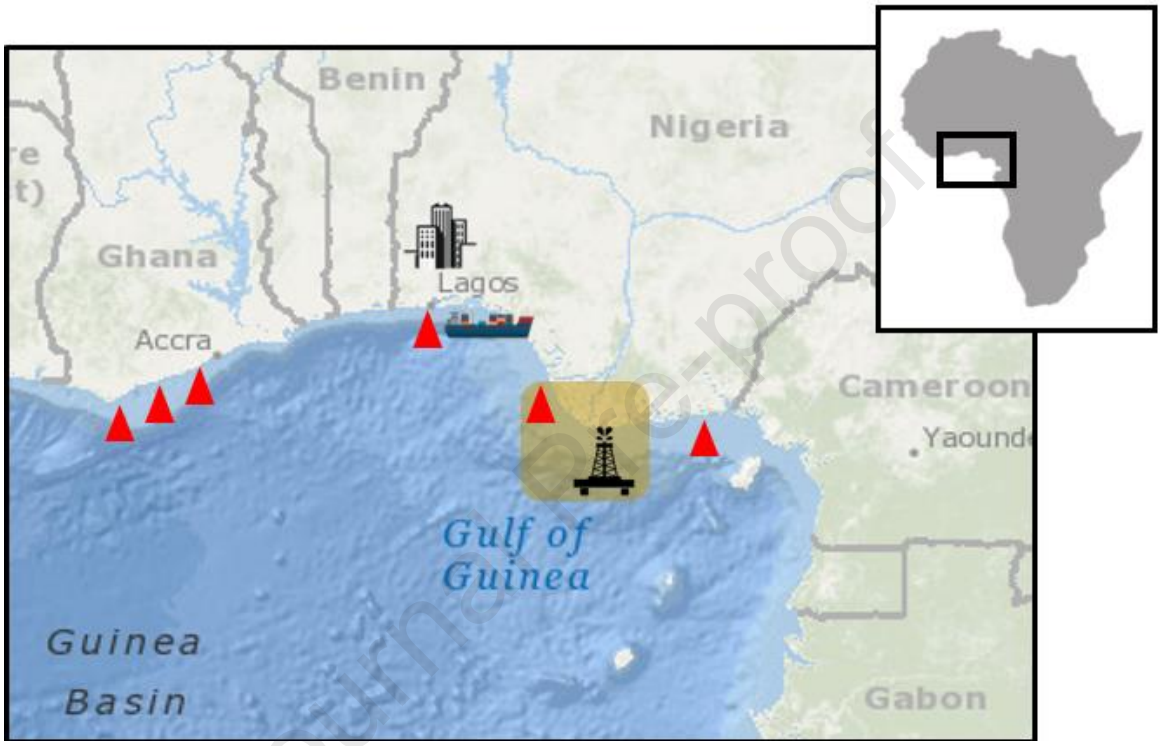


Figure 1. Map showing the Gulf of Guinea. Red points indicate approximate sample collection points in Nigeria and Ghana in January 2022. Vessel and oilrigs indicate higher anthropic activity in Nigeria compared to Ghana.

2.2. Sample Collection

In Nigeria, samples were collected at Ibeno Beach, Ibeno (Akwa Ibom State), Ogulagha, Warri (Delta State) and Makoko, Lagos Island (Lagos State) in southern Nigeria, while in Ghana, samples were collected at Apam Beach, Apam; Elmina Beach Market, Cape Coast (both in Central Region) and Albert Bosomtwi Sam Fishing Harbour, Sekondi-Takoradi (Western Region). Croakers and shrimps freshly caught (<24 h) with fishing nets were purchased from boat landings of artisanal fishers at the various coastal communities between 10 and 23 January

2022. Collected seafood were wrapped in aluminium foil, stored in dry ice and shipped to the organic contaminant laboratory at IFREMER, Nantes, France, and kept in cold storage (-20 ℃) until further treatment. At the lab, vital field information such as sampling date, time, location and coordinates was retrieved and documented.

2.3. Sample Preparation

At the lab, all fish were defrosted, measured (total length, cm), weighed (total weight, g) and sexed for maturity staging by dissection and macroscopic examination of gonads (Supplementary file). Fish were dissected using stainless steel scalpels and blades. The dissecting tools were rinsed with Milli-Q water, ethanol and methanol (MeOH) between each sample to prevent cross-contamination. After sex determination, selected individuals were gutted and the muscles, liver, digestive tract (after emptying the stomach), gills and the rest of the fish were harvested for analysis. The tissues were weighed and placed in appropriate glass containers and were kept in cold storage (-20 ℃) prior to further treatment. For age determination, the cranium of each fish was cut and the otoliths were extracted, cleaned with water and shipped to the IFREMER Sclerochronology Laboratory in Boulogne sur mer (France) to estimate the individual age of each fish according to the international ageing protocol (Mahé et al., 2009; Vitale et al., 2019). The age and growth of fish were determined using external and internal growth rings of the sagittal otoliths. Individual shrimps (whole-body) were cut into bits and stored at -20 °C. All samples were freeze-dried for 48 - 72 h (depending on the size), homogenized using a blender with stainless steel blades and then ground into powder using a ball mill (Retsch, MM400) with bowls and marbles made of stainless steel. The powdered samples were kept in amber bottles in storage cabinets at room temperature before extraction. In total, 81 samples were analysed consisting of 59 muscles of male and female croakers (20 cassava male, 20 longneck male, 9 longneck female and 7 bobo male) and 5 extra tissues of

longnecks (gills, rest of fish, digestive tracts, and liver); 6 samples of shrimps were extracted and analysed (Table S1). Further information on sample preparation is available in the supplementary file.

2.4. Extraction and Analysis

The list of reagents for sample preparation and analysis is given in Table S2. Seafood were extracted and analysed at the Organic Contaminants Laboratory, Ifremer Atlantic Centre using methods described by Munschy et al. (2020) with slight modifications. About 0.2 g of freeze-dried sample was transferred to a 15 mL polypropylene tube and then spiked with ^{13}C-labelled surrogate standards. A liquid-solid extraction (LSE) was performed using 6 mL of a blend of MeOH/KOH (0.01M KOH). The sample was then mechanically agitated and left in contact for one night. The sample was centrifuged (Sigma 3-16L, Sigma Laborzentrifugen, Germany) for 5 min, the supernatant was collected into a 10 mL tube and was evaporated to 1 mL under a gentle stream of nitrogen, at 40 °C. The evaporated sample was purified a on weak anion exchange stationary phase (Waters® Oasis WAX, 150 mg, 6 mL), eluted with MeOH for a neutral fraction containing FOSA and with MeOH/NH_4OH (99.5:0.5, v/v) for an anionic fraction containing the remaining perfluoroalkyl acids. Each fraction was evaporated to 1 mL and further purified using a graphitized carbon stationary phase (Supelco® ENVI-Carb™, 500 mg, 6 mL) eluted with 6 mL of MeOH:acetic acid (80:1, v/v). The extracts were evaporated to 250 µL, transferred into polypropylene injection vials (700 µL), evaporated to dryness under a gentle stream of nitrogen at 40 °C and then reconstituted in 200 µL of a mixture of MeOH:H_2O (50:50, v/v) to which injection standards were added (Table S3).

Analysis was performed using an Acquity ultra-performance liquid chromatograph (UPLC®, Waters Corp.) coupled to a triple quadrupole mass spectrometer (Xevo® TQ-S micro, Waters Corp.) interfaced with a Z-spray™ electrospray ionisation source (Waters Corp.); UPLC

separation was achieved using an Acquity UPLC BEH C_{18} reversed-phase column (1.7 μm, 50 × 2.1 mm, Waters Corp.). Elution solvents were ammonium acetate in water (20 mM) (A) and methanol (B). The mobile phase flow rate was 0.5 mL min^{-1}. The gradient started at 25% B for 0.5 min, was increased to 85% in 4.5 min and 100% in 0.1 min, held for 0.9 min, returned to initial conditions in 0.1 min and held for 2.9 min. A 10 μL of the sample was injected with an automatic injector. The mass spectrometer was operated in negative ionization mode using multiple reaction monitoring (MRM) with argon as the collision gas. Two transitions were recorded per analyte. The capillary voltage was 2.8 kV. The source temperature and probe temperature were 150 °C and 500 °C, respectively. Nitrogen was used as the nebulizing and desolvation gas. Instrumental operations (Table S4), data acquisition and peak integration were performed with MassLynx Software (v.4.1, Waters). The target analytes include PFBS, PFPS, PFHxS, PFHpS, PFOS, PFNS, PFDS, PFDoDS, PFBA, PFPA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnDA, PFDoDA, PFTrDA, PFTeDA, PFPeDA, PFECHS, 6:2 Cl-PFAES, FOSA, 5:3 FTCA, 7:3 FTCA, 6:2 FTS, 8:2 FTS and 10:2 FTS following the abbreviation of PFASs according to Buck et al. (2011). Targeted analytes were quantified using the corresponding isotope labelled standard: PFBS $^{13}C_3$ (used to quantify PFBS and PFPS), PFHxS $^{18}O_2$ (PFHxS), PFOS $^{13}C_8$ (PFHpS, PFOS, PFNS, PFDS, PFDoDS, 6:2 Cl-PFAES and PFECHS), PFBA $^{13}C_4$ (PFBA), PFPA $^{13}C_5$ (PFPA), PFHxA $^{13}C_5$ (PFHxA), PFHpA $^{13}C_4$ (PFHpA), PFOA $^{13}C_8$ (PFOA and 5:3 FTCA), PFNA $^{13}C_9$ (PFNA), PFDA $^{13}C_6$ (PFDA and 7:3 FTCA), PFUnDA $^{13}C_7$ (PFUnDA), PFDoDA $^{13}C_2$ (PFDoDA), PFTeDA $^{13}C_2$ (PFTrDA, PFTeDA and PFPeDA), FOSA $^{13}C_8$ (FOSA), 6:2 FTS $^{13}C_2$ (6:2 FTS), 8:2 FTS $^{13}C_2$ (8:2 FTS) and 10:2 FTS $^{13}C_6$ (10:2 FTS). PFOS $^{13}C_4$, PFBA $^{13}C_3$, PFOA $^{13}C_2$, PFDA $^{13}C_2$ and 8:2 FTS $^{13}C_2D_4$ were added to the purified extracts prior to analysis as injection standards. All standards were purchased from Wellington Laboratories (Guelph, Canada) with a purity > 98%, except for PFPeDA (Chiron AS, Trondheim, Norway, purity 87.6%) and 8:2 FTS $^{13}C_2D_4$ (Cambridge Isotope Laboratories,

Andover, USA, purity >98%). The MRMs of 9:3 FTCA were acquired, but in the absence of a commercial standard, reported values are semi-quantitative only, and based on the response coefficient of 7:3 FTCA. Further details can be found in the supplementary file.

2.5. Quality Assurance and Quality Control (QA/QC)

In order to minimize external and cross-contamination, all samples were processed in a clean laboratory (low dust atmosphere and positive pressure with UV-protected windows) under a fume hood. Chemicals and reagents were of LC-MS grade. Glassware used for sample preparation and extraction was baked overnight at 450 °C. All polypropylene materials were washed twice with MeOH before use. PFASs were quantified by isotopic dilution using ^{13}C-labelled compounds. A nine-point calibration curve ranging from 0.05 to 25 pg μL^{-1} was used to calculate relative response factors and check for linearity.

Laboratory blanks were simultaneously processed and monitored in parallel with the samples to assess contamination throughout the analytical procedure. A commercially purchased mussel sample sequentially shelled, homogenized, freeze-dried and spiked at 0.2 ng g^{-1} dw of each target PFAS was used as an in-house QC and included in each series of analyses to assure repeatability. The results obtained for this QC were used to set up a quality control chart guaranteeing the robustness of the entire analytical procedure. In addition, the laboratory regularly participates in QUASIMEME intercomparison exercises for PFASs, and obtained satisfactory Z-scores, i.e., between -0.9 and +0.2, on a 2022 mussel sample. For the samples, labelled standard recoveries were above 50% (between 51 ± 22% for PFTeDA $^{13}C_2$ and 91 ± 11% for PFNA $^{13}C_5$), except for 6:2 FTS $^{13}C_2$ (117 ± 67%). Laboratory blank levels were low, between 4 pg g^{-1} ww (PFTrDA) and 129 pg g^{-1} ww (PFBA). Limits of quantification (LOQ) values were determined for each target compound in each analysed sample according to Wenzl et al. (2016), i.e., using a signal-to-noise ratio of 3 (peak-to-peak) for the less intense

MRM (qualifier ion). Mean LOQs were between 13 pg g^{-1} ww (PFHpS) and 256 pg g^{-1} ww (PFBA), which are satisfactory to detect marine trace levels. The relative standard deviations of the in-house QC mussel versus target values indicated good accuracy, between 0.2% (PFOA) and 8.1% (PFHpA). Extended uncertainties derived from the method validation were between 24% (PFHpS) and 50% (PFTrDA). Concentrations in samples were blank-corrected and are provided as wet weight (ww). Dry weight content was consistent among croaker muscles (mean of 23.1%, range 18.3 - 25.6%), and among shrimps (22.4%, 21.2 - 23.9%). ∑PFCA concentrations refer to the sum of compounds from PFNA to PFTeDA, with concentrations below LOQs counted as zero, while $\sum_7$ PFAS refers to the sum of PFOS and ∑PFCA. Detailed QA/QC parameters are given in Table S5.

2.6. Relative body burdens

To evaluate the contribution of specific tissues to the whole-body contaminant burden of PFASs, we determined the relative body burdens (RBB) as shown in the equation:

$$RBB_{tissue} = 100 \times \frac{C_{tissue} \times m_{tissue}}{\sum (C_{tissue} \times m_{tissue})}$$

where C_{tissue} is the concentration in a particular tissue (pg g^{-1} ww) and m_{tissue} is the mass of the tissue (g ww).

2.7. Health Risk Assessment

The potential health risk to the human population through the consumption of contaminated seafood (croakers and shrimps) with PFASs was assessed by using the estimated daily intake (EDI, ng kg^{-1} day^{-1}) and hazard ratio (HR). EDI was calculated using methods described by previous authors (Wei et al., 2018; Barhoumi et al. 2022) with the equation:

242

243 $EDI = C \times CR / BW$

244

245 Where C is the mean concentration (ng g^{-1}, ww) of PFASs in seafood (muscles, whole fish and
246 shrimp) samples, CR is the consumption rate of seafood per day (g day^{-1}, ww) and BW is the
247 average body weight (60.7 kg for people in the Gulf of Guinea as reported by Walpole et al.
248 2012). The estimated daily average consumption of fish and shrimp was obtained from the most
249 recent Food Balance Sheets data of the Food and Agriculture Organization (FAO, 2022) using
250 the data for Nigeria in this calculation. The estimated daily consumption was 23.73 and 0.77 g
251 day^{-1} for fish and shrimp respectively. The calculated EDIs were compared to the current
252 tolerable daily intake (TDI) established by the European Food Safety Authority (EFSA). The
253 most recent risk assessment by EFSA set a new TDI of 0.63 ng kg^{-1} day^{-1} (i.e. 4.4 ng kg^{-1} $week^{-1}$) for the sum of PFOA, PFOS, PFNA and PFHxS (Σ_4 PFAS) to evaluate the safety of seafood
255 for human consumption (EFSA et al. 2020). The HR was calculated using the following
256 equation:

257

258 $HR = EDI / RfD$

259

260 The oral reference dose (RfD, ng kg^{-1} day^{-1}) was computed using the TDIs from EFSA. An HR
261 value less than 1.0 indicates a low risk, while a value above 1.0 means a high risk of
262 contaminants to public health.

263

264 **2.8. Data Analysis**

265 All statistical analyses were performed using XLSTAT v.2022 (Addinsoft). Data were
266 summarized into descriptive statistics (mean, standard deviation and percentages). Non-

parametric tests (Mann-Whitney for two samples and Kruskal-Wallis test for independent groups) were applied with a significance level set at $p<0.05$. PFAS concentrations <LOQs were equated to zero and were not included in the data analysis. Figures were plotted using Microsoft Excel, v.2019 (Microsoft Corporation). All the contaminant concentrations in fish and shrimps were reported in pg g^{-1} or ng g^{-1} ww.

Fish biomarkers such as the condition factor (CF), gonadosomatic index (GSI) and liver somatic index (LSI) of croakers were computed following the equations:

$$CF = [\text{weight of fish}/\text{total length of fish}^3] \times 100$$

$$GSI = [\text{weight of gonads}/\text{weight of gutted fish}] \times 100$$

$$LSI = \text{weight of liver}/\text{weight of gutted fish}] \times 100$$

3. Results and Discussion

3.1. Fish Biometrics

Fish biometric data is essential for understanding vital fish statistics for ecological, economic and contaminant studies. The general information on total length, total weight, age class, CF, GSI and LSI of cassava, longneck and bobo croakers is shown in Table 1. As we made an effort to collect fish samples with biometric similarities across the six study sites, the three species showed similar standard lengths in males (26.5±4.8 cm for cassava, 25.2±2.5 cm for longneck and 24.4±3.7 cm for bobo croakers). However, inter-species differences ($p=0.02$) exist for fish weights between cassavas (308.3±143.9 g) and longnecks (198.2±59.5 g), resulting in a significant difference of CF between the two species ($p<0.0001$). Bobo croakers showed significantly higher CFs than did longnecks ($p=0.014$), which relates to their morphology. LSI was similar for all species. Otolith analysis indicated that there were no differences in age between the three species (2 - 3 yrs). In relation to sex, the GSI of female longnecks was

292 significantly (p = 0.001) higher than that of males, while no differences in CF or LSI were
293 observed between the two sexes. No difference (p=0.462) in GSI was observed between old (3-
294 4 yrs) and young (1 yr) female longneck croakers.
295 Biometric characteristics showed a decline in mean fish sizes compared to the period of big
296 croakers in the 1980s (Okpanefe, 1991). Since the mid-1980s, there has been a steady decline
297 in the average size of croakers in captured fishery due to overfishing and bycatch in shrimp
298 trawling (Ambrose et al., 2005; Polidoro et al., 2016). Croakers are estimated to have a lifespan
299 of 15 years, with rapid growth and the attainment of sexual maturity at a very early age
300 (Longhurst, 1964; Francis et al., 2007; Okyere and Blay, 2020). This agrees with most of the
301 samples in our study having fully developed reproductive organs and sexual maturity. The CF
302 reported in this study is similar to the values observed for *P. senegalensis* in Sierra Leone
303 (Oladipo and Tarawallie, 2014), but the GSI and LSI values were 2 - 3 times lower than those
304 reported in the same study (Oladipo and Tarawallie, 2014), and in *P. elongatus* and *P. typus*
305 from Nigeria (Ekanem et al., 2004; SPDC, 2019).
306
307 Table 1. Biometric information (mean±SD and range) for seafood collected from the Gulf of
308 Guinea in January 2022. TL = total length (cm), SL= standard length (cm), GSI =
309 gonadosomatic index, LSI = liver somatic index, CF = condition factor
310

Country State	Location (n)	Weight (g)	TL (cm)	SL (cm)	Sex (age in years)	GSI	LSI	CF
Longneck croakers (29 individuals)								
Nigeria Delta	Ogulagha (7)	128.6±15.3 (112.6-150.0)	25.6±1.3 (24.5-27.5)	22.2±0.6 (21.5-23.0)	M (2-5)	0.2±0.1 (0.1-0.3)	0.3±0.1 (0.1-0.4)	0.8±0.1 (0.6-1.0)
Nigeria Delta	Ogulagha (9)	168.9±44.2 (109.4-239.5)	28.7±2.4 (26.0-33.0)	24.6±2.2 (22.0-28.5)	F (1-4)	1.9±0.3 (1.4-2.7)	0.4±0.1 (0.2-0.6)	0.7±0.1 (0.6-1.0)

Nigeria Akwa Ibom	Ibeno (5)	251.8±33.3 (206.2-283.5)	31.8±1.1 (30.5-33.0)	26.9±1.1 (25.5-28.0)	M (3-4)	0.4±0.2 (0.2-0.6)	0.6±0.1 (0.5-0.7)	0.8±0.1 (0.7-0.8)
Nigeria Lagos	Makoko (3)	221.8±29.6 (200.3-255.5)	31.0±1.8 (29.0-32.5)	25.8±1.6 (24.0-27.0)	M (1-2)	0.2±0.1 (0.2-0.2)	0.4±0.2 (0.2-0.6)	0.7±0.1 (0.7-0.8)
Ghana Sekondi	Albert Harbour (5)	227.8±37.0 (168.1-265.9)	32.4±1.4 (30.0-33.5)	27.4±1.4 (25.0-28.5)	M (1-2)	0.1±0.1 (0.0-0.1)	0.2±0.1 (0.2-0.3)	0.7±0.1 (0.6-0.7)
Cassava croakers (20 individuals)								
Nigeria Lagos	Makoko (5)	287.7±45.5 (242.0-351.1)	31.4±2.8 (26.8-34.0)	26.7±1.6 (25.0-28.5)	M (1-3)	0.3±0.1 (0.2-0.5)	0.3±0.1 (0.3-0.5)	0.9±0.2 (0.8-1.3)
Ghana Sekondi	Albert Harbour (5)	385.9±84.0 (252.5-451.3)	35.6±2.7 (31.5-38.0)	29.5±1.9 (26.5-31.0)	M (1-2)	0.3±0.2 (0.1-0.6)	0.5±0.2 (0.2-0.8)	0.8±0.1 (0.8-0.9)
Ghana Cape Coast	Elmina Beach (5)	111.0±74.1 (65.4-241.2)	23.1±3.7 (20.0-29.5)	19.4±2.7 (17.0-24.0)	M (1-2)	0.2±0.1 (0.1-0.3)	0.3±0.1 (0.2-0.4)	0.8±0.1 (0.6-1.0)
Ghana Apam	Apam Beach (5)	448.7±49.3 (403.1-524.1)	35.7±0.6 (35.0-36.5)	30.4±0.9 (29.5-31.6)	M (2-3)	0.2±0.1 (0.2-0.3)	1.0±0.4 (0.4-1.4)	1.0±0.1 (0.9-1.1)
Bobo croakers (7 individuals)								
Nigeria Delta	Ogulagha (5)	182.8±43.3 (153.2-256.7)	26.1±2.1 (23.0-28.0)	23.5±3.9 (21.0-30.5)	M (1-5)	0.2±0.1 (0.2-0.3)	0.4±0.1 (0.4-0.5)	1.1±0.4 (0.8-1.6)
Nigeria Akwa Ibom	Ibeno (2)	272.5±80.9 (215.3-329.8)	31.7±3.2 (29.5-34.0)	26.5±2.8 (24.5-28.5)	M (3-4)	0.3±0.1 (0.2-0.3)	1.1±0.1 (1.0-1.1)	0.8±0.1 (0.8-0.8)
Giant tiger shrimps (6 individuals)								
Nigeria Delta	Ogulagha (6)	17.9±1.8 (15.5-20.7)	15.1±0.7 (14.2-16.1)	3.2±0.1 (3.1-3.4)	-	-	-	0.5±0.1 (0.5-0.5)

311

312 **3.2. PFASs Distribution in Relation to Site and Species**

313 Among the 28 targeted PFASs quantitatively analysed, 17 were found above the LOQ in all
314 samples (n=82) with 7 compounds having detection frequencies (DFs) above 60% (PFOS,
315 PFNA, PFDA, PFUnDA, PFDoDA, PFTrDA and PFTeDA, 61 - 94%), while PFHpS, PFPA,

PFHxA, PFHpA, PFOA, FOSA, 5:3 FTCA and 7:3 FTCA were seldom detected (1-37%) in the samples (Table S5).

The emphasis in our study is on male croakers and this will be reflected in the results for the seven PFASs highlighted above. The $\sum_7$ PFAS concentrations in the muscles of male croakers (n=47) ranged from 91 to 1510 pg g^{-1} ww with a mean of 465±313 pg g^{-1} ww (mean of individual PFAS presented in Figure 2). Detailed concentrations per individual compound and species are given in Table S6.

In relation to profiles of PFASs in the muscles of male croakers, a non-parametric Kruskal-Wallis test showed that the level of PFOS was significantly ($p<0.0001$) higher than that of PFNA, PFDA, PFUnDA, PFDoDA, PFTrDA and PFTeDA (Figure S1). The PFOS concentration (52±11% of $\sum_7$ PFAS) was 3 - 5 times higher than that of PFTrDA (15±6%), PFUnDA (14±6%) and PFDA (10±3%) and 6 - 8 times higher than that of PFNA (8±6%), PFDoDA (7±3%) and PFTeDA (6±7%).

In this study, the presence of PFASs (PFOS and PFCAs) in the muscle of croakers collected from the study sites in the Gulf of Guinea supports the evidence of the ubiquity of PFASs in the global environment. PFASs presence in the gulf could be from both atmospheric and transboundary deposition of precursors (Yamashita et al., 2005; Muir and Miaz, 2021) and from the local use and degradation of PFAS-laden industrial and domestic products, although understanding the relative importance of these sources would require further work.

The elevated level of long-chain PFASs reported in this study is consistent with previous studies indicating higher bioaccumulation propensities for long-chain PFASs in fishes compared to short-chain compounds (Fujii et al., 2019; Pan et al., 2021; Vi et al., 2022). In our study, PFOA was below the LOQ in all samples, which is consistent with previous studies also reporting the detection of little or no PFOA in fish tissues (Carlsson et al., 2016; Diao et al., 2022). In contrast, the PFOS concentration was significantly higher than that of the rest of the long-chain

compounds. Previous laboratory and field studies have demonstrated that precursors like FOSA and FTOHs are biotransformed into PFOS and PFCAs, respectively, in soil or sediments via microbial mediation, ingestion or in vivo metabolization in higher organisms (Martin et al., 2010; Butt et al., 2014). Among PFCAs, long- and odd-chain compounds like PFTrDA and PFUnDA dominated the distribution of PFASs in croakers compared to even-chain compounds (PFDA, PFDoDA and PFTeDA). Fujii et al. (2015) and Carlsson et al. (2016) reported higher odd-chain lengths (PFTrDA and PFUnDA) compared to even-chain compounds in fish and shrimp. This trend which is prevalent with PFCA distribution in marine biota is associated with the transformation of fluorotelomers such as FTOHs and has been discussed elsewhere (Martin et al., 2004; Butt et al., 2010; Rotander et al., 2012; Munschy et al., 2020).

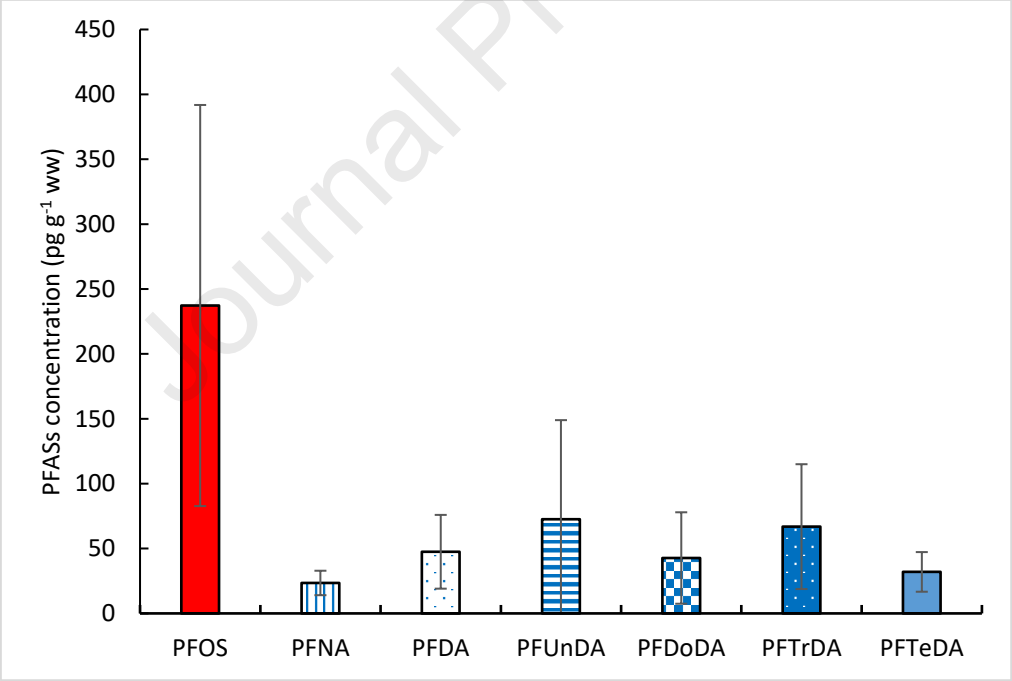


Figure 2. PFASs concentration (mean±SD in pg g^{-1} ww) in the muscle of male croakers collected from the Gulf of Guinea in January 2022

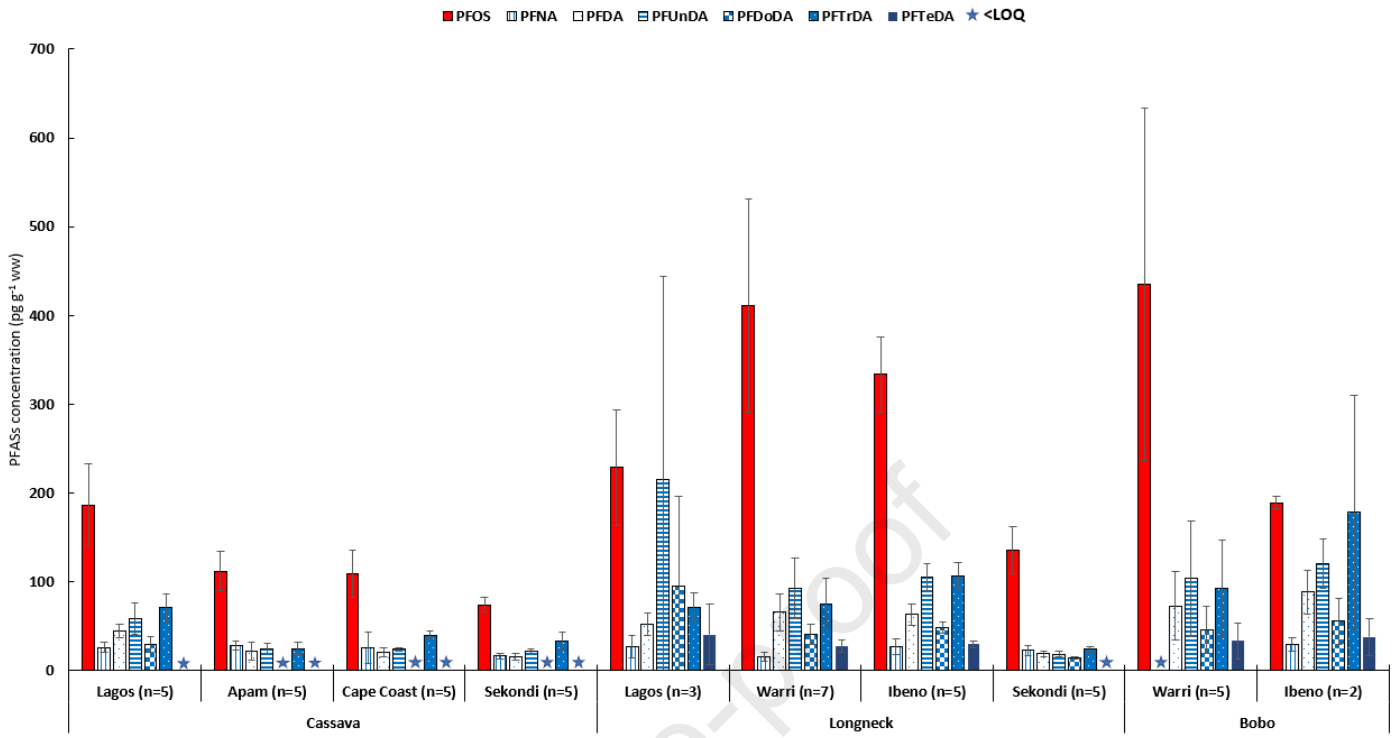


Figure 3. Individual PFAS concentrations (mean±SD in pg g^{-1} ww) in the muscle of male croakers across the sampled sites in Nigeria and Ghana.

An overview of PFASs distribution recorded across the six sites is shown in Figure 3 and concentrations are given in Table S6. For the three species of croakers, the $\sum_7$ PFAS ranges were 91 - 572, 168 - 1073 and 524 - 1510 pg g^{-1} ww for cassava, longneck and bobo croakers, with a mean of 232±126, 594±274 and 761±351 pg g^{-1} ww, respectively.

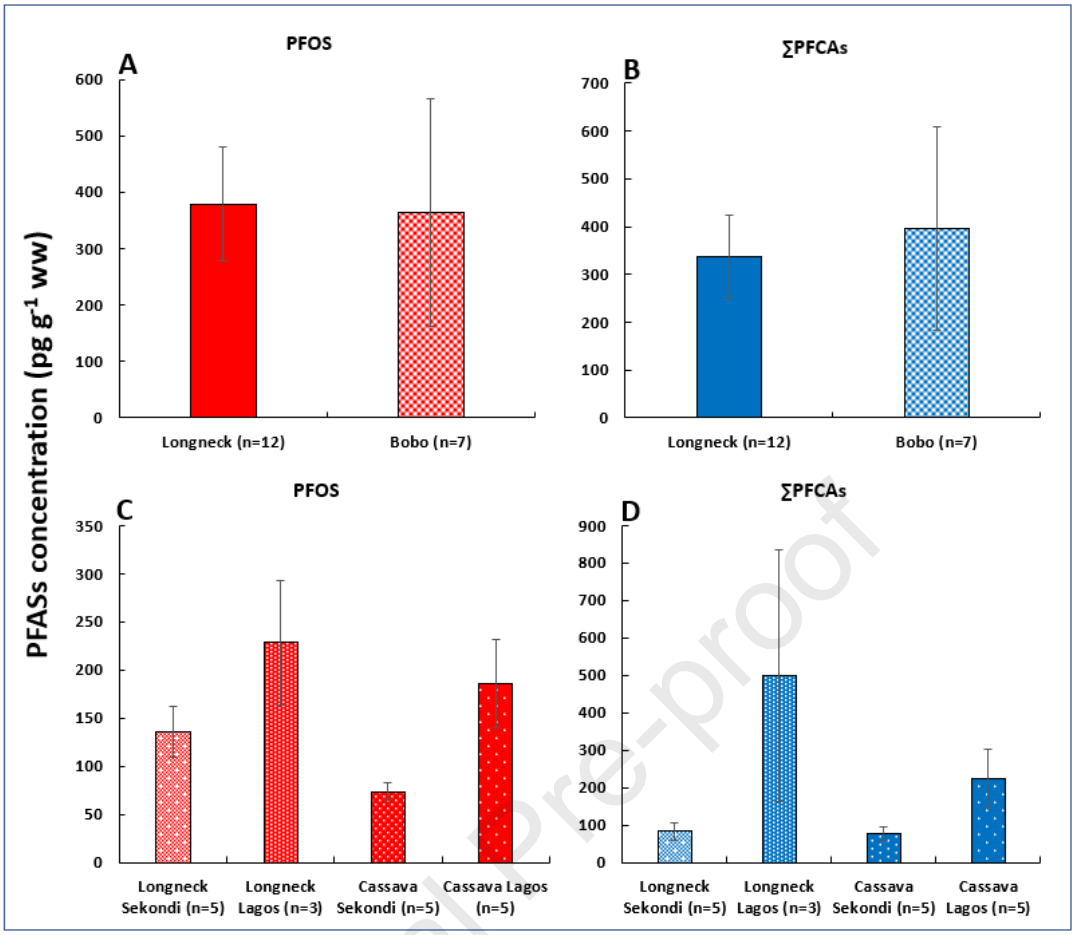
The concentrations of PFASs in the three species showed variation with respect to study sites. For cassava croakers along the four study sites, the PFOS concentration in Lagos (Nigeria) was significantly ($p<0.0001$) higher than the values reported in Sekondi ($p=0.002$) in Ghana. A similar trend was observed for PFDA and PFUnDA, across the four study sites. For longnecks, the PFOS concentration in Warri was significantly higher ($p=0.0021$) than that in Sekondi ($p=0.001$). Significant differences ($p<0.0001$) were also reported for the rest of the compounds (PFNA, PFDA, PFUnDA, PFDoDA, PFTrDA, PFTeDA) across the four sites except for PFUnDA and PFTrDA in Lagos which were not different (Figure 3).

Differences in PFOS concentrations between species showed that longnecks exhibited higher levels than cassavas in Sekondi ($p=0.02$) and to a lesser extent in Lagos (non-significant but $n=3$ for longnecks), while bobos and longnecks did not show any significant differences in Warri or Ibeno. Given this trend, the variation in contaminant distribution in croakers appears to be species-specific and could be traceable to their habitat. Although the ecology of croakers is still only partly known, cassava croaker has been described as mainly restricted to the coastal region, while longneck and bobo enter estuaries and lagoons, with bobo moving further into creeks and rivers (Ajayi, 1981; Isangedighi, 2014). The ecology of bobo and longneck croakers predisposes them to greater exposure to contaminants entering aquatic ecosystems, compared to cassava croakers that are restricted to the coastal zone (Longhurst, 1964; Isangedighi, 2014). Fishes found in estuarine areas may show elevated PFAS exposure compared to species in offshore coastal waters (Munoz et al., 2017).

The higher level of PFASs in cassava croakers from Lagos compared to any of the three sites in Ghana is indicative of the existence of local sources of PFASs in the Gulf of Guinea, as increased anthropic stress from the Lagos region is expected to enter the gulf. Over 80% of all industries in Nigeria and within the gulf are situated in the Lagos metropolitan area, coupled with a population density 10 times higher than any of the three cities in Ghana. Industrial emissions and municipal wastewater enter streams, rivers and the Lagos lagoon before reaching the coastal region, polluting nursery grounds for fish and shrimp - the main prey items for croakers (Oyewo and Don-Pedro, 2003; Oguguah et al., 2017).

As observed with cassava croakers, PFAS concentrations in longneck croakers from any of the three sites in Nigeria were generally higher than those reported in Sekondi. The high levels of PFOS and long-chain PFCAs recorded in croakers from Warri and Ibeno compared to Lagos seem unexpected given the lower anthropic pressures in both sites compared to Lagos, but might reflect location-specific sources possibly from industrial activities within the region.

Warri and Ibeno are part of the Niger Delta region with over five decades of oil exploration and production as the main industrial and economic activities. Increased levels of PFASs in both sites could be associated with the application of PFAS surfactants in oil production (Gluge, et al., 2020), oil spill control and combating fire incidents in the petroleum industry. PFAS products have been used extensively in oil drilling and in enhanced oil recovery in the petrochemical industry due to their unique physicochemical properties, especially as surfactants and oil additives (Bao et al., 2017; Xu et al., 2017; Meng et al., 2021). They are also used as aqueous film-forming foams (AFFFs) for combating fire incidents and for in-situ burning of spilt oil in soil and water (Meng et al., 2021). Elevated levels of short- and long-chain PFASs, associated with oil exploitation and refinery activities have been reported in surface water and sediment from Dagang Oilfield, Tianjin, China compared to areas far from the oilfields (Meng et al., 2021). Another possibility is the introduction of contaminants from the Niger River into the gulf. However, the association of increased PFAS levels in Warri and Ibeno judging from a single study remains speculative. The specific sources of contaminants in the area have not been fully described, and would include freshwater inputs, runoff from urban areas, as well as direct release from industries (including petrochemistry). The site differences in Nigeria require future investigation of environmental matrices and biota to elucidate potential localized sources of PFASs in the region.



418

419 Figure 4. PFOS and ∑PFCA concentrations in muscles of male croakers collected from the Gulf of Guinea in
420 January 2022. A) PFOS in longneck and bobo croakers from Warri and Ibeno, B) PFCAs in longneck and bobo
421 croakers from Warri and Ibeno, C) PFOS in longneck and cassava croakers from Sekondi and Lagos, D) PFCAs
422 in longneck and cassava croakers from Sekondi and Lagos (∑PFCA concentrations refer to the sum of compounds
423 from PFNA to PFTeDA).

424

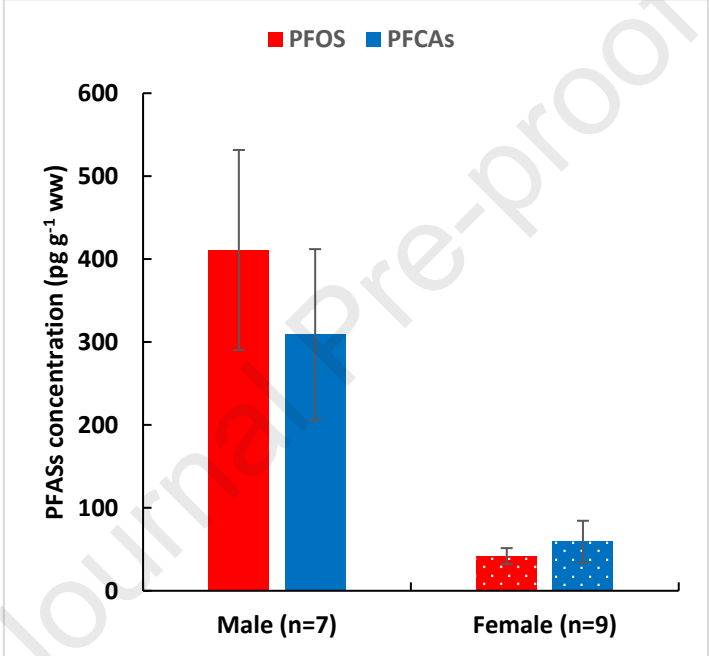
425 Inter-species comparison of PFOS and ∑PFCA concentrations was tested for longneck and
426 bobo croakers from Warri and Ibeno as well as for longneck and cassava croakers collected
427 from Lagos (Nigeria) and Sekondi (Ghana). In reference to longneck and bobo croakers from
428 Warri and Ibeno, no significant differences were reported in PFOS or PFCAs. For croakers
429 collected from Nigeria and Ghana, we observed that the PFOS concentration in longnecks from
430 Lagos was significantly ($p=0.002$) higher than that from Sekondi, while in cassava croakers
431 from Lagos, it was also higher ($p=0.0001$) than in those from Sekondi. A similar trend was also

observed for PFCAs between Lagos and Sekondi for longneck ($p<0.0001$) and cassava ($p=0.0001$) croakers respectively (Figure 4).

3.3. Influence of Sex and Age on PFAS Levels in Croakers

The influence of biological parameters such as sex and age on PFAS distribution was assessed in longneck croakers from Warri. The mean concentrations of PFASs in male and female longneck croakers are shown in Figure 5. Highly significant differences were observed in PFOS ($p=0.003$) and PFCA ($p=0.001$) concentrations between male (n=7) and female (n=9) longneck croakers, with 5 - 10 times higher levels in males. The influence of sex on bioaccumulated PFAS levels has been documented in a limited number of studies, which generally concluded in the absence of an influence of sex on concentrations (Schultes et al., 2019; Arinaitwe et al., 2020; Macorps et al., 2022). In a study in German freshwater fish, slightly higher levels were reported in male vs female (both in fillets and whole fish extracts), sex being one of several confounding factors (Rudel et al., 2022). The lower levels of PFASs reported in female croakers in our study could be attributed to a loss of contaminants associated with spawning. Croakers are asynchronous partial spawners with continuous spawning throughout the year in Nigeria, Cameroon, Ivory Coast and Sierra Leone (Ekanem et al., 2004; Francis et al., 2007; Olapade and Tarawallie, 2014; Tia et al., 2017). Aside from having year-round spawning, two peak spawning periods are recognized in the species across the gulf, which synchronize with the temperature regimes in the region (Longhurst, 1964; Ekanem et al., 2004). The first reproduction peak is between December and March, which reflects the high temperatures of the dry season, and the second reproduction peak is from July to September, which conforms with the high temperatures of August (Longhurst, 1964; Ekanem et al., 2004). Croakers for this study were collected in January, coinciding with the first peak period for spawning which could explain the decreased level of PFASs found in female fishes. The year-round spawning of

457 croakers could aid the elimination or depuration of PFASs in female croakers. In a synthesized
458 review, Madenjian (2011) inferred that spawning causes the drastic reduction of
459 polychlorinated biphenyls in female trout and salmon compared to males. Another potential
460 factor could be more PFAS intake by higher food consumption in male croakers (Madenjian et
461 al., 2016). In both genders, no difference was found for PFAS concentration in relation to the
462 age of croakers (Figure S2).
463



464

465 Figure 5. PFOS and ∑PFCA distribution with relation to sex in the muscles of longneck croakers collected from
466 Nigeria (Warri) in January 2022
467

468 **3.4. Tissues Distribution of PFASs in fish**

469 Besides those in muscles, PFAS concentrations were investigated in four tissues for longneck
470 croakers. Aside from PFOS and long-chain PFCAs (PFDA, PFUnDA, PFDoDA, PFTrDA and
471 PFTeDA) having DFs of 70 - 100%, FOSA had a DF of 100% and 7:3 FTCA was found in the
472 liver only. The $\sum_9$ PFAS (i.e. the summation of $\sum_7$ PFAS, as previously defined, FOSA and 7:3
473 FTCA) concentration in the tissues of longnecks ranged from 644 pg g^{-1} ww in muscles to 16775

474 pg g^{-1} ww in the liver of longneck croakers. For the respective tissues, the highest mean
475 concentration of $\sum_9$ PFASs was found in the liver (8342±5372 pg g^{-1} ww), followed by the
476 digestive tract (3854±450 pg g^{-1} ww), gills (2456±750 pg g^{-1} ww), rest of the fish (1859±355 pg
477 g^{-1} ww) and the muscle (817±178 pg g^{-1} ww), (Figure 6). Across the tissues, the concentrations
478 of PFOS and PFCAs (PFDA, PFUnDA, PFDoDA, PFTrDA and PFTeDA) were significantly
479 ($p<0.0001$) higher in the liver compared to the muscle.
480 Previous studies also reported higher concentrations of PFASs in the liver compared to the
481 muscles of fish (Zafeiraki et al., 2019; Fauconier et al., 2020). PFASs readily partition into
482 phospholipids and specific proteins in biota (De Silva et al., 2021). They also bind preferentially
483 to blood serum and albuminous proteins. Albumin, an important carrier of PFOS and PFOA, is
484 principally synthesized in the liver (Wang et al., 2014; Forsthuber et al., 2020). As the primary
485 site for bioaccumulating and detoxifying certain pollutants in animals, the liver is the target
486 organ for contaminants including PFASs (Wang et al., 2014). As observed in our study,
487 precursors like FOSA and 7:3 FTCA which were rarely observed in muscles were found in
488 other tissues, particularly in the liver, which could result from either increased detection of the
489 generally higher levels of PFASs in the liver, or from their presence as intermediates in hepatic
490 metabolism of precursors. As observed in our study, Soerensen et al. (2023) also found the
491 highest concentration of PFOS in the liver and kidneys in fish and the lowest concentrations in
492 muscle.
493

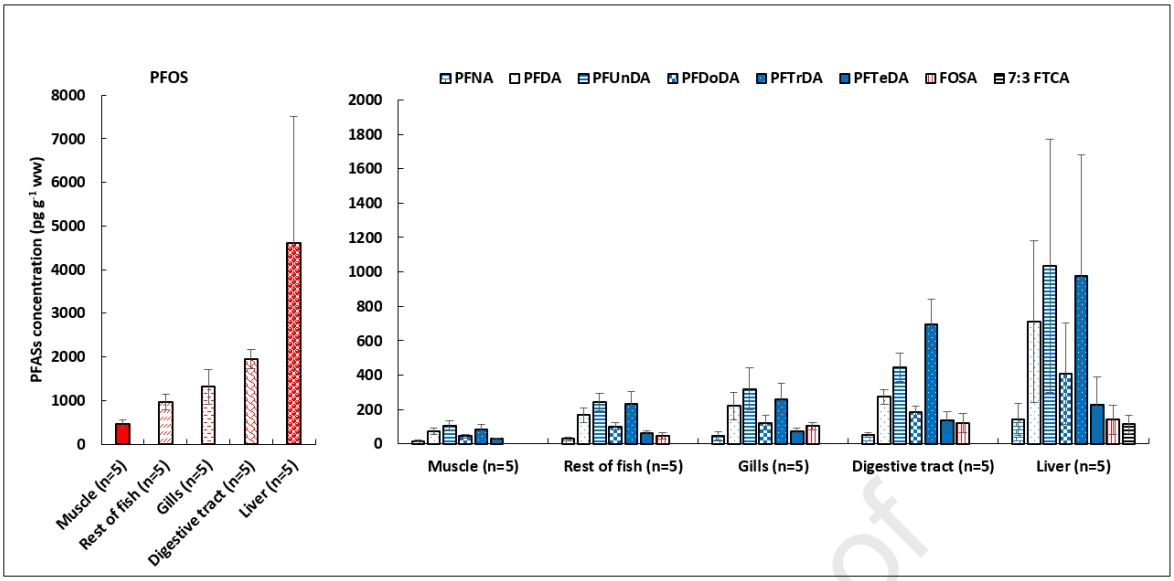


Figure 6. Tissue distribution in longneck croakers collected from Warri in Nigeria from January 2022 (concentrations, pg g^{-1} ww).

The relative body burden (RBB) helps to ascertain the contribution of each tissue to the levels of contaminants present in longneck croakers with respect to the mass of the fish (Figure S3). Among the various tissues, in longneck croakers, the highest RBB for PFASs was reported in the rest of the fish, accounting for 55-68% depending on the compound, compared to the muscles (32-38%), gills (3-5%), digestive tract (3-5%), and liver (1-2%). PFOS and PFCAs (PFDA, PFUnDA, PFDoDA, PFTrDA and PFTeDA) showed similar contributions in all tissues, suggesting the absence of a significant compound-specific partitioning. The high contribution of the rest of the fish, composed of kidneys, heart, blood, bones, brain, eyes, skin and carcass, in the body burden is attributed to both the elevated mass of those remains and the expected higher concentrations likely found in blood-rich tissues. This is similar to the findings of Soerensen et al. (2023) where RBB was dominated by carcass and muscle in marine and freshwater fishes from the Baltic Sea and Northern European lakes. Despite its lower concentration, the muscle is an important tissue to the contribution of the fish's total contaminant burden. In our study, PFOS and PFCAs had similar values for all tissues reported

in croakers. This is consistent with the report of Soerensen et al. (2023) where PFASs followed an overall similar pattern to PFOS for RBB values.

3.5. Trophic Transfer and Biomagnification of PFASs

Pseudotolithus are carnivorous predatory feeders, preying mainly on shrimps and juvenile fish (Tientcheu and Djama, 1994; Osibona and Eniola, 2011; Nunoo et al., 2013; Olatunji, 2021). This is further confirmed by the increased number of shrimps recovered from the stomach of dissected croakers during sample preparation in the lab. The giant tiger shrimp (*Penaeus monodon*, Family: Penaeidae, Fabricius, 1798) is an economically important and abundant seafood in the Gulf of Guinea. It also serves as a vital food source for several fish species in the gulf including croakers (Nunoo et al., 2013; Olatunji, 2021). To ascertain the trophic transfer and bioaccumulation potential of PFASs, we investigated the levels of PFASs in shrimps - a direct prey item for croakers. Residents around the gulf majorly consume whole shrimps (non-gutted) so we analysed whole shrimps to reflect the actual exposure to PFASs.

The $\sum_7$ PFAS concentration in shrimps (n=6) from Warri ranged from 204 to 461 pg g^{-1} ww, with a mean of 326±98 pg g^{-1} ww. PFOS (114±31 pg g^{-1} ww) and PFTrDA (85±22 pg g^{-1} ww) were the most abundant in shrimps, with mean levels 2 to 4 times higher than for other PFCAs. Traces of precursors such as FOSA, 5:3 FTCA and 9:3 FTCA were also found in shrimps close to the LOQ.

Investigating the uptake of contaminants from the environment and their potential transfer from one trophic level to another is essential in understanding contaminant transfer across the food chain. Whole-body ww concentrations were calculated in longneck croakers (n=5) from the contribution of all tissues, as discussed in the previous section. PFOS and long-chain PFCA concentrations in longneck croakers were significantly higher ($p=0.004$) than those in shrimps except for PFTeDA which was similar in both species (Figure 7).

The elevated levels of PFOS and PFCAs observed in longnecks compared to shrimps indicate the likelihood of a trophic transfer of these compounds from a lower to a higher trophic level and is consistent with previously reported results in tropical oceans (Fauconier et al., 2020; Munschy et al., 2020; Barhoumi et al., 2022). Munoz et al. (2017) investigated the trophic transfer of PFASs in whole individuals and determined a significant biomagnification between common seabass and white shrimps for PFOS, PFDA, PFDoDA and PFTrDA. Although the evaluation of biomagnification requires analysis of whole individuals (Borgå et al., 2012), more references are available in the literature on targeted organs and are discussed below. Focusing on targeted tissues, Barhoumi et al. (2022) reported lower PFOS levels in the muscles of shrimps compared to the muscles of sea bass at the Bizerte lagoon in Tunisia. Similarly, Fauconier et al. (2020) also indicated that the increased levels of PFOSs in the muscle of *Ambassis natalensis* (feeding predominantly on crustaceans, fish fry, fish eggs and larvae) compared to whole shrimp was influenced by their diet and the higher trophic level they occupy in the food chain. However, Carlsson et al. (2016) reported significantly higher levels of PFOS in whole shrimps compared to halibut fillets, which was attributed to the protein-rich soft tissues of the head in whole shrimp analysis compared to the fillets of halibut. The low levels of FTCAs found in shrimps, close to the LOQ, warrant caution, but the metabolization of precursor compounds in higher trophic levels (Simonnet-Laprade et al., 2019) could explain their absence in fish tissues in our study.

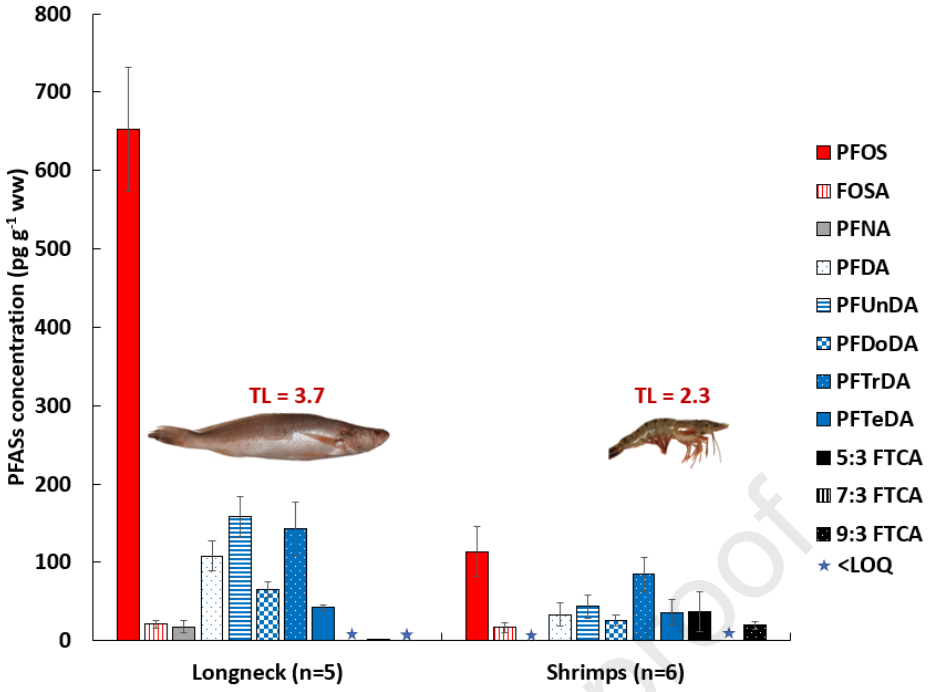


Figure 7. PFAS concentration (pg g^{-1} ww) in whole fish and whole shrimps from Warri, Nigeria. Trophic levels (TL) associated with fish and shrimp are values from Fishbase.

Predator-prey biomagnification factors (BMFs) were calculated by dividing the mean whole-body ww PFAS concentration in the predator by that of its prey. Mean BMFs were above 1 for PFOS and long-chain PFCAs, suggesting biomagnification. The BMF for PFOS was 5.7 while it was 1.2-3.6 for long-chain PFCAs (Table 2), with values decreasing with chain length. Our result is similar to that reported in inland and coastal food webs in Canada (PFOS 5.9), the United States (PFOS = 6.3) and China (PFCAs 2.1-3.7), (Martin et al. 2004; Houde et al. 2006; Xu et al. 2014), but higher than the values reported between fish and shrimp species from Chinese waters (Loi et al. 2011, BMFs of PFOS, PFDA, PFUnDA and PFDoDA between 1.8 and 3.3) and fish and shrimp species from western Europe (Munoz et al. 2017 BMFs of PFOS, PFDA, PFDoDA and PFTrDA between 1.2 and 2.7).

Table 2. Predator/prey relationship (concentration means in pg g^{-1} ww and biomagnification factor) for PFASs in longnecks and shrimps from the Gulf of Guinea.

BMF	PFOS	PFDA	PFUnDA	PFDoDA	PFTrDA	PFTeDA
Longneck (n=5)	652	108	159	66	143	43
Shrimp (n=6)	114	33	44	26	85	36
Predator/Prey BMF	5.7	3.2	3.6	2.5	1.7	1.2

574

575 **4. Human Health Risk Assessment**

576 The potential health risk to the human population through the consumption of contaminated
577 seafood in the Gulf of Guinea was assessed by calculating the EDIs and HR for the
578 concentration of PFASs in the muscles and whole fish and shrimps. Croakers, being a high-
579 protein, lean fish with good nutritional value and minerals, it is essential to investigate the
580 potential health risk of PFASs in croakers to human health (Abimbola, 2016; Njinkoue et al.,
581 2016; Oppong et al., 2021). The calculated EDIs for PFOS in whole fish (longnecks) and shrimp
582 were 0.26 and 0.0014 ng kg^{-1} day^{-1}, respectively, resulting in HR values less than 1 for fish
583 (0.40) and shrimps (0.0023), while in the muscles of the three croakers, EDIs were 0.048, 0.12
584 and 0.14 ng kg^{-1} day^{-1} and HR values were 0.076, 0.18 and 0.23 (Table 3) for cassava, longneck
585 and bobo, respectively. These values are lower than the EFSA-recommended TDIs for PFOS
586 (1.8 ng kg^{-1} day^{-1}) and below the HR safety threshold of 1. In addition, the levels of PFOS
587 recorded in this study (64 - 789 pg g^{-1} ww) are well below the environmental quality standard
588 (EQS_{biota} 9.1 ng/g ww PFOS) threshold value set by the European Union for fishery products.
589 In summary, the current concentrations of PFOS reported in croakers and shrimps from the Gulf
590 of Guinea do not pose a health risk to the human population consuming seafood from the region.
591 The low values reported in this study are similar to those reported in cod across the Pacific
592 Ocean (Fujii et al., 2019), but higher than the values reported in seafood from Tunisia and
593 Vietnam (Barhoumi et al., 2022; Vi et al., 2022). Although different forms of seafood
594 preparation for human consumption have been demonstrated to increase PFAS levels due to

packing materials, loss of moisture during cooking and use of non-stick cookware (Domingo, 2012; Vassiliadou et al., 2015; Taylor et al., 2019), the potential combination from these sources is still far below the threshold to cause immediate health risk to human health. However, prolonged exposure to PFASs through subtle dosage from various food consumption and non-dietary routes may pose human health risks in the long term. As a protein-rich seafood, croakers provide a good source for the regular assessment of proteinophilic contaminants in the Gulf of Guinea on the basis of their PFOS contamination. However, the commonly targeted PFASs such as those determined in the present study are far from representative of the wide variety of PFASs to which ecosystems are exposed (Wang et al., 2017). Besides that, other major contaminants, such as the legacy POPs for which safety thresholds for human health have been defined, would warrant some more investigations.

Table 3. Estimated daily intakes (EDIs) and hazard ratio (HR) of PFASs in whole-body and fillets of seafood for local residents in the Gulf of Guinea

	EDI (ng kg^{-1} day^{-1})							Hazard ratio
Whole-body	PFOS	PFNA	PFDA	PFUnDA	PFDoDA	PFTrDA	PFTeDA	PFOS
Longneck	0.26	0.01	0.04	0.06	0.03	0.06	0.02	0.40
Shrimp	1.4E-03	-	4.2E-04	5.5E-04	3.3E-04	1.1E-03	4.5E-04	2.3E-03
Muscle								
Cassava	4.8E-02	9.2E-03	1.1E-02	1.3E-02	9.5E-03	1.6E-02	1.5E-02	7.6E-02
Longneck	0.12	0.01	0.02	0.04	0.02	0.03	0.01	0.18
Bobo	0.14	0.01	0.03	0.04	0.02	0.05	0.01	0.23

5. Global Comparison of PFASs from Species of Similar Trophic Level

Considering the global distribution of PFASs in seafood with similar trophic levels to those of croakers, we observed generally that most of the studies reported higher PFAS levels compared to croakers from the Gulf of Guinea (Table S7). The values of PFASs reported in this study are

similar to those reported in Brazil, Tunisia and China (Quinete et al., 2009; Pan et al., 2018; Pan et al., 2021; Barhoumi et al., 2022) and lower than those reported in Sweden, the Netherlands and Markets in China (Eriksson et al., 2013; Zafeiraki et al., 2019; Jin et al., 2020). Lower values reported in the Gulf of Guinea and elsewhere are associated with low economic activity, distance from industrial centres, oceanic transport and long-range atmospheric depositions, in contrast with western Europe, North America and China where historical and present-day use and production of industrial chemicals including PFASs are reported (Zhang et al., 2020).

Higher values were attributed to long-range transport and industrial and municipal wastewater in Norway (Carlsson et al., 2016), South Africa (Fauconier et al., 2020), France (Lebigre et al., 2022), Vietnam (Vi et al., 2022) and the United States (Fair et al., 2019); proximity to a fluorochemical industry and direct industrial emissions in China (Wang et al., 2020; Diao et al., 2022), Japan (Fujii et al., 2015; Fujii et al., 2019) and Belgium (Cara et al., 2022); the use of AFFFs for extinguishing a petrochemical fire in the United States (Nolen et al., 2022); and the metal plating industry in China (Chen et al., 2018). Studies in Australia showed particularly high levels of PFASs due to the proximity of the studied areas to an airforce base (Taylor and Johnson, 2016; Taylor et al., 2018, 2019; Taylor, 2020).

6. Conclusion

This study provides the first insight regarding the distribution of PFASs in seafood from the tropical Atlantic region of the Gulf of Guinea. Perfluoroalkyl substances, particularly PFOS and long-chain PFCAs, were prevalent at varying concentrations in croakers and shrimps from the region. However, the values reported are below the recommended daily intake for PFOS and represent no obvious human health risk (HR < 1) to local communities and the seafood supply chain. The values are also below the environmental quality safety limits for seafood set

639 by the European Union. The various concentrations of PFASs reported in the three species
640 across the study sites indicate location-specific and habitat-specific contamination sources in
641 the region, particularly in the Niger Delta region and Lagos in Nigeria. Future studies should
642 focus on surface water, sediments and indicator species to improve our understanding of the
643 sources of contamination in the region, and elucidate the potential effects of PFAS
644 contamination on the environment and seafood from the region. Although the levels reported
645 in this study are lower than those reported in various studies worldwide, the increasing
646 urbanization and industrialization in the region coupled with the lack of adequate effluent
647 treatments should be a concern to stakeholders in the region. A more comprehensive assessment
648 is essential to understand the overall exposure of PFASs to seafood safety and public health.
649 There is a need to establish and implement guidelines for persistent contaminants, both legacy
650 and emerging ones, as governments in the Gulf of Guinea look towards the blue economy
651 potential of the region for its economic prosperity.
652

653 **Competing Interest**

654 The authors declare no competing interest
655

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Highlights and Graphical Abstract

Highlights

PFOS and long-chain PFCAs predominant PFASs in marine seafood from the Gulf of Guinea

The concentrations of PFASs in croakers were species- and location-dependent

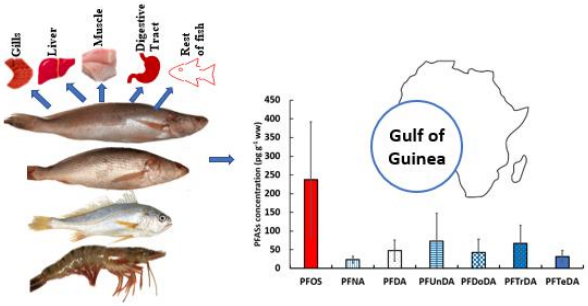
Significantly higher contamination levels in male croakers compared to females

Biomagnification from shrimps to croakers was evidenced for PFOS and long-chain PFCAs

Calculated BMFs decreased with increasing PFCA chain lengths

Based on present safety limits, PFOS levels in seafood show no risk to human health

Graphical Abstract



Competing Interest
The authors declare no competing interest