

Original Research Article

Levels, Spatial and Seasonal Distribution of Polybrominated Diphenyl Ethers (PBDEs) in Water, Sediments and *Sarotherodon melanotheron* from Ologe Lagoon

ABSTRACT

Polybrominated diphenyl ethers (PBDEs) are chemical additives of increasing concern in water bodies associated with industrial and urban areas. In this study, they were quantified in sediments, surface water and Black Chin Tilapia, *Sarotherodon melanotheron* from three sampling zones of Ologe lagoon, Nigeria. The lagoon receives effluents and runoffs from the neighbouring industrial town of Agbara. Quantification of PBDE congeners in the samples was done using a gas chromatograph coupled electron capture detector (GC-ECD). Sediment Σ PBDEs levels of 0.05 ± 0.006 to 0.09 ± 0.006 ng/g were below Environment Canada's (EC's) safe limit (6124 ng/g) while levels in water ranged from 0.01 ± 0.003 to 0.02 ± 0.005 mg/L and were above EC's safe limit (0.0002 mg/L). Σ PBDEs levels in the Tilapia ranged from 0.06 ± 0.01 to 0.12 ± 0.01 ng/g and were below EC's safe limit (631 ng/g). There was no seasonal variation ($p > 0.05$) of Σ PBDEs concentrations in the assessed fish. The low levels of PBDEs in *Sarotherodon melanotheron* suggest that the fish are safe for consumption. However, there is need for continuous efforts to improve industrial effluents and manage urban run-offs into waters in order to avoid potentially catastrophic levels of PBDEs in urban water bodies.

Keywords: Aquatic toxicology; Polybrominated diphenyl ether; Urban runoffs; Industrial effluents; Waste management.

1. INTRODUCTION

PBDEs are a group of chemicals that have been used as additive flame retardants in products such as building materials, plastics, and electronic equipment for decades [1,2]. They are hydrophobic and lipophilic [3], and could enter into aquatic environments through wet and dry deposition [4,5], surface runoff, and municipal and industrial wastes [5]. Despite the importance of PBDEs in consumer products, the characteristics of these chemicals make them persistent in aquatic environments [6]. They tend to stick to sediments which usually act as a sink for hydrophobic contaminants [7]. Their presence has been detected in water [8-10],

37 and aquatic organisms such as molluscs [11] and fish [12,13] which are able to
38 bioaccumulate them, and most importantly, traces of PBDEs have been found in humans [14-
39 16]. Their detection in humans has led to the study of their biological effects [17-21]. PBDEs
40 are endocrine disrupting chemicals that can interfere with the production, secretion, transport,
41 binding, and elimination of hormones that are responsible for development, behavior,
42 fertility, and the maintenance of homeostasis in the body [22-24]. There have also been
43 reports that PBDEs could be carcinogenic [25,3]. These effects make the investigation of the
44 occurrence of PBDEs in aquatic environments essential.

45 Ologe lagoon has been receiving a variety of wastes from its surroundings and more notably
46 from Agbara Industrial Estate. The ecological and economic services of the lagoon include
47 mining, transportation, waste disposal, and fishery [26]. It is vital to note that effluents from
48 Agbara Industrial Estate, which is close to the lagoon, are discharged into it after treatment in
49 a central effluent treatment plant. There are over 20 factories in the estate [27]. Currently,
50 there is no literature on the levels of PBDEs in the lagoon. It is vital to investigate the levels
51 of toxicants such as PBDEs that could end up in abiotic and biotic matrices of the lagoon.
52 The aim of this study was to assess the spatial and seasonal distribution of PBDEs in water,
53 sediments, and *Sarotherodon melanotheron* from Ologe Lagoon.

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56 **2. MATERIALS AND METHODS**

57 **2.1 Study Area**

58 The study area, Ologe lagoon, is in the south-western part of Nigeria and an extension of the
59 Badagry creek (Figure 1). The main body of the lagoon borders Badagry in Lagos State as
60 well as Agbara Township in Ogun State, Nigeria (Figure 1). Agbara is an industrial town
61 with industrial estates, and adjoining residential and business areas.

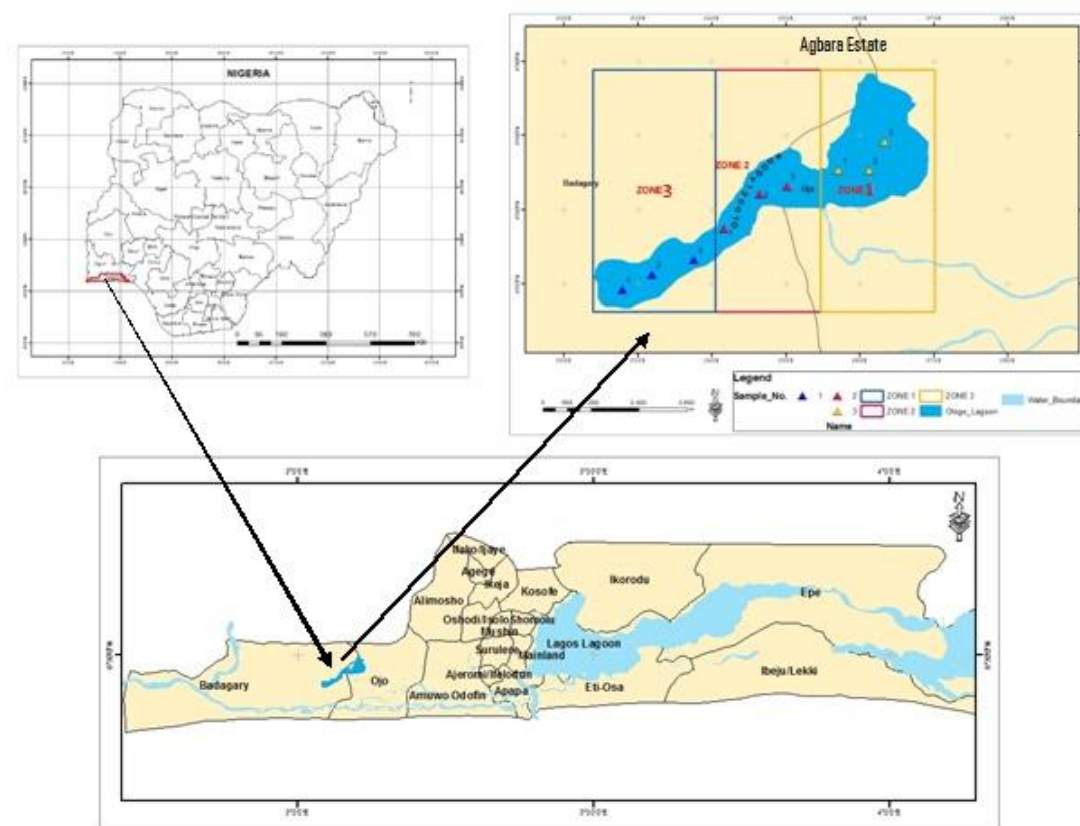


Figure 1: Map of Ologe Lagoon, Lagos, Nigeria, showing Sampling Zones

2.2 Sample Collection

Samples were collected in triplicates from three sampling zones in the rainy (May, 2017) and dry seasons (November, 2017).

Water samples were collected using a 2.5 L amber glass bottle at the water surface. Samples were filtered through 0.45µm fiberglass filters to remove sand and debris. Sediments were collected with a Van Veen Grab sampler. The sediments were wrapped with aluminum foil and appropriately labeled.

Fish were collected and wrapped in hexane-rinsed aluminium foil, labelled and placed inside a closed-glass vessel containing ice packs before they were transported in coolers to the laboratory for analysis within 24 hours. The fish species, *Sarotherodon melanotheron*, used for this study, is commonly exploited as a food resource in its native range and has been utilized as an aquaculture and non-native fishery species [28].

2.3 Extraction and Gas Chromatograph Analysis

The methods used were adapted from [29]. Specifically, 1L of water was extracted thrice with 300 ml mixture of hexane and acetone (3:1v/v). The extract was combined and dried with 20g of anhydrous Na_2SO_4 . After 24 hours, a rotary evaporator was used to concentrate the extract to 2ml.

Extraction of PBDEs from sediments and fish tissues was done by grinding 5grams of each sample and mixing with 5g of anhydrous Na_2SO_4 for homogenization. The homogenate was then extracted with 200 ml n-Hexane (solvent). The solution was shaken for 1 hour with an electronic shaker and the solution was stored overnight for separation and evaporation of the solvent. The extract was cleaned up using solvent-rinsed chromatographic columns (15 mm - 250 mm), packed with a plug of glass wool followed by 3 g deactivated silica gel and topped up with Na_2SO_4 . The column was pre-rinsed with 15 mL hexane after which 2 mL of the extract was added to the column and eluted with 60 mL hexane. The extract was then concentrated to approximately 2 mL using a rotary evaporator and kept in a sample vial for gas chromatographic analysis.

Quantification of PBDE congeners was done with a gas chromatograph coupled electron capture detector (GC-ECD) equipped with 30 m $\times$ 0.25 mm $\times$ 0.25 μm VF-5ms fused capillary column. The carrier gas was helium. The operating guidelines were as follows: column head pressure was 2 psi for 1.5 min with a constant flow rate of 150 mL min^{-1} at 4 min, the front injector line was programmed at 250°C. Injection volumes were 2.0 μL in the splitless mode. The column temperature was set at 50°C for 3 minutes initially and increased to 320°C at a rate of 20°C per minute and then maintained at 320°C for 20 minutes. Quantification of PBDE by the electron capture detector was based on sample peak areas or peak heights relative to standard peak areas or peak heights.

2.4 Biota-Sediment Accumulation Factor (BSAF)

The biota-sediment accumulation factor (BSAF) was calculated as a factor of the levels of PBDEs in the sediment and fish samples [30].

$$\text{BSAF} = \frac{\text{mean concentration of PBDEs in fish}}{\text{mean concentration of PBDEs in sediment}}$$

2.5 Bioaccumulation Factor (BAF)

The bioaccumulation Factor (BAF) was calculated as a ratio of PBDEs in the fish and surface water [31].

$$\text{BAF} = \frac{\text{mean concentration of PBDEs in fish}}{\text{mean concentration of PBDEs in water}}$$

2.6 Statistical Analysis

All readings were done in triplicates and results were expressed as Mean $\pm$ Standard Error. Significant differences in the obtained data were determined at $P < 0.05$ using One-way analysis of variance (ANOVA) and significant means were separated using Duncan (SPSS version 20 (IBM)).

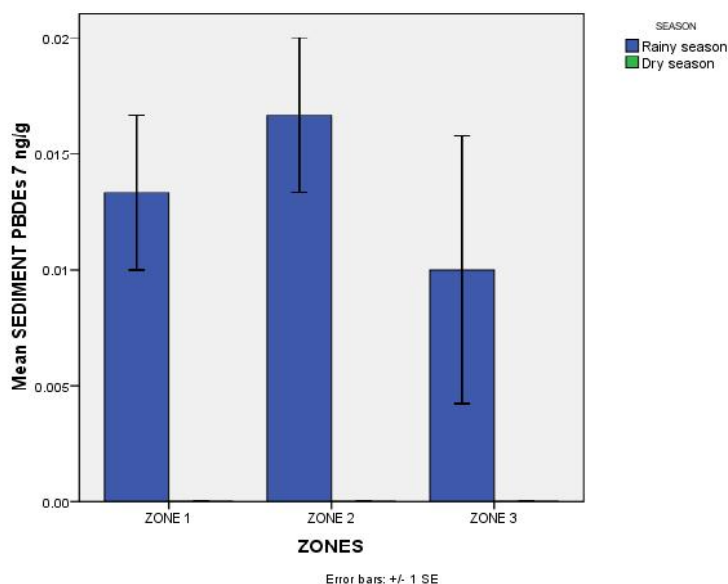
3. RESULTS AND DISCUSSION

3.1 Congeneric Patterns

Only two congeners (BDE 7 and 28) were detected in this study (Figures 2 to 7). Tri-BDE (BDE 28) was the predominant congener. TetraBDE (tetrabromodiphenyl ether), hexaBDE (hexabromodiphenyl ether), heptaBDE (heptabromodiphenyl ether), nonaBDE (nonabromodiphenyl ether) and decaBDE (decabromodiphenyl ether) were not detected. This could be as a result of photodecomposition or photolytic debromination of large BDEs into smaller molecules BDEs [32]. [32] assessed the photodegradation of BDE 47, BDE 99, BDE 100, BDE 153, and BDE 183 dissolved in hexane. The researchers observed that higher brominated BDE congeners degraded at a faster rate than the lower brominated congeners. It was concluded that the main decomposition mechanism induced by photolysis was reductive

127 debromination to form lower brominated BDE congeners. [33] also investigated the
128 photodegradation of BDE 209 in plastics and obtained similar results.

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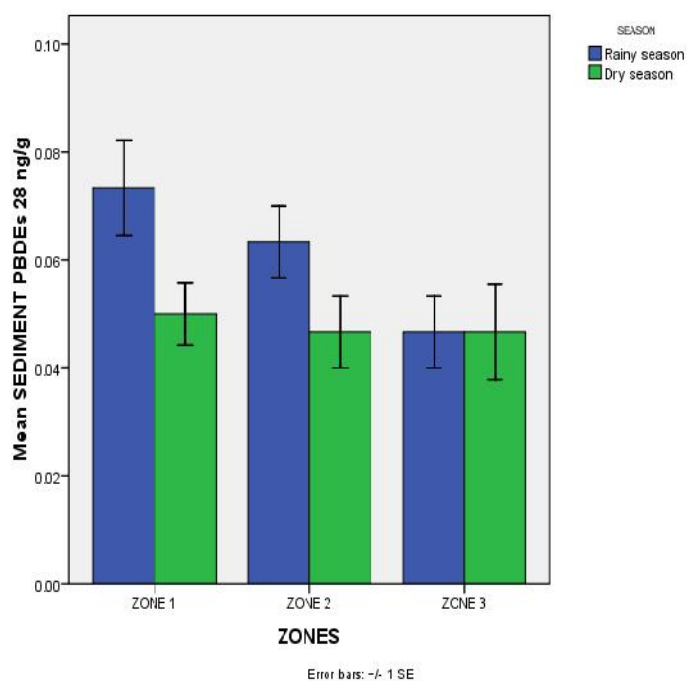


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Figure 2: Concentrations of BDE 7 in Sediments samples of the Ologe lagoon

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Figure 3: Concentrations of BDE 28 in Sediments samples of the Ologe lagoon

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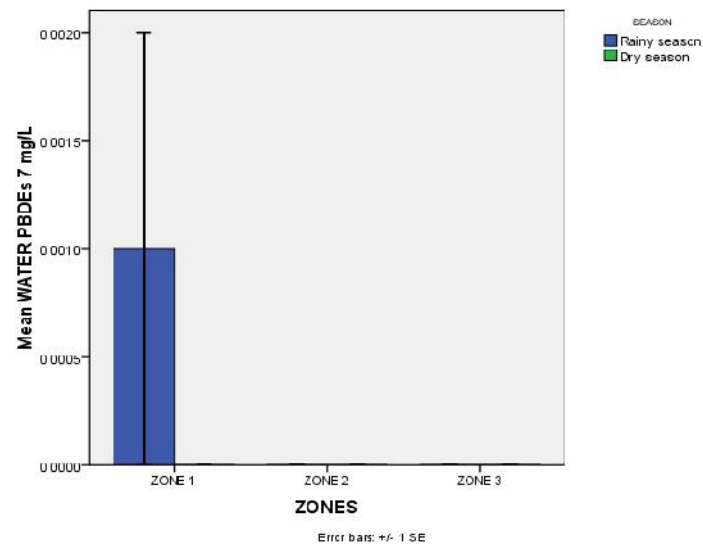


Figure 4: Concentrations of BDE 7 in the surface water samples of the Ologe lagoon

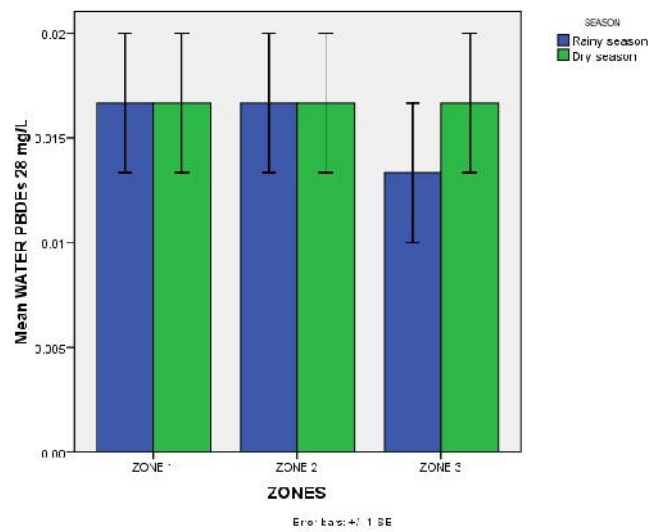


Figure 5: Concentrations of BDE 28 in the surface water samples of the Ologe lagoon

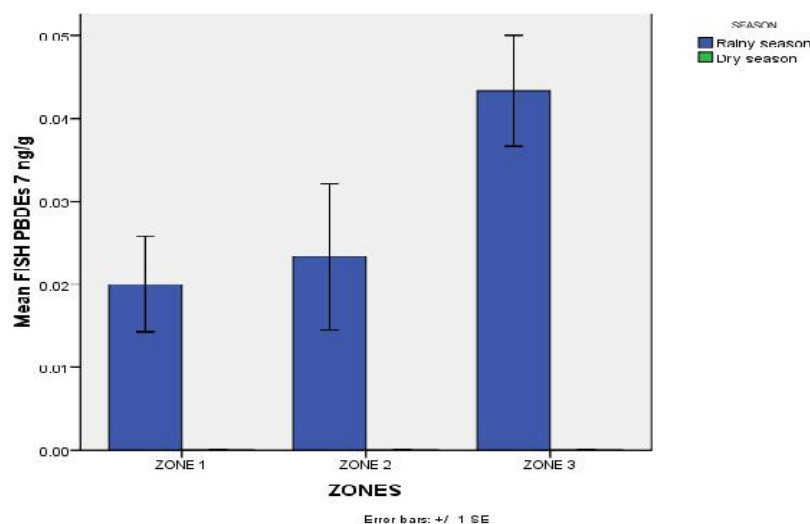


Figure 6: Concentrations of BDE 7 in Fish samples of the Ologe lagoon

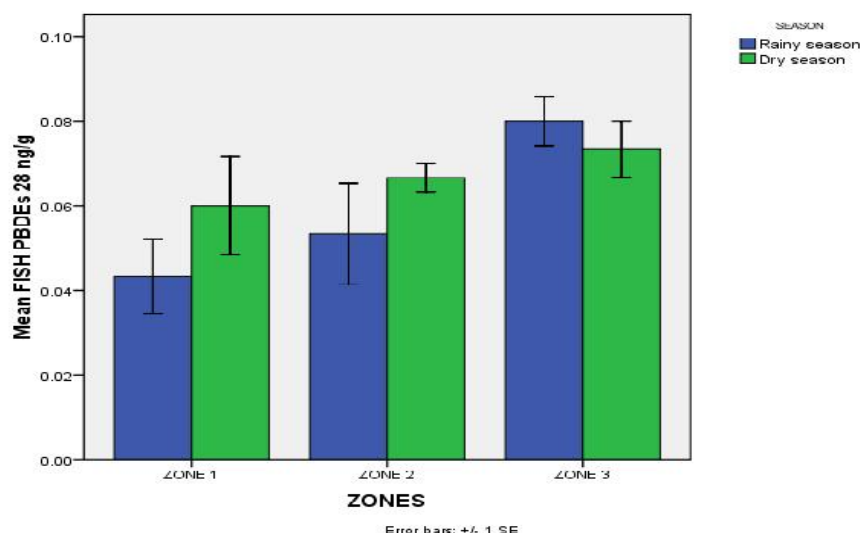


Figure 7: Concentrations of BDE 28 in Fish samples of the Ologe lagoon

3.2 Seasonal and Spatial Distribution of PBDEs in Sediments

There was a significant ($P < 0.05$) spatial difference in the PBDE levels in sediments in the rainy season, however, the concentrations did not significantly ($P > 0.05$) differ in the dry season (Figure 8). PBDEs levels of 0.05 ± 0.006 to 0.09 ± 0.006 ng/g in sediments recorded in this study were within the range reported by [34] in China and [35] in Japan and were below Environment Canada (EC) safe limit (6124 ng/g). Σ PBDEs levels in sediments from Zones 1

and 2 were significantly ($p < 0.05$) higher in the rainy season. The variation might have been caused by the seasonal differential influx of industrial wastes into the lagoon from nearby industries (Agbara industrial estate precisely).

In the dry season, there is less surface runoff which might cause contaminants to accumulate in the surface soil while in the rainy season, the contaminants previously accumulated in the surface soil are flushed into the lagoon through surface runoff due to the heavy rains [36-38].

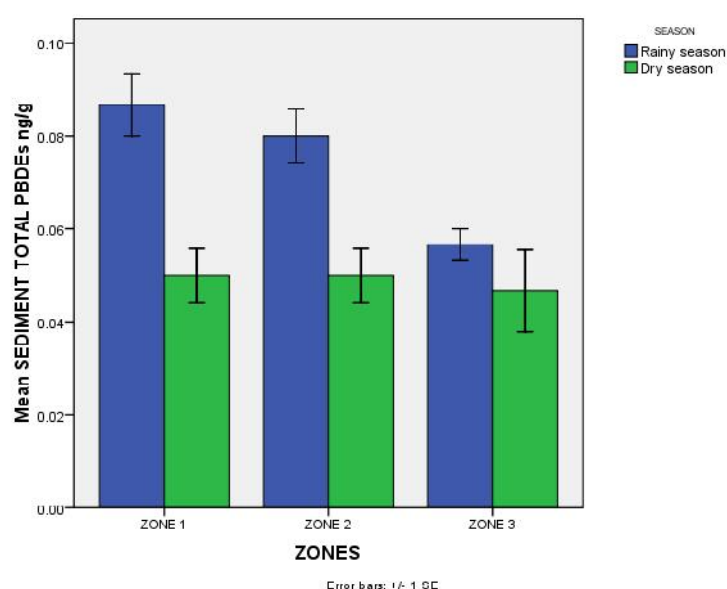


Figure 8: Concentrations of Σ PBDEs in Sediments samples of the Ologe lagoon

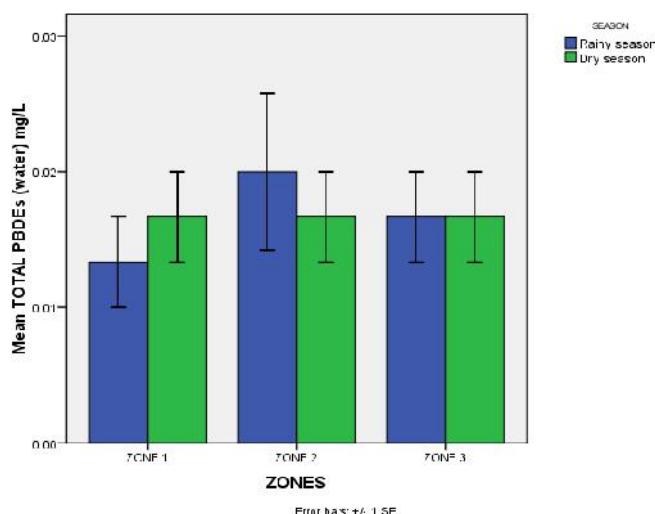
3.3 The Occurrence of PBDEs in Water

The Σ PBDEs concentrations in surface water samples from the lagoon ranged from 0.01 ± 0.003 to 0.02 ± 0.005 mg/L (Figure 9). There was no significant spatial and seasonal variation ($P > 0.05$) in the Σ PBDEs concentrations. [39] assessed the effects of seasonal change on PBDEs concentrations in water but did not report significant seasonal variations as well. The Σ PBDEs levels in water in this study and in Ile Ife [10], another town in Nigeria with much less industries were found to be above ECs' safe limit (0.0002mg/L). This raises

180 concern on the diffuse nature of this pollutant and the potential for environmental and public
181 health risks.

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Figure 9: Concentrations of Σ PBDEs in the surface water samples of the Ologe lagoon

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188 3.4 Occurrence and Bioaccumulation of PBDEs in *Sarotherodon melanotheron*

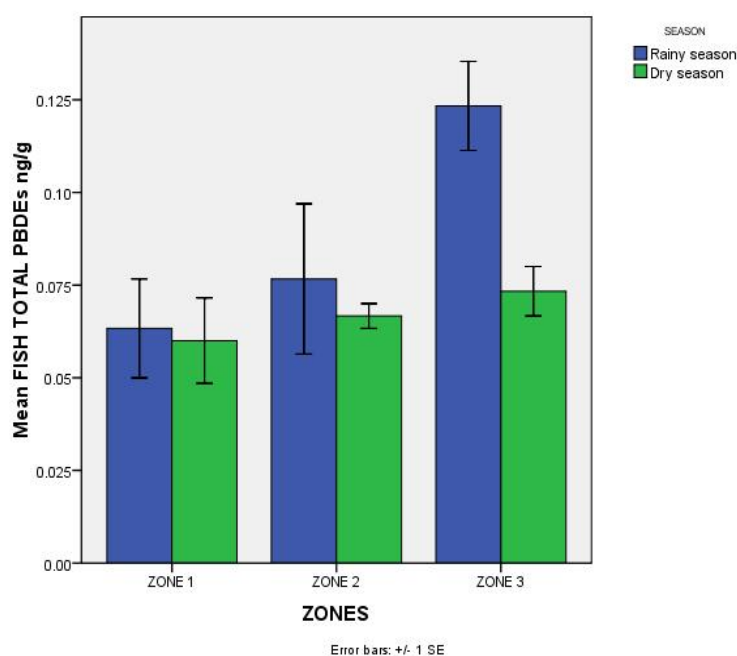
189 Σ PBDEs concentrations ranged from 0.06 ± 0.01 to 0.12 ± 0.01 ng/g in the assessed tilapia fish
190 (Figure 10) . There was a significant ($P < 0.05$) difference in PBDE concentrations across the
191 sampling zones in the rainy season conversely, the concentrations did not significantly ($P >$
192 0.05) differ in the dry season. The spatial difference could be associated with the difference
193 in the proximity of the zones to the source(s) of effluents. The PBDEs values in *S.*
194 *melanotheron* were below EC's safe limit of 631ng/g. There was no statistical seasonal
195 variation ($p > 0.05$) in Σ PBDEs levels in fish.

196 The levels of PBDEs in the assessed tilapia were low when compared to data from other
197 countries (Table 1). The low levels of PBDEs in fish found in the present study suggest that
198 the levels of PBDEs bioavailable for aquatic organisms in Ologe lagoon were low. Nigeria is
199 much less industrialized than most of the countries listed in the table while for the case of

200 Ghana the differences might lie on the specific sources of effluents and runoffs entering the
201 water bodies examined.

202 In the rainy season, Tilapia from zone 3 had the highest BSAF and BAF values which
203 suggest that accumulation of PBDEs in the fish from zone 3 was highest (Table 2). In the dry
204 season, BSAF and BAF values were the same except BSAF of PBDEs in fish from zone 1
205 (Table 2). [40] noted that the mobility and reproductive behavior of an aquatic organism
206 could influence the extent of bioaccumulation of contaminants.

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Figure 10: Concentrations of Σ PBDEs in Fish samples of the Ologe lagoon

Table 1: PBDEs Levels in Fish from other countries

Country	Levels	References
U.S.A.	1140 ng/g ww	[41]
Sweden	36,900 ng/g lw	[42]
Norway	72–1120 ng/gww	[12]
Ghana	7.3 ng/g lw	[13]
China	18 to 1100 ng/g lw	[43]
Nigeria (Ologe lagoon)	0.06±0.01 to 0.12±0.01 ng/g lw	

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217 **Table 2: Biota-Sediment Accumulation Factor (BSAF) and Bioaccumulation Factor**
 218 **(BAF)**

PBDEs concentrations	Sampling Zones					
	Rainy Season			Dry Season		
	1	2	3	1	2	3
BSAF	0.67	1.00	1.33	1.2	1.4	1.4
BAF	0.005	0.00	0.00	0.00	0.00	0.00
		4	7	4	4	4

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220 4. CONCLUSION

221 The levels of PCBs in this study suggest that the release of the PBDEs into the lagoon occurs
 222 at a low rate considering the vast array of anthropogenic activities that is carried out in the
 223 area. This can be attributed to the fact that industries around the area are more concerned with
 224 production of pharmaceuticals and beverages rather than electronics and equipment. Despite
 225 this public suspicion, it is noteworthy to state that *S. melanothron* inhabiting the water body
 226 are considered safe for human consumption because the PBDEs levels were below EC safety
 227 limits for fish food. On the contrary, the Σ PBDEs levels in the water were above EC safe
 228 limit confirming the surface runoffs as possible route of entry into the water bodies. The
 229 need for continued environmental monitoring of PBDE levels is advised in view of their
 230 potential to act as potent carcinogens.

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