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

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Authors' contributions

This work was carried out in collaboration between all authors. All authors read and approved the final manuscript.

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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 ∑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). ∑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 significant ($P > 0.05$) seasonal variation of ∑PBDEs concentrations in the assessed fish. The low levels of PBDEs in *Sarotherodon melanotheron*

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suggest that the fish are safe for consumption. However, there is a need for continuous efforts to improve industrial effluents treatment and manage urban surface run-offs into water bodies in order to avoid potentially catastrophic levels of PBDEs in urban water bodies.

Keywords: Aquatic toxicology; polybrominated diphenyl ethers; urban runoffs; industrial effluents; waste management.

1. INTRODUCTION

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

Ologe Lagoon has been receiving a variety of wastes from its surroundings and more notably from Agbara Industrial Estate. The ecological and economic services of the lagoon include mining, transportation, waste disposal, and fishery [26]. It is vital to note that effluents from Agbara Industrial Estate, which is close to the lagoon, are discharged into it after treatment in a central effluent treatment plant. There are over 20 factories (food and beverages, pharmaceutical, breweries, metal finishing, chemical, pulp and paper industries) in the estate [27]. Currently, there is no literature on the levels of PBDEs in the lagoon. It is therefore vital to investigate the levels of toxicants such as PBDEs that could end up in abiotic and biotic matrices of the lagoon. The aim of this study was to assess the spatial and seasonal distribution of PBDEs in water and sediments from Ologe Lagoon using *Sarotherodon melanotheron* bioindicator.

2. MATERIALS AND METHODS

2.1 Study Area

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

2.2 Sample Collection

Samples were collected from Zones 1 (3º05' N and 6º28'E), 2 (3º04' N and 6º27'E), and 3 (3º03' N and 6º26'E) of Ologe Lagoon in the rainy (May, 2017) and dry seasons (November, 2017). Each Sampling Zone was divided into 3 sampling stations at 930 m intervals.

Water samples were collected using a 2.5 L amber glass bottle at the water surface. Samples were filtered through 0.45 µm 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.

Fishes 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 utilised as an aquaculture and non-native fishery species [28].

2.3 Extraction and Gas Chromatograph Analysis

The methods used were adapted from [29]. Specifically, 1 L of water was extracted thrice with 300 mL mixture of hexane and acetone

(3:1v/v). The extract was combined and dried with 20 g of anhydrous Na_2SO_4. After 24 hours, a rotary evaporator was used to concentrate the extract to 2 mL.

Extraction of PBDEs from sediments and fish tissues was done by grinding 5 g of each sample and mixing with 5 g of anhydrous Na_2SO_4 for homogenisation. 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×0.25 mm×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 PBDEs by the electron capture detector was based on sample peak areas or peak heights relative to standard peak areas or peak heights.

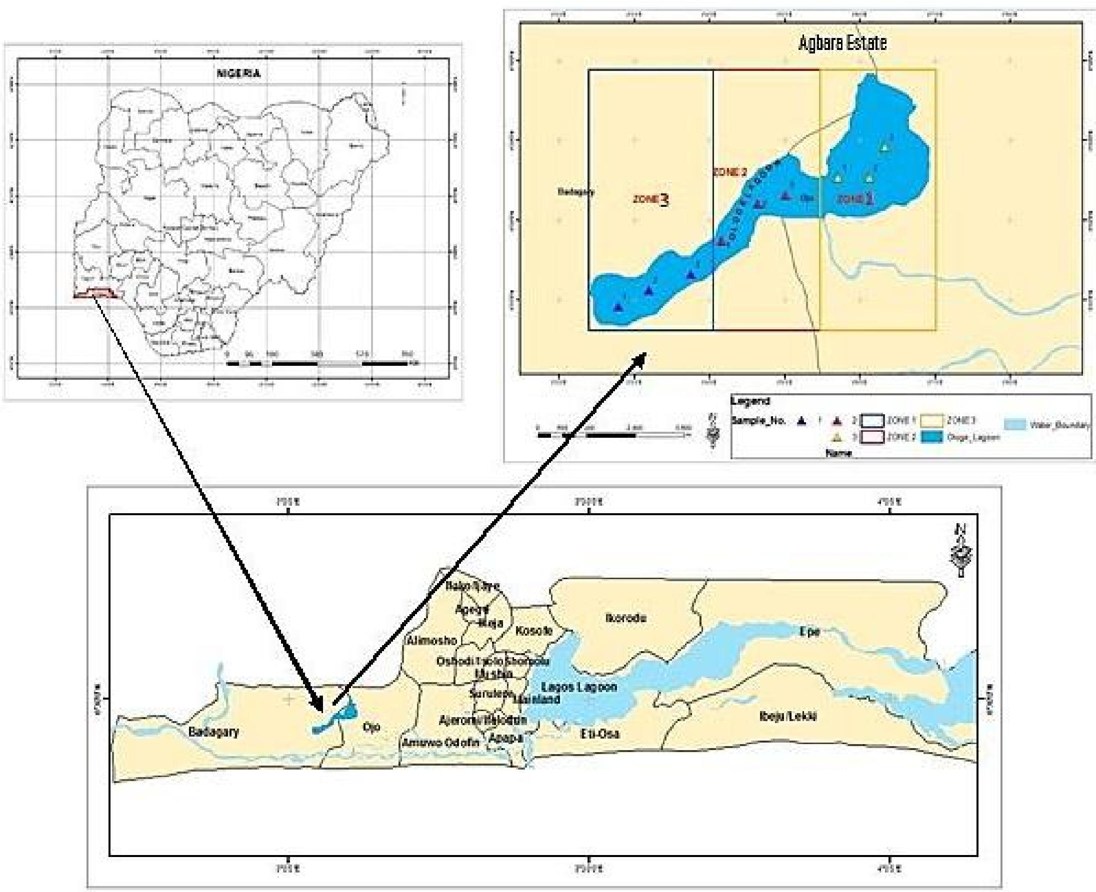

Fig. 1. Map of Ologe Lagoon, Lagos, Nigeria, showing the sampling zones

2.4 Biota-Sediment Accumulation Factor (BSAF)

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

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

2.5 Bioaccumulation Factor (BAF)

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

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

2.6 Statistical Analysis

All results were expressed as Mean ± Standard Error. Two-way analysis of variance (ANOVA) with Duncan's Multiple Range (DMR) test was used to determine the significance of spatial and seasonal differences in the levels of PBDEs in water, sediments, and fish using SPSS version 20 (IBM).

3. RESULTS AND DISCUSSION

3.1 Congeneric Patterns

PBDEs comprise 209 related molecules, known as congeners [32]. The lower brominated PBDEs are known to exhibit higher toxicity than higher brominated PBDEs [33,34]. Only two congeners (BDE 7 and 28) were detected in this study (Figs. 2 to 7). Tri-BDE (BDE 28) was the predominant congener. Higher brominated PBDEs were not detected in the present study. This could be as a result of photodecomposition or photolytic debromination of higher brominated PBDEs into smaller molecules BDEs [35]. Fang L et al. [35] assessed the photodegradation of BDE 47, BDE 99, BDE 100, BDE 153, and BDE 183. [35] 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 debromination to form lower brominated BDE congeners. Ramu K et al. [36] investigated the photodegradation of BDE 209 in plastics and obtained similar results.

3.2 Seasonal and Spatial Distribution of PBDEs in Sediments

There was a significant (*$P < 0.05$, $F= 4.00$*) spatial difference in ∑PBDEs levels in sediments. This could be attributed to the varying distances of the zones to the source of effluents. ∑PBDEs levels of 0.05±0.006 to 0.09±0.006 ng/g (Fig. 8) in sediments recorded in this study were within the range reported by [37] in China and [38] in Japan and were below Environment Canada's (EC's) safe limit (6124 ng/g). ∑PBDEs levels in sediments were significantly (*$P < 0.05$, $F=25.190$*) higher in the rainy season than the levels recorded in the dry 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 was 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 being flushed into the lagoon through surface runoff due to the heavy rains [39-41].

3.3 The Occurrence of PBDEs in Water

The ∑PBDEs concentrations in surface water samples from the lagoon ranged from 0.01±0.003 to 0.02±0.005 mg/L (Fig. 9). There was no significant spatial (*$P > 0.05$*, *$F = 0.375$*) and seasonal (*$P > 0.05$*, *$F = 0$*) variation in the ∑PBDEs concentrations. Harrad S et al. [42] assessed the effects of seasonal change on PBDEs concentrations in water but did not report significant seasonal variations as well. The ∑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 EC's safe limit (0.0002 mg/L). This raises concern on the diffuse nature of this pollutant and the potential for environmental and public health risks.

3.4 Occurrence and Bioaccumulation of PBDEs in *Sarotherodon melanotheron*

∑PBDEs concentrations ranged from 0.06±0.01 to 0.12±0.01 ng/g in the assessed tilapia fish (Fig. 10). There was a significant (*$P < 0.05$, $F=4.675$*) difference in PBDE concentrations across the sampling zones. The spatial difference could be associated with the difference in the proximity of the zones to the source(s) of effluents. The ∑PBDEs values in *S. melanotheron* were below EC's safe limit of 631 ng/g. There was no statistical seasonal variation (*$P > 0.05$, $F = 4.349$*) in ∑PBDEs levels in fish.

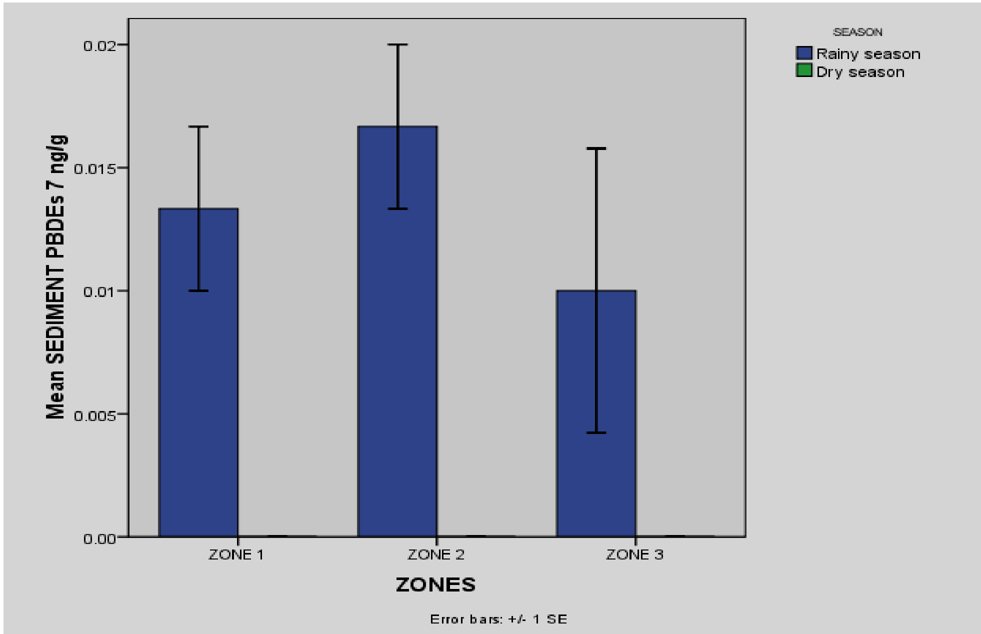

Fig. 2. Concentrations of BDE 7 in Sediments samples of Ologe lagoon

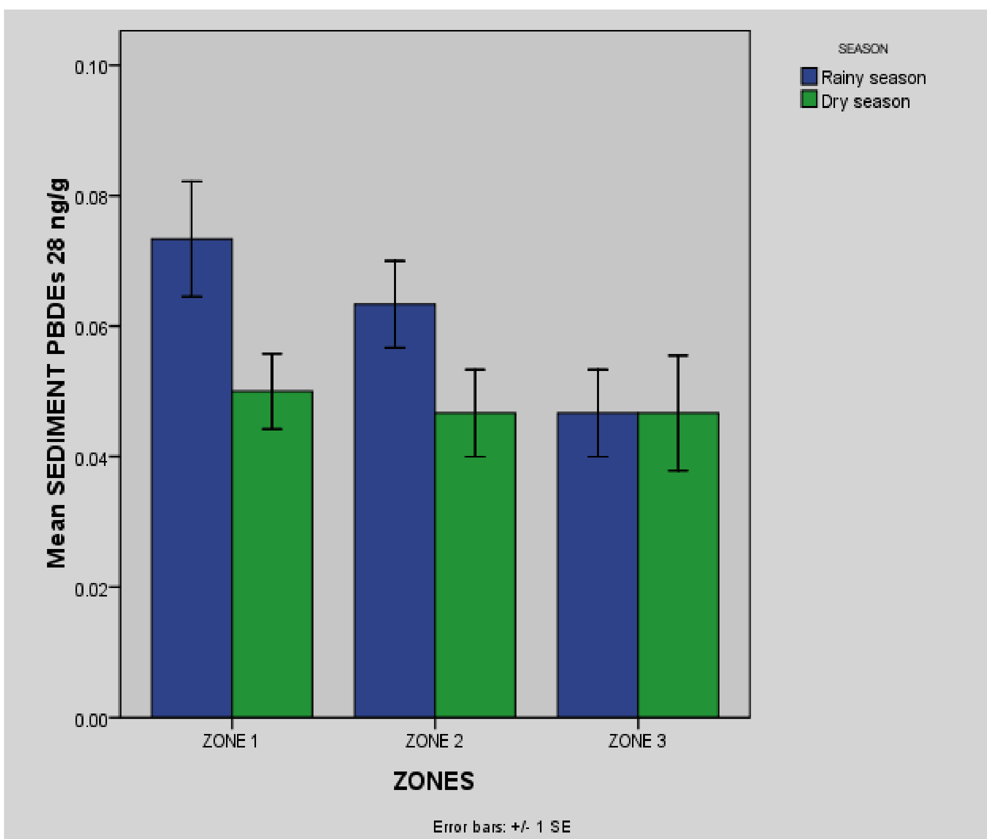

Fig. 3. Concentrations of BDE 28 in Sediments samples of Ologe lagoon

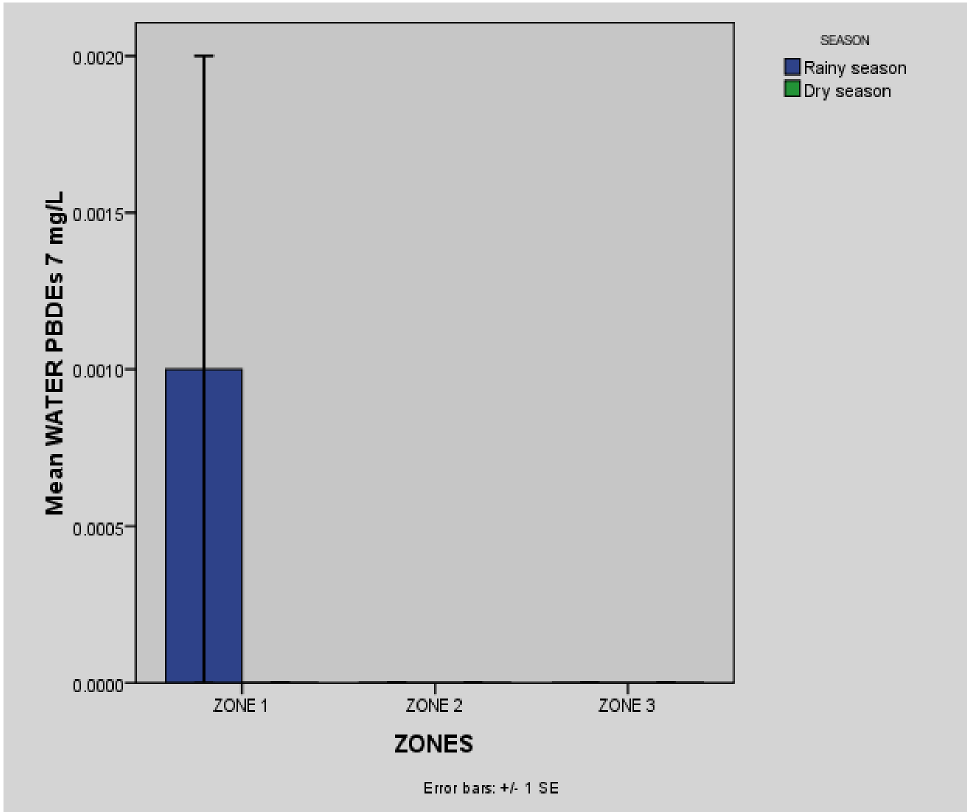

Fig. 4. Concentrations of BDE 7 in the surface water samples of Ologe lagoon

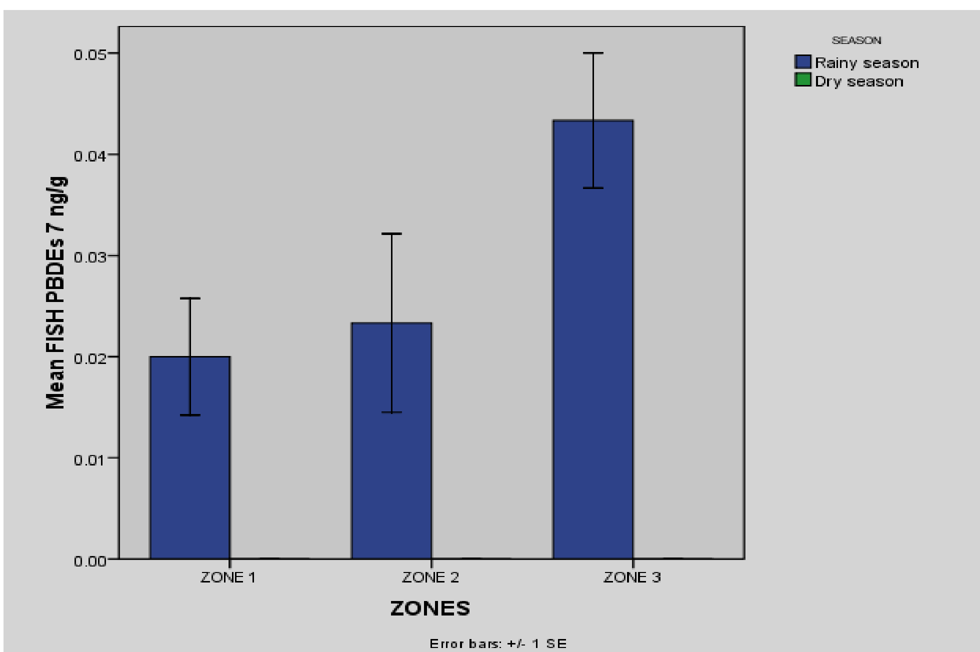

Fig. 5. Concentrations of BDE 7 in Fish samples of Ologe lagoon

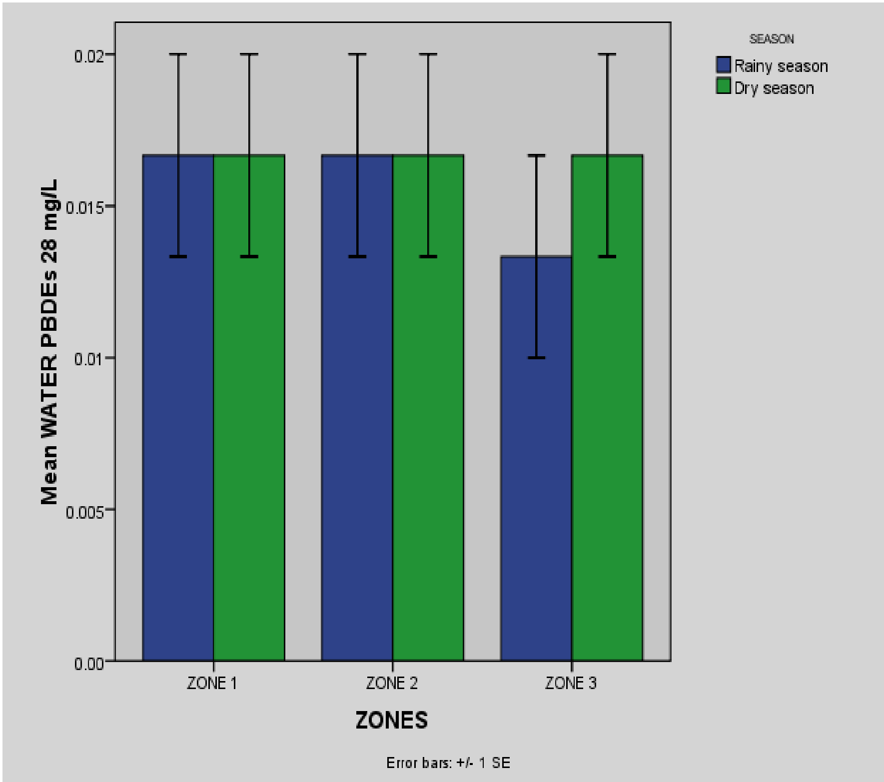

Fig. 6. Concentrations of BDE 28 in the surface water samples of Ologe lagoon

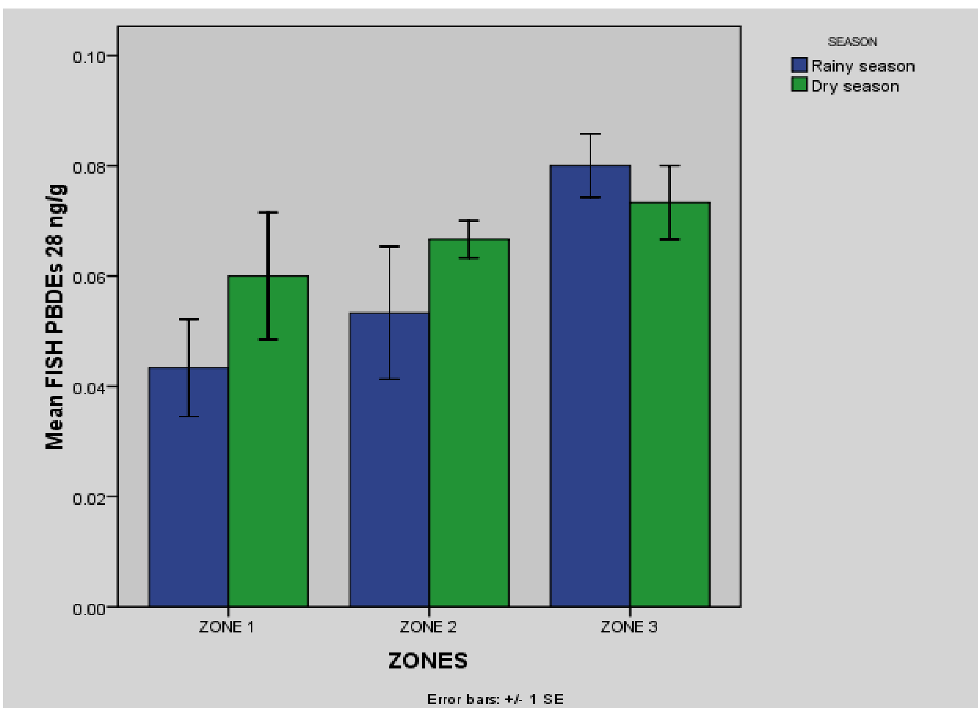

Fig. 7. Concentrations of BDE 28 in Fish samples of Ologe lagoon

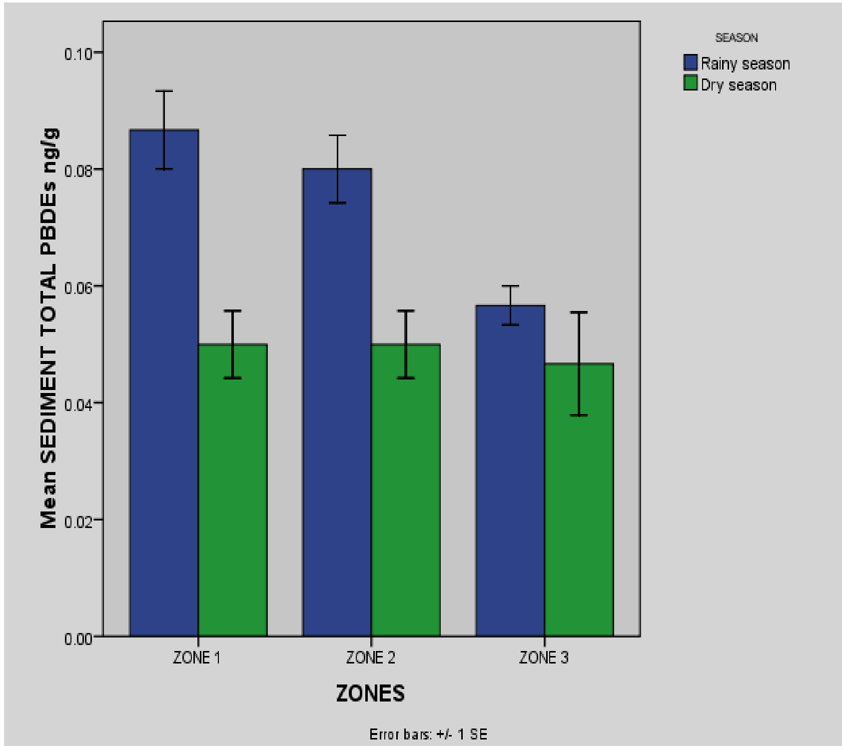

Fig. 8. Concentrations of ∑PBDEs in Sediments samples of Ologe lagoon

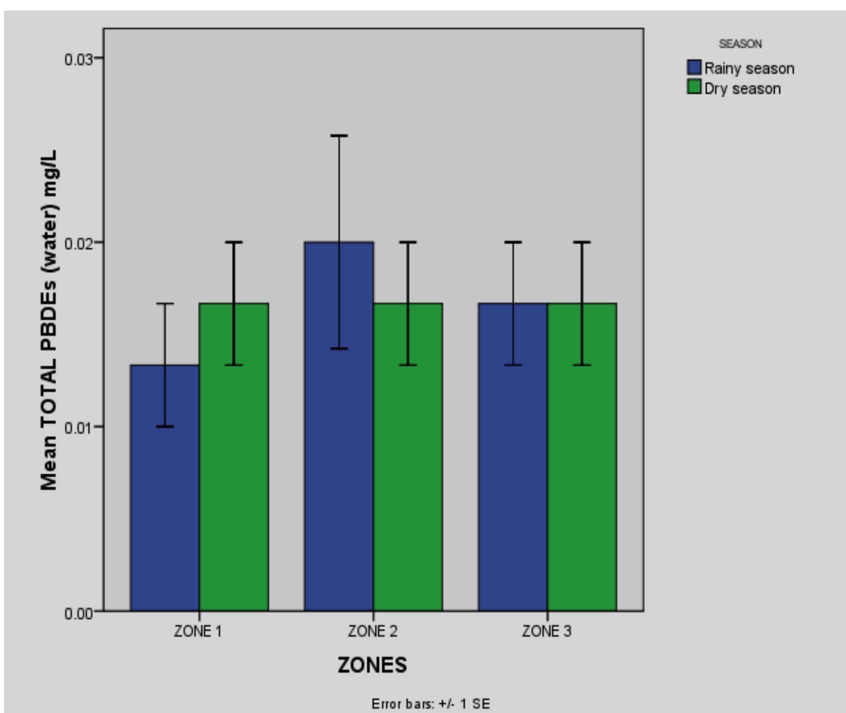

Fig. 9. Concentrations of ∑PBDEs in the surface water samples of Ologe lagoon

The levels of ∑PBDEs in the assessed tilapia were low when compared to data from other countries (Table 1). The low levels of PBDEs in fish found in the present study suggest that the levels of PBDEs bioavailable for aquatic organisms in Ologe lagoon were low. Nigeria is much less industrialised than most of the countries listed in Table 1 while for the case of Ghana the differences might lie on the specific sources of effluents and runoffs entering the water bodies examined.

In the rainy season, Tilapia from zone 3 had the highest BSAF and BAF values which suggest that accumulation of PBDEs in the fish from zone 3 was highest (Table 2). In the dry season, BSAF and BAF values were the same except BSAF of PBDEs in fish from zone 1 (Table 2). Howell NL et al. [43] noted that the mobility and reproductive behavior of an aquatic organism could influence the extent of bioaccumulation of contaminants.

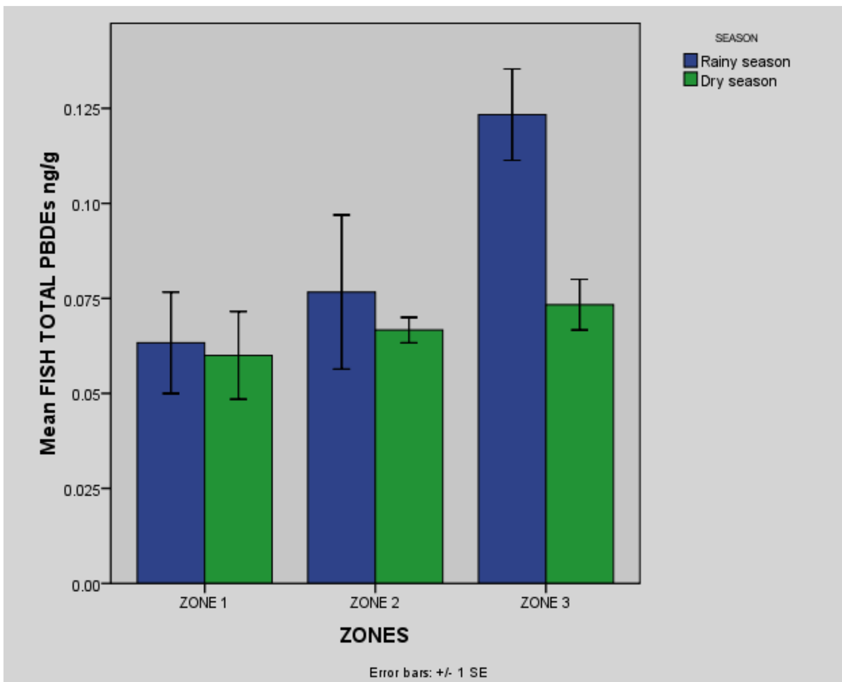

Fig. 10. Concentrations of ∑PBDEs in Fish samples of Ologe lagoon

Table 1. PBDEs Levels in Fish from other countries

Country	Levels	References
U.S.A.	1140 ng/g ww	[44]
Sweden	36,900 ng/g lw	[45]
Norway	72–1120 ng/gww	[12]
Ghana	7.3 ng/g lw	[13]
China	18 to 1100 ng/g lw	[46]
Nigeria (Ologe lagoon)	0.06±0.01 to 0.12±0.01 ng/g	Present study

Table 2. Biota-Sediment Accumulation Factor (BSAF) and Bioaccumulation Factor (BAF)

	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	0.004	0.007	0.004	0.004	0.004

4. CONCLUSION

The levels of PBDEs in this study suggest that the release of PBDEs into the lagoon occurs at a low rate considering the vast array of anthropogenic activities that is carried out in the area. This may be attributed to the fact that industries around the area are more concerned with the production of pharmaceuticals and beverages rather than electronics and equipment. The present research findings indicate that *S. melanotheron* inhabiting the water body are safe for human consumption with regard to PBDEs level. On the contrary, the ∑PBDEs levels in the water were above EC's safe limit confirming the surface runoffs as possible route of entry into the water bodies. The need for continued environmental monitoring of PBDEs levels is advised in view of their potential to act as potent carcinogens.

COMPETING INTERESTS

Authors have declared that no competing interests exist.

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