Original Research Article

MICROPELLET PARTICLES: A VECTOR OF HYDROPHOBIC ENDOCRINE DISRUPTING CHEMICALS IN LAGOS LAGOON

ABSTRACT

The occurrence of plastic waste materials in the aquatic environment is receiving enormous attention all over the world due to its negative impacts on aquatic organisms. Micropellet litters have been found to adsorb and absorbs persistent organic endocrine disrupting chemicals (EDCs). Endocrine disrupting chemicals are recognized toxic chemicals to humans and organisms. This study quantifies occurrence of micropellet particles in Lagos Lagoon and their EDCs contents. Surface water and sediment sample collection was conducted from 2016 to 2017 at eight sampling locations with three points established in each of the sampling station. The chemical analysis of EDCs was conducted by gas chromatography coupled with electron capture detector and flame ionization detector. Micropellet particles occurrence was highest in surface water (67%) compared to (33%) in sediment during the period of sampling. EDCs contents reflect contamination of PCBs and PAHs in the extracted micropellet particles. Some sampling stations contained relatively higher PAHs concentrations but very low concentration of PCBs. Since, micropellet particles and EDCs cannot be removed completely from the aquatic environment, reduction of impending hazards ought to rely on curtailing disposal of plastic materials and sensitizing the populace on general disposal methods in order to minimize interaction of plastic particles with EDCs which are likely to pose significant effects on aquatic fauna.

Keywords: Micropellet particles, endocrine disrupting chemicals (EDCs), PCBs; PAHs, Lagos Lagoon

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I. 0 INTRODUCTION

Lagos lagoon in Nigeria supplies a number of essential services for indigenous and non-indigenous people in Lagos (Ajagbe *et al.,* 2012). High influence of solid waste litters in and around the Lagos lagoon has been reported by several authors (Ajao, 1996; Amaeze *et al.*, 2012; Abiodun and Oyeleke, 2016, Soneye *et al.,* 2018). In Nigeria, plastic litter materials in coastal areas as been reported by (Amaeze *et al.*, 2012; Abiodun and Oyeleke, 2016, Soneye *et al.,* 2018) as one of the most common litters in Lagos lagoon and are likely to be a major sources of hydrophobic endocrine disrupting chemicals contributing to the significant decline in aquatic resources thus threatening their long-term sustainability. Plastic wastes materials is one of the most documented waste in aquatic ecosystem globally (Derraik, 2002;UNEP, 2016) with greater negative impact on aquatic fauna (Gibb *et al.*, 2O17). In the last few decades, plastic products have developed into one of the most largely used materials for many applications. World total plastic production continues to increase yearly (Duis and Coors, 2016) owing to the plasticity and the durability of most of the plastic products (PlasticEurope, 2015, 2017). Due to the extensive utilization of plastic in many applications coupled with improper disposal methods of plastic waste materials consistently in waterways leading to fragmentation into microparticles due to weathering action and ultra violet solar radiation (Ryan *et al.*, 2009; Andrady, 2011).

Plastic wastes materials was proposed as hazardous materials (Rochman *et al.*, 2013) when found in the aquatic environment (UNEP, 2016). Due to increase in population density and economic growth rate there is a clamour for affordable products which have led to increased plastics production as well as indiscriminate increase in plastic waste generation in Nigeria (Olanrewaju and Ilemobade, 2009). Despite the intervention of the government in Nigeria, on proper waste disposal methods, solid waste still find their way into the Lagos lagoon at an

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alarming rate (Olanrewaju and Ilemobade, 2009; Babayemi, *et al.,* 2018).Unlike other substances, majority of plastic waste materials are not easily biodegradable, but instead photodegradable into smaller fragment (Arthur *et al.*, 2009) from macroplastic, >5 mm into microplastics<5mm that has increased conspicuously (Thompson *et al.*, 2004, 2009) in the aquatic environment. Several authors reported the ability of microplastics particles to adsorbed and absorbed hydrophobic endocrine disrupting chemicals (HEDCs) at a several magnitude higher than their surrounding water (Rochman *et al.*, 2013; Velzeboer *et al.,* 2014).

Plastic waste materials and hydrophobic endocrine disruption chemicals has been reported by Vethaak and Leslie (2016) to form multifaceted mixture of contaminants in the aquatic environment that increase the availability HEDCs to be readily bioavailability to wide variety of aquatic faunas and eventually to humans in contrast to other naturally sorbent (Bakir *et al.,* 2016; Koelmans *et al.*, 2016). Hydrophobic endocrine disrupting chemicals like polychlorinated biphenyls (Mato *et al., 2*001, Bakir *et al.,* 2012, 2014) and polycyclic aromatic hydrocarbons (Rochman *et al.*, 2013) are known to adsorb on microplastic surfaces (Teuten *et al.*, 2009). Presently, there is increasing concern that aquatic fauna declines in population and increasing occurrence of endocrine-related syndrome in aquatic organisms are connected to chemicals compound adsorbed on plastic waste materials (Bergman *et al.,* 2013). These chemicals compounds include but not limited to polychlorinated biphenyl (PCBs), organochlorine pesticides (OCPs) and polycyclic aromatic hydrocarbon (PAHs). Most of these pollutants are well known hydrophobic persistent organic pollutants that are constrained in most countries Nigeria inclusive may interfering with the endocrine system as reported by (Bergman *et al.,* 2013). Some HEDCs are known to cause effects at the present levels found in biota and the environment (Vethaak and Legler, 2012; Bergman *et al.,* 2013). However, the absorption ability

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of micropellet particles of hydrophobic endocrine disrupting chemicals has not been sufficiently studied. Hence, this study is aimed at evaluating the associated hydrophobic endocrine disrupting chemicals in micropellet particles extracted from surface water and sediment of Lagos lagoon.

2.0 MATERIALS AND METHODS

2.1. Description of sampling area

The study was carried out in one of the biggest estuary in Nigeria Lagos lagoon is located between longitude 3°23" and 3°53" and latitude 6°26" and 6°37"N. The lagoon empties into the Atlantic Ocean through the Lagos harbour, an important channel through the heart of Lagos. Within the Lagos lagoon eight (8) sampling stations were established based on solid waste characteristics of each of the sampling area as reported by past literature (Abiodun and Oyeleke, 2016) (Figure 1 and Table 1). In each of the sampling stations three (3) points were selected to represent the true conditions of the sampling locations (Figure 1) with different wastes littering the surface of the water.

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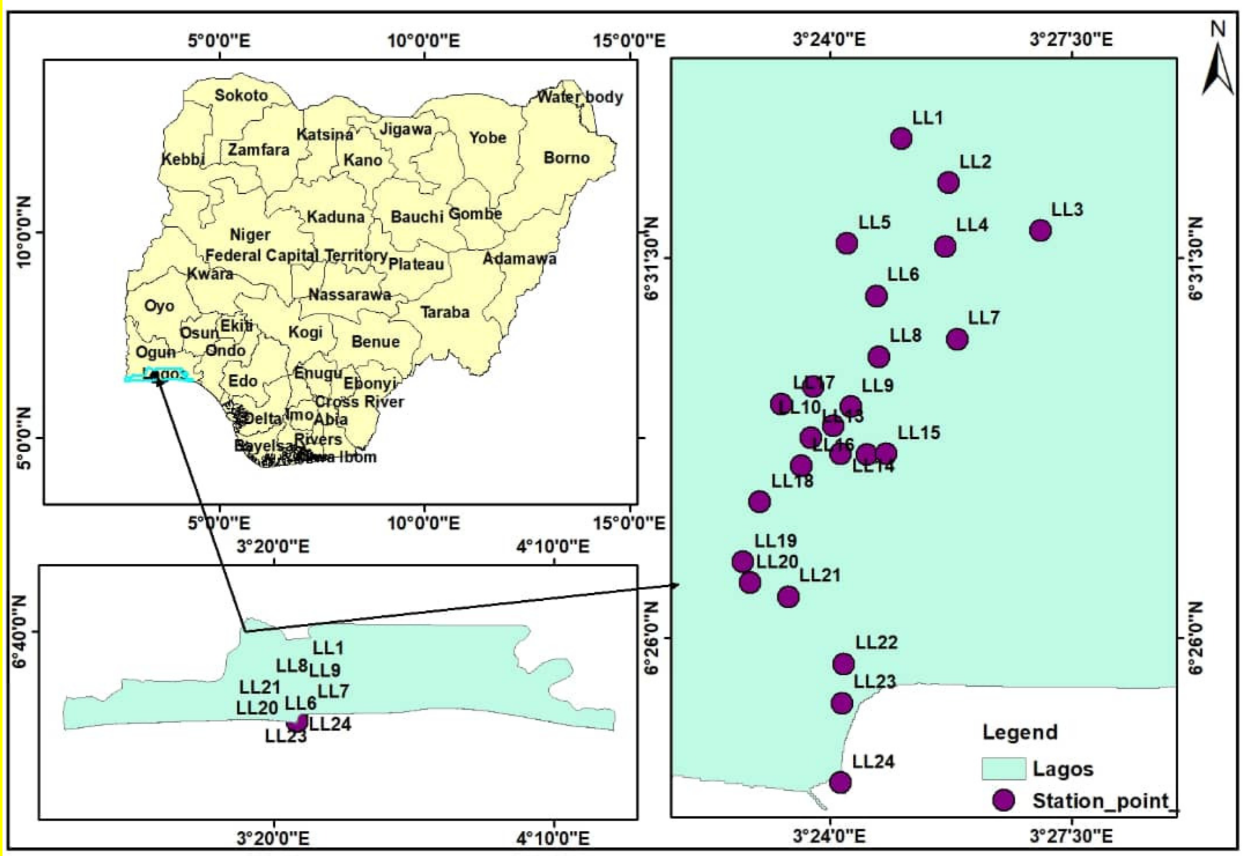

Figure1: Map of the Sampled Study Area

2.2 Sample Collection Methods

- Microplastic Samples

The surface water was collected by means of manta trawl net with a circular opening of 15cm by 45cm wide with iron frame, 60 cm length 1.62mm mesh net with 20X 5 collecting bottle was towed behind a speed boat for 30 minutes at each site to sampled microplastic by tow speeds below 3 knots, while speed boat maintains a consistent heading. At each sample site, a target tow length of 500 to 2000 m was established with length based upon the amount of floating debris and waste samples captured at the base of the net end was placed in a clean pre labelled glass sample bottle. To avoid contaminating samples, the manta net and collection vessel were rinsed

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methodically (Eriksen *et al.*, 2013). Sediment sampled for the analysis of micropellet particles were collected with a Van veen grab sampler ($0.1m^2$) in areas of low flow velocity (<0.3m/s) in each of the stations. Thereafter, sediment was gently stirred and carefully sieved through a 0.5mm mesh sieve. The content of the sieve after washing was transferred into a pre-labelled container and 10% formalin was added and transfer to the laboratory for further analysis.

2.3 Extraction of microplastics

The method of extraction employed involved filtration of solids wastes obtained in manta trawl net surface sampling and sediment sampled while plastic waste materials of appropriate size were isolated. The sieved plastic waste materials were air dried under the fume hood to determine the mass in the microplastics sampled. The micropellet particles extracted were subjected to wet peroxide oxidation (WPO) in the presence of a Fe (II) catalyst to absorbed organic matter and sediment attached to the plastic waste. In addition the floating plastics were further isolated from the denser undigested mineral components with a density separator using a custom 0.45mm filter, air-dried, and plastic material were removed and weighed to determine the microplastics concentration (Free *et al.*, 2014; Masura *et al.,*, 2015).

Table 1: Description of sampling stations and solid waste characterization

Station Name	Sampling points	Solid waste characterization
Oworonshoki (ST 1)	LL1	Plastic litters, glass, paper, domestic organics, cloths, and human waste.
	LL2	
	LL3	
AbuleEledu (ST 2)	LL4	Plastic litters, glass, paper, domestic organics, human waste, and wood logs.
	LL5	
	LL6	
Makoko(ST 3)	LL7	Plastic litters, glass, paper, domestic organics, human waste, cloths and wood log
	LL8	
	LL9	
Okobaba (ST 4)	LL10	Sawdust waste, wood log, plastic waste, organic wastes and human
	LL11	

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	LL12	waste
Iddo (ST 5)	LL13	solid waste dump, cement bag washing, and rubber waste
	LL14	
	LL15	
Marina (ST 6)	LL16	plastics, glass, paper, vegetable waste, human waste
	LL17	
	LL18	
Apapa Port (ST 7)	LL19	Oil and grease, spillages, ship garbage and plastic debris
	LL20	
	LL21	
Commandor channel (ST 8)	LL22	Marine litters
	LL23	
	LL24	

2.4 Test chemicals

Analytical grade solvents hexane and dichloromethane were purchased from Sigma Aldrich. Standards of PCBs, and PAHs were purchased from Accustandard (New Haven, CT, USA). PCB 65 and PAHs mixture Z-014J-0.5X (Naphthalene -d8, acenapthene-d10, phenathrene-d10, chrsyene-d12 and perylene-d12) and CLP-LC-SS1 (Nitrobenzene-d5, 1-1'-biphenyl 2-fluroene-d10 and ptertphenyl-d14) were purchased from Accustandard. These standards were diluted with analytical grade hexane to make calibration, internal, and recovery standards.

2.5 Solvent Extraction Procedure

Micropellet particles were extracted for determination of contents of PCBs and PAHs. Approximately 1 g of micropellets was used for extraction, Samples were placed in labelled amber glass bottle and matrix blanks were used as controls. Matrix blank consisted of virgin polyethylene pellets that were not exposed to environmental factors outside of production. About 0.3 g of sodium sulphate was added to each amber bottle in order to remove any moisture present in each sample. Each sample was then spiked with known amounts of internal standards. N-Hexane was added to each sample in equal amounts of 30ml and shaker for 30 minutes using a mechanical shaker (Fisher Scientific, Fair Lawn, NJ, USA). The extraction procedure was then

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repeated a three time (3x) with 15ml of hexane were added to the same amber bottle Extracted samples were then placed in a round bottom flask and attached to rotary evaporator and concentrated to about 2 ml at 40°C.

2.6 Silica gel- solid Phase Extraction (SPE)

Silica gel based SPE cartridges, Sep-Pak, from (Waters, Milford, MA, USA) and glass syringes were assembled on a Visiprep SPE vacuum (Supelco, Bellefonte, PA, USA). After thorough cleaning and conditioning of SPEs, each extract was loaded into SPE and fractionized with 10 ml of hexane and 10 ml dichloromethane (7:3). Flow rate of solvent through SPEs was carefully monitored at this time. The two fractions were combined and concentrated by the TurboVap, transferred to 2 ml amber vials.

2.7 Identification and Quantification of PCB Congeners and PAHs derivative

Identification and quantification of 28 PCB congeners (congeners (PCB 8, PCB18, PCB 28, PCB 44, PCB 52, PCB 60, PCB 77, PCB 101, PCB 81, PCB 105, PCB 114, PCB 118, PCB 123, PCB 126, PCB 128, PCB 138, PCB 153, PCB 156, PCB 157, PCB 167, PCB 169, PCB 170, PCB 180, PCB 185, PCB 189, PCB 195, PCB 206 and PCB 209) was performed with a gas chromatograph (GC)-electron capture detector (ECD) (Agilent 7890A GC- (ECD Detector) using USEPA Method 608. sixteen(16)PAHs (naphthlaene (NAP), acenaphthylene (ACY), acenaphthene (ACE), fluorene (FLU), phenanthrene (PHE), anthracene (ANT), fluoranthene (FLT), pyrene (PYR), benzo[a]anthracene (BaA), chrysene (CHR), benzo(b)fluoranthene (BbF), benzo[k]fluoranthene (BkF), benzo[a]pyrene (BaP), benzo[ghi]perylene (BghiP), dibenzo[a,h]anthracene (DahA), indeno[1,2,3-cd]pyrene (IND)). Analysis was conducted

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utilizing Agilent Gas Chromatography (GC-7890A) coupled with Flame ionization Detector (FID) Column: HP5 (30m x 320um x 0.25um) along with internal and recovery standards.

2.8 Quality Assurance

All glassware was thoroughly cleaned and baked in the oven at 140°C for thirty minutes (30mins) and glass syringes at 45°C for one hour. During the extraction procedures, samples were all carefully covered with aluminium foil in order to prevent contamination. All glass pipettes, test tubes, and vials were discarded after single usage.

2.9 DATA ANALYSIS

Analysis of results was completed by using the sum totals of 28 PCBs and 16 PAHs. One way Analysis of variance (ANOVA) with Pos –Hoc Duncan multiple range test was conducted coupled with descriptive analysis means and standard deviations) from the sums of each EDCs compound analyzed. Data was sorted micropellets extracted from surface water and sediment according to sampling locations.

3.0 RESULTS

3.1. Micropellet particles occurrence and distribution among the sampled environmental matrices

The occurrence of micropellets sampled within the environmental matrices indicates pellet particles occurs more in surface water than sediment. Almost all of the plastic micropellet particles (91.6%) were round in shape, with only 8.4% non-cylindrical in shape; maximum occurrence in surface water (67%) and (33%) in sediment sampled (Figure. 2). Most common colour in all size class of micropellet particles extracted in surface water and sediment in all the

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sampling stations was white (46.97) and opaque in sediment (53.03) (Figure 3). Majority (63.20%) fell between 2–3mm size classes in surface water while 26.8 % size class in sediment was between 1-2mm. The highest mean occurrence concentration of micropellet particles was observed in surface water samples from the marina axis at sampling point LL16 (4692 micropellets particles/L), followed closely by samples collected at Commodore channel at sampling points LL22 (4165 Micropellets particles/L) and the lowest occurrence is observed at Makoko station at sampling point LL7 (figure 4).

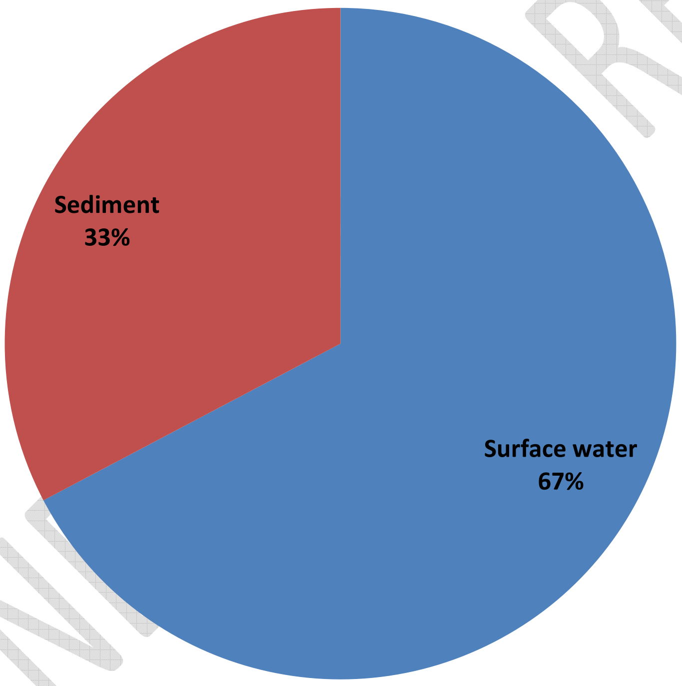

Figure 2: Distribution of micropellet particles in environmental matrices

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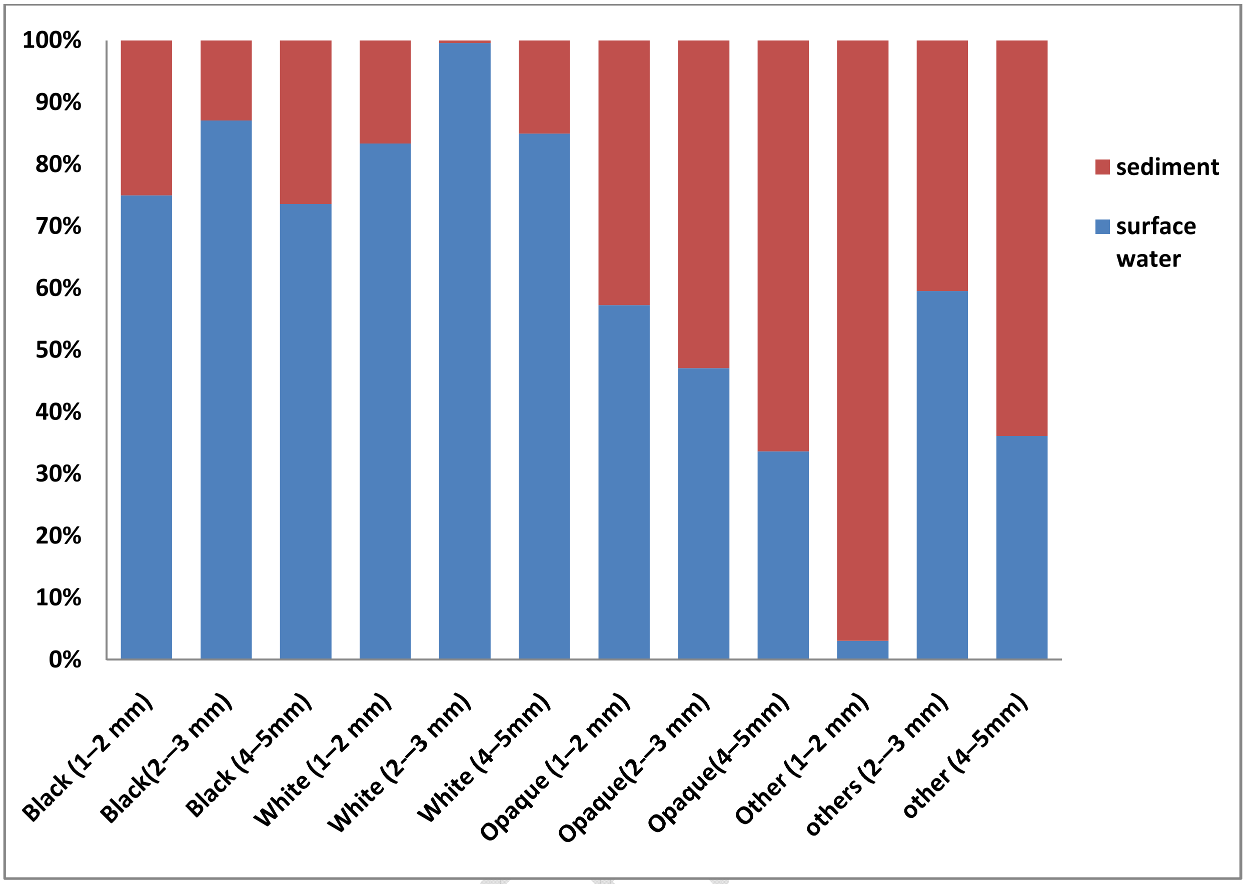

Figure 3: Percentage of plastic micropellet particles with each colour in each size class from each environmental matrix.

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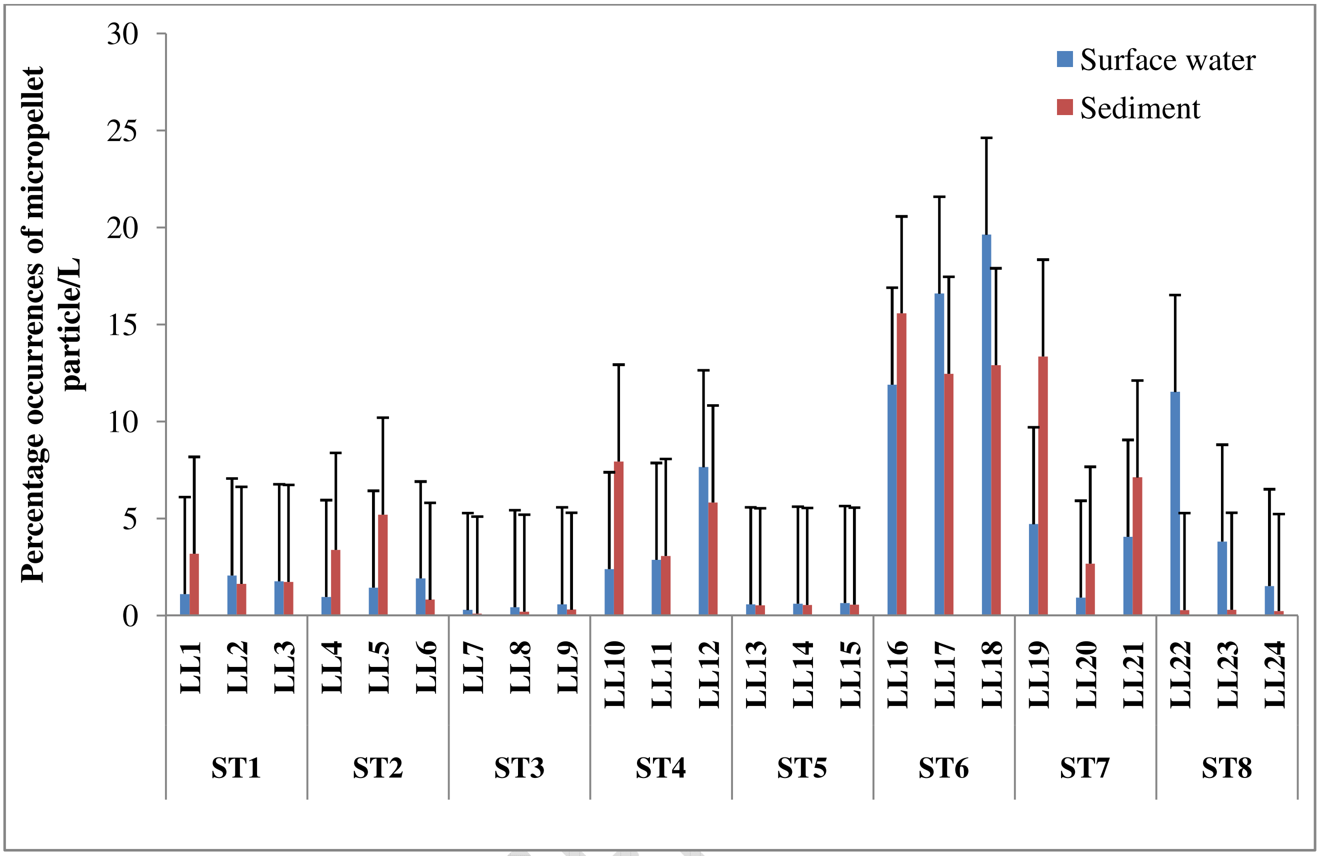

Figure 4: percentage occurrences of micropellet particles in each sampling points

3.2 EDCs contents in extracted pellet particles from surface water and sediment

All micropellets samples contained detectable amount of persistent organic EDCs (figure 5-10), demonstrating the ubiquitous nature of these contaminants. Inter-stations differences in the concentrations of individual EDCs were apparent in all the sampling stations.

3.2.1 PCBs in Micropellet particles extracted from surface water and sediments

The ΣPCBs concentration varied between 76 and 1043 ng g^{-1}, which was significantly P (< 0.01 and 0.05) higher in the surface water than in sediment (Figure 5). The maximum ΣPCBs concentration was found at ST 5 from micropellet particles extracted from surface water while

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the highest notably concentration of PCBs in micropellet particles extracted from sediment was detected in ST 8 (873 ng g^{-1}), with two to three orders of magnitude higher than that recorded for some of the other stations. In respect of sampled matrices PCB 52 and PCB 77 are the most abundance in surface water and sediment while PCB 195 was relatively low in the pellet particles extracted from surface water and sediment (Figure 6 and 7)

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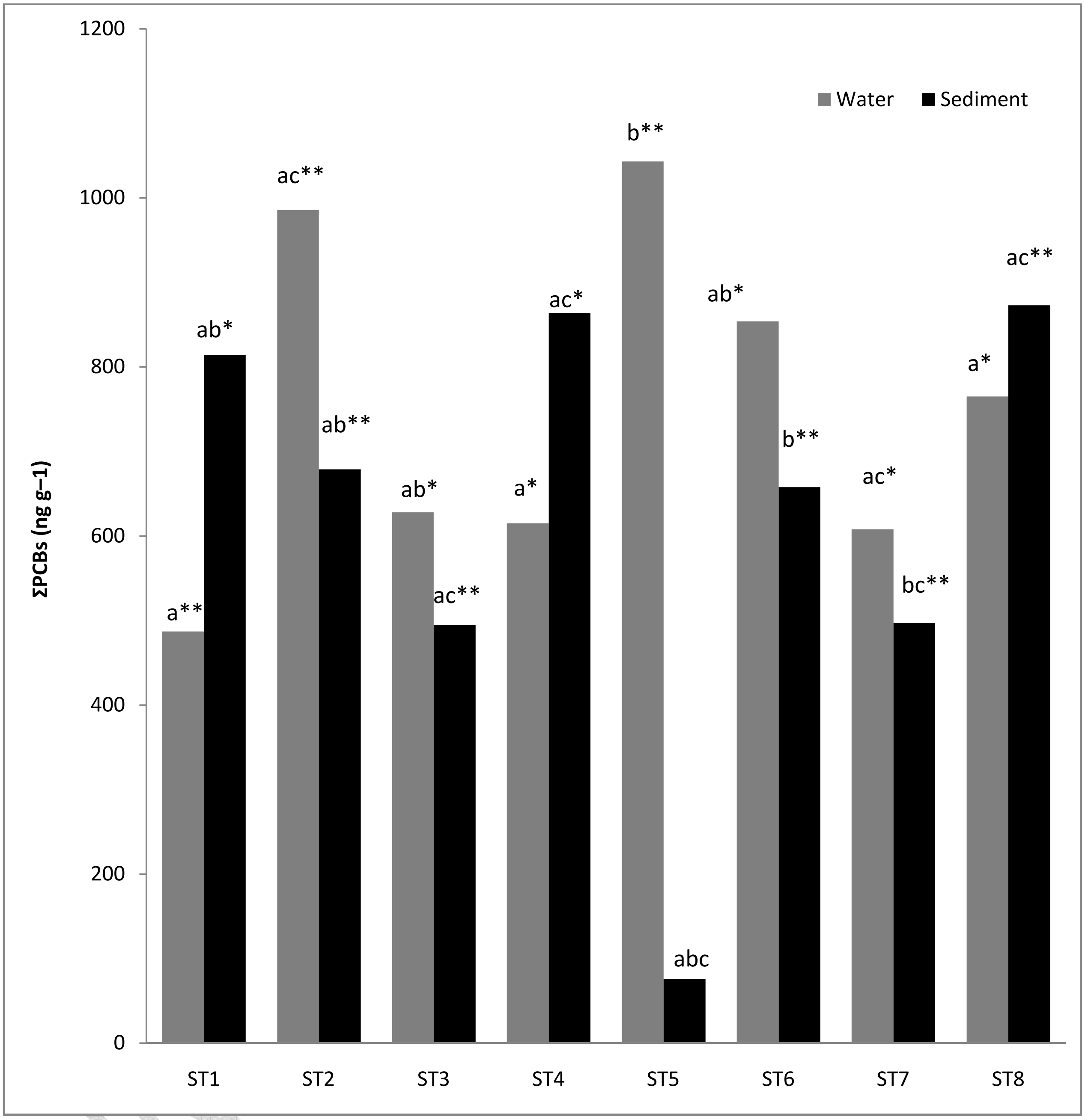

Figure 5: Mean concentrations of ΣPCBs (28 congeners) in microplastics (ng g^{-1}) in Surface water and sediments.

Means and standard deviations of three replicates are shown; bars with different letters indicated significant differences among sampling stations at the level of $p < 0.05$ according to one-way ANOVA and post-hoc Duncan multiple range test; single asterisks (*) and double asterisks (**) indicated a significant difference between environmental matrices at the level of $p < 0.01$.

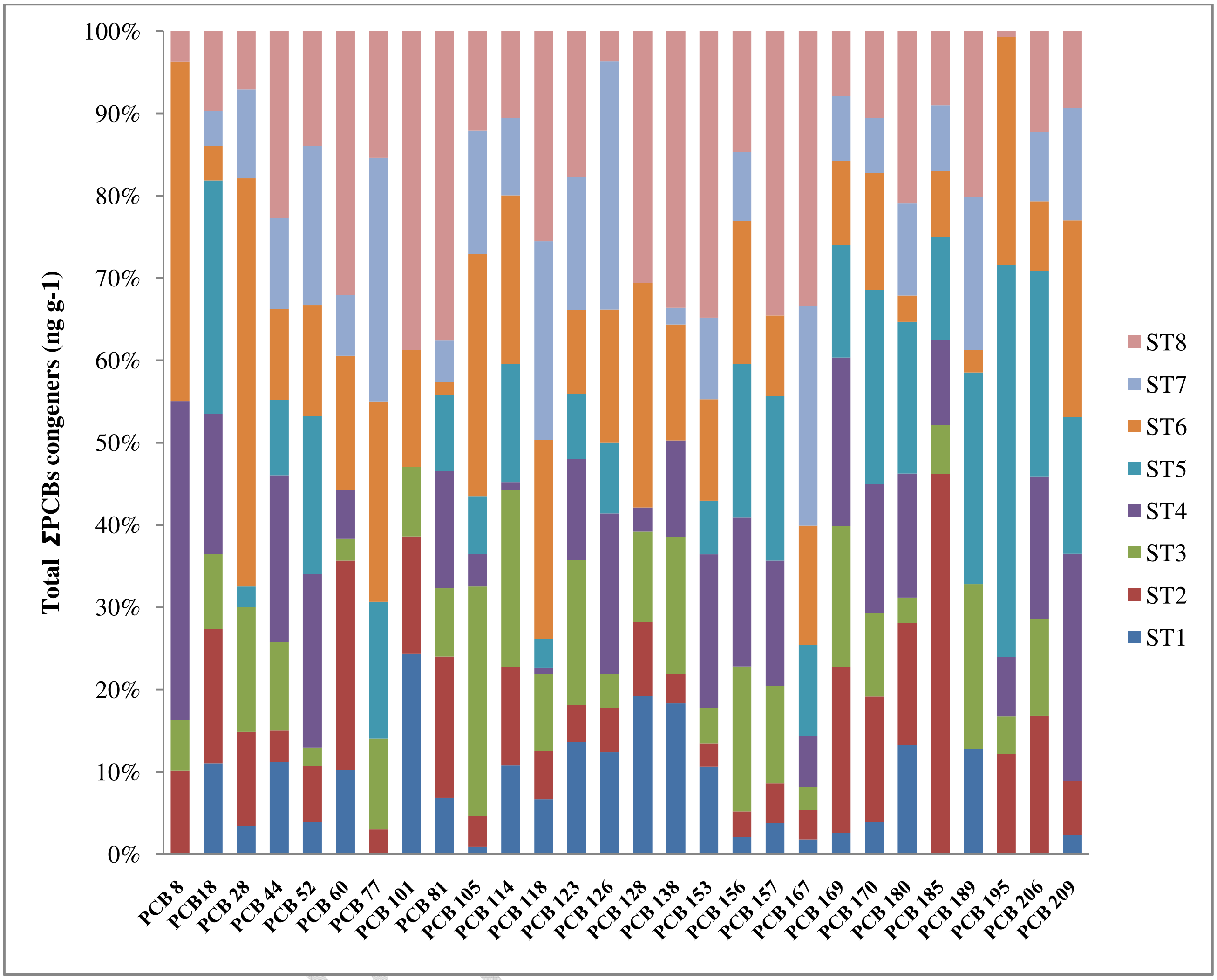

Figure 6: Mean concentrations of ΣPCBs (28 congeners) extracted from micropellets in surface water

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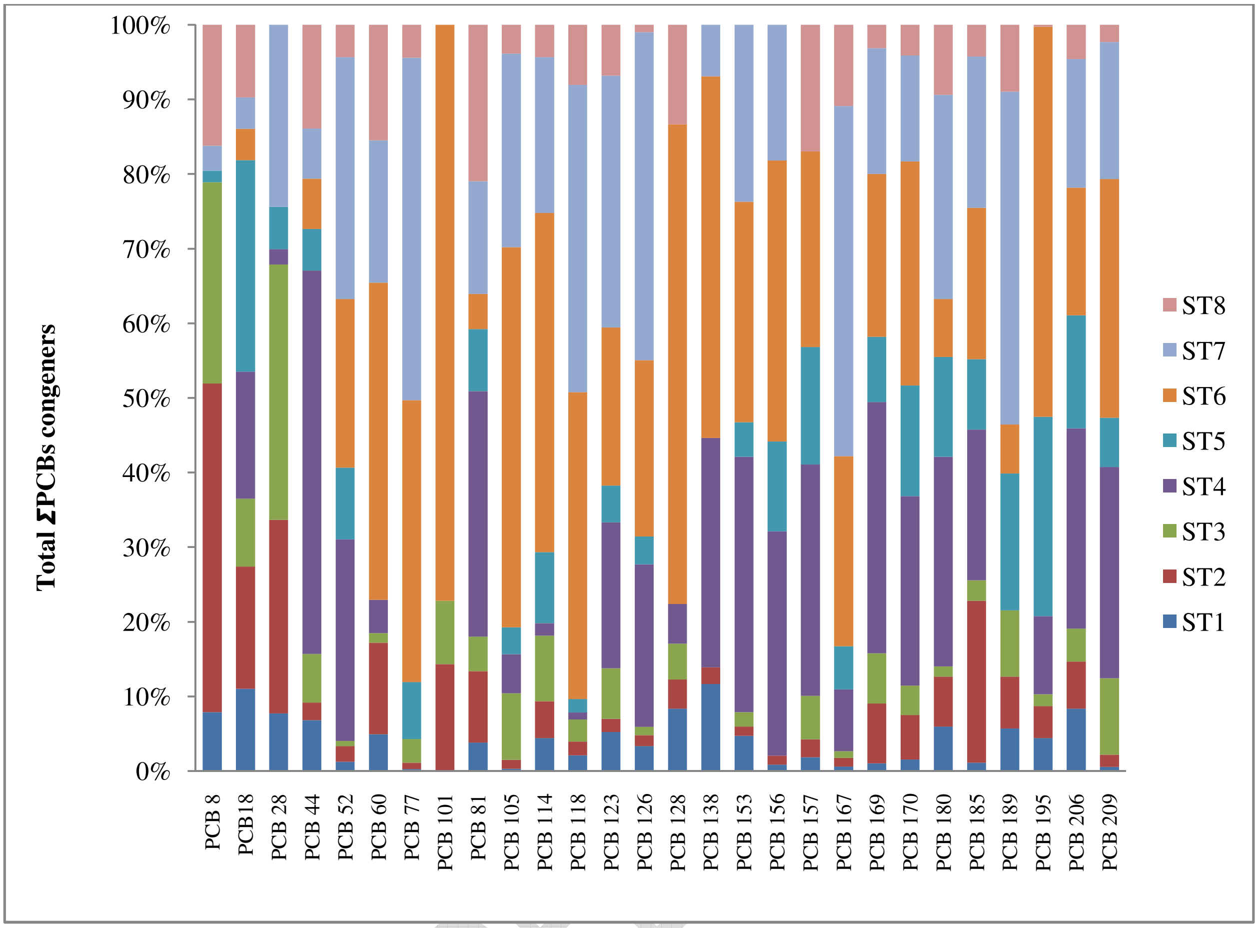

Figure 7: Mean concentrations of ΣPCBs (28 congeners) extracted from micropellets in sediment

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3.2.2 PAHs in Micropellet particles extracted from Surface water and sediments

The total PAHs concentration ranged between 46.05 ng g^{-1} (ST1) and 3984.04ng g^{-1} (ST 6) within micropellet particles extracted in the environmental matrices (Figure 8). When individual station were compared in regards to the environmental matrices, all the stations have PAHs types three 5–6 rings PAHs .On the other hand, sites ST1 and ST6 were greatly affected by 2–5rings PAH. Regardless of stations, 3–4 rings PAH dominated in this study (Figure 9 and 10). The PAH diagnostic ratios (Figure8) indicated PAHs that varied among Stations as well as within the environmental matrices). The related distribution pattern for EDCs displayed in ST 7 and ST 6; contamination of PAHs could be an indication that local contamination sources probably would have contributed to such difference observed across the sampling station.

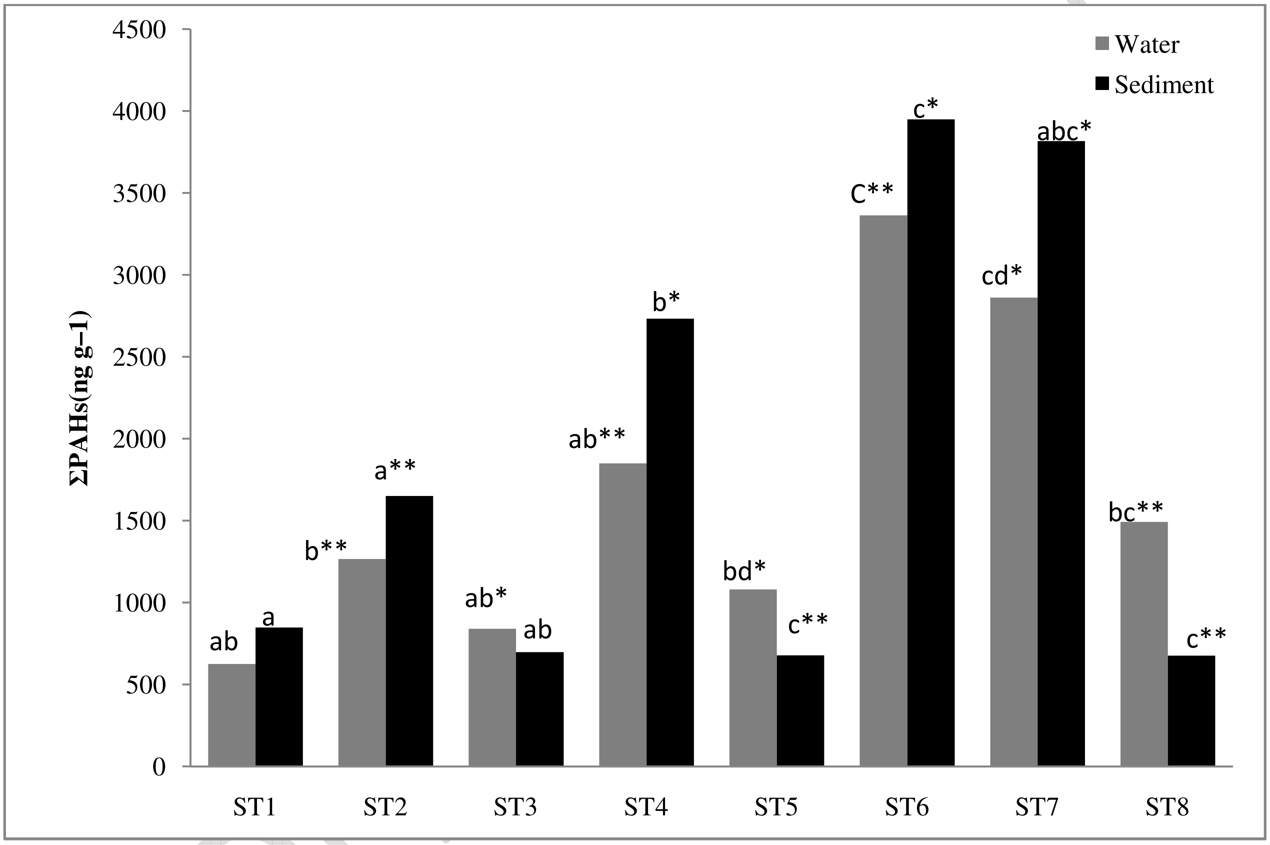

Figure 8: The concentrations of HEDCs in micropellet (ng g–1) of ΣPAHs (16 congeners).

Means and standard deviations of three replicates are shown; bars with different letters indicate significant differences among sampling stations at the level of $p < 0.05$ according to one-way ANOVA and post-hoc Duncan multiple range test; single asterisks (*) and double asterisks (**) indicate a significant difference between matrices at the level of $p < 0.01$

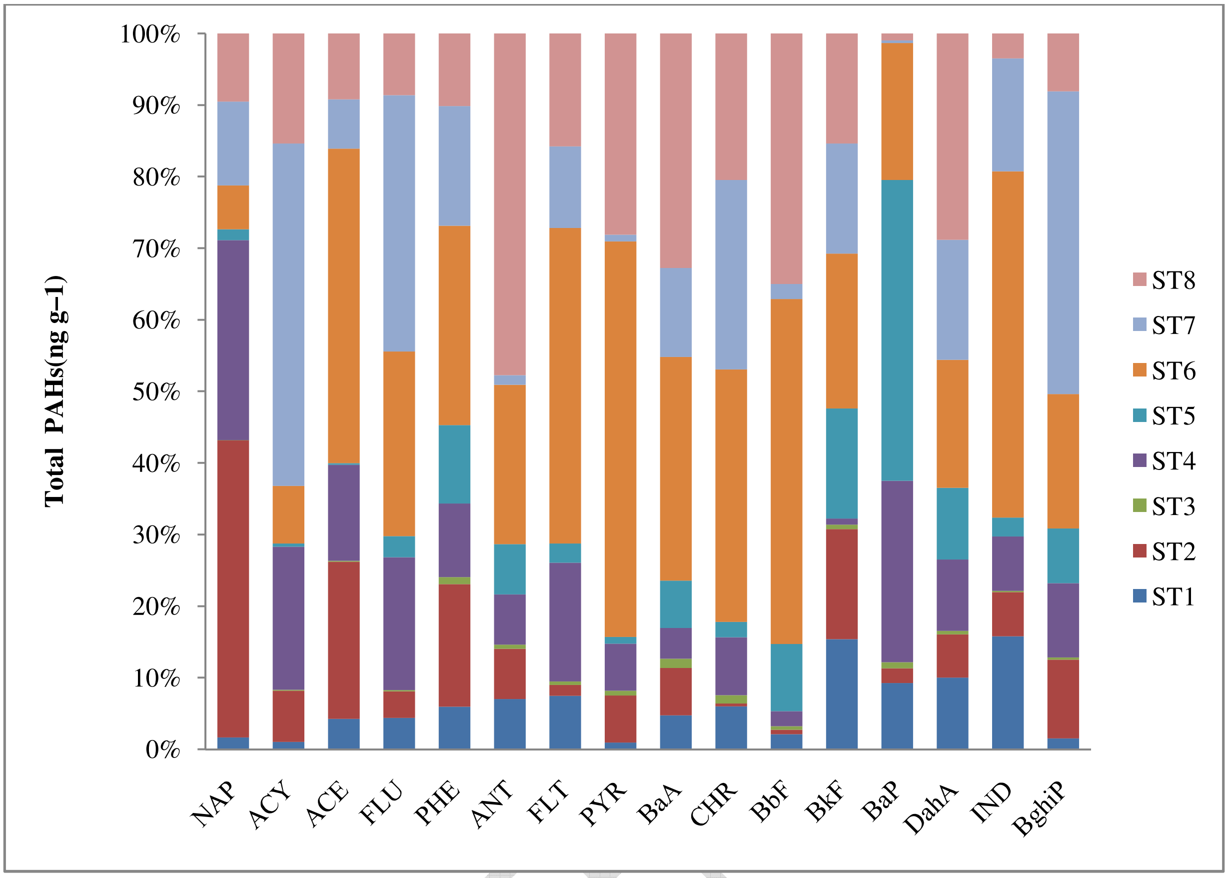

Figure 9**:** The concentrations of ΣPAHs (16 congeners) extracted from micropellets in surface water

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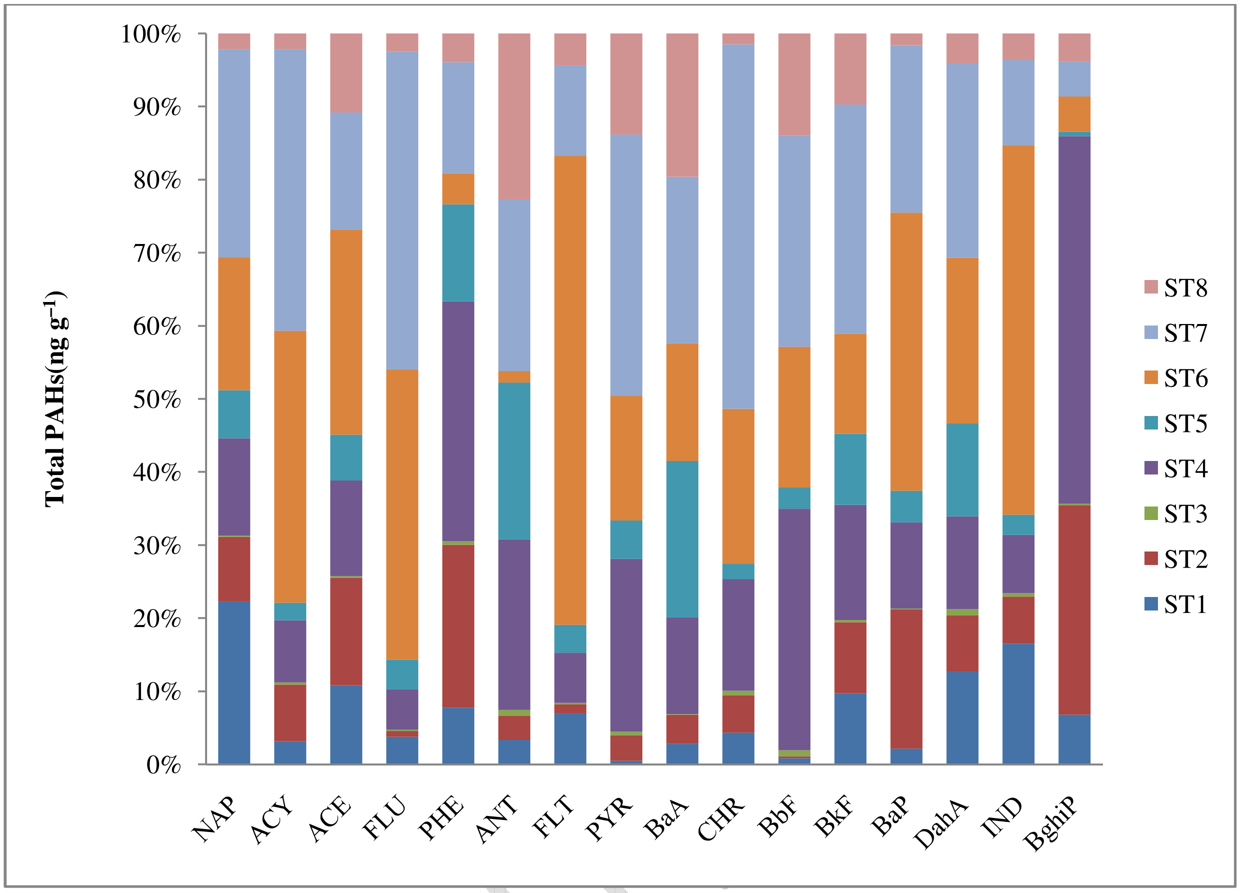

Figure 10**:** The concentrations of ΣPAHs (16 congeners) extracted from micropellets in sediment

4. Discussion

This study evaluates micropellet particle occurrence and distribution in (surface water and sediment) and their associated hydrophobic endocrine disrupting chemicals in the extracted micropellet sampled. In the present study, the micropellet particles collected were higher in surface water than sediment were mostly opaque and white, a finding in agreement with other studies that have reported that most micropellet found in environmental matrices are often white or opaque (Heo *et al.,* 2013; Corcoran, 2015;Veerasingam *et al.*, 2016). This finding is not surprising because white micropellet particles are the most common colour manufactured (Redford *et al.*, 1997) worldwide. However, one author have recorded frequency of yellow

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micropellet (Karapanagioti and Klontza, 2007) and according Veerasingam *et al.* (2016) they are the second most frequently observed micropellet particles often associated with micropellet particles in environmental matrices, a results that was not established in this present study. Although, the difference observed probably may be due to difficulties in colour definition by those researchers counting opaque micropellet particles as "yellow" whereas in the present study they were counted as opaque colour. According to Wright *et al.* (2013), white color composition of micropellet particles are similar in colour to most plankton organisms, a primary food source for most aquatic organisms dwelling in the pelagic zone. The occurrence of micropellets particles within the marine environment is currently well recognized in the water column, at the sea surface and sediments (Law and Thompson, 2014). It has been documented that micropellet particles also accounted for about 10% of all reports of ingestion of aquatic debris, highlighting their importance as a component of aquatic debris (Gall and Thompson, 2015). The size of micropellet particles makes them accessible to organisms with a range of feeding methods, including: filter feeders (mussels, barnacles), deposit feeders (lugworms) and detritivores (amphipods, sea cucumbers) and zooplankton (Wright, *et al.*, 2013). Due to diversities in their size, a substantial proportion could possibly be discharged into creek, river, estuaries and the oceans. Micropellet particles have been reported to generally concentrated in the areas of nutrient upwelling (Doyle *et al.*, 2011), which possibly accounted for the significant numbers detected around solid waste litter and probably influenced by local weather condition systems (Moore *et al.*, 2002; Lattin *et al.*, 2004; Reisser *et al.* 2013). PCBs were the most frequently encountered organic contaminant, and total PCBs on micropellet particles were higher and varied according to geographical location and frequency of pellets occurrence (Rochman, 2015; Fotopoulou and Karapanagioti, 2017), nevertheless, PAHs concentrations in micropellet particles obtained in

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this study were generally lower than the values reported elsewhere. Some authors reported high concentrations of priority PAHs contamination in micropellet particles collected in coastal region (Ziccardi *et al.*, 2016; Ivleva, *et al.,* 2017;Mendoza, *et al.,* 2016). Sixteen priority PAHs concentration was detected in the floating marine plastic debris collected from Northern Pacific Gyre varied between undetected and 14459 ng g^{-1} (Rios *et al.,* 2010). When compared with the concentrations found in marine environment in previous studies, much higher concentrations of PAHs were obtained in micropellet in this study.

Differences in PAHs level across the sampling stations were apparent, even for stations very close to each other. This probably indicated that there is possibility of input PAHs at preproduction of plastic pellets. The presence of EDCs in the environment may have ecological and health consequences not only for aquatic fauna but also for humans, as EDCs can enter the food chain and bioaccumulates. The range of values of polycyclic aromatic hydrocarbons and polychlorinated biphenyl studied confirmed large fluctuations within the period of study possibly influenced by anthropogenic activities. This study corroborated with the reports of previous authors (Nubi, *et al.*, 2010; Amaeze, and Abel-Obi, 2015; Abiodun and Oyeleke, 2016; Soneye, *et al.*, 2018) in South-West Nigeria that plastic waste materials litter the Lagos Lagoon environment. Furthermore, a potential problem associated with micropellet particles contamination is the likelihood of transport of endocrine disrupting chemicals contaminants by plastic waste particles which have been established in this study to adsorbs onto surface of plastic waste materials and may transfer to biota upon ingestion as reported by many authors (Teuten *et al.*, 2009;Devriese *et al.*, 2015; Koelmans *et al.*, 2016;Suaria *et al.*, 2016).

Conclusion

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The present study showed that micropellet particles are present in environmental matrices with differential affinities for sorption of endocrine disrupting chemicals that may alters the hormonal behavioural and physiology of aquatic fauna if injected are likely to threat aquatic resource. This calls for urgent monitoring of Lagos lagoon (where plastics products are abundantly manufactured and used with reckless abandon) and other coastal region in Nigeria in order to mitigate the danger of plastic waste materials in our coastal bodies.

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