

# Levels and Distribution of Polycyclic Aromatic Hydrocarbons (PAHs) in Marine Sediments of Eastern Bank Weshia Peninsular, Pemba Island, Tanzania

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## Abstract

Levels of Polycyclic aromatic hydrocarbons (PAHs) from sediments collected in July 2017, a week after tanks oil leak in the bay of Weshia Peninsular, Pemba Island are reported. Four sampling points were chosen alongside the mangroves where a total of twenty samples were collected during the low tides. Eight PAHs were detected with concentrations ranging from ND to 53.42 ng/g (dw). Total PAHs in sediments was 158.38 ng·g<sup>-1</sup>. Phenanthrene residues (lower molecular weight) were dominant followed by pyrene (higher molecular weight PAHs) accounting for 90% and 40% detection frequencies, respectively. Statistical analysis showed no significant difference in the concentrations of PAHs among sites ( $p < 0.05$ , ANOVA). Vertical distribution of PAHs shows no definite patterns with sediment depth, however, composition patterns of PAHs along sediment depth indicate that higher molecular weight PAHs tend to remain in middle layer (2 - 4 cm). The results advocate the needs of proper monitoring to avoid the possible consequences to the public health.

## Keywords

PAHs, POPs, Sediments, Marine Pollution, Weshia

## 1. Introduction

There is a growing concern about persistent organic pollutants (POPs) and polycyclic aromatic hydrocarbons (PAHs) in marine environment. The distribu-

tion of these chemical compounds is globally in a variety of environmental compartments including marine [1]. Marine pollution is a subject of global interest due to the large number of toxic substances transported to marine environments [2]. The pollutants are capable of residing in marine sediments and water due to their physical-chemical properties [3].

In May 2001, a global treaty for the regulations of POPs was signed the “Stockholm convention” for the purposes of total elimination of the 12 POPs on a global scale [4]. Although PAHs are not part of the 12 priority pollutants of the Stockholm convention but were listed by the United States Environmental Protection Agency and the European Commission as priority pollutants. Previous studies have shown the presence of large amount of organic compounds in the marine environment, the most important of which belong to PAHs and OCs [5].

These ubiquitous environmental pollutants have been extensively studied due to their toxicity to ecosystem and environment as whole [6]. The occurrence of PAHs pollutants is a result of both natural and anthropogenic sources and normally found in marine water and sediment where they can be taken up by marine animals [7]. Due to the low solubility, hydrophilic characteristics and higher persistence, POPs tend to adsorb and highly accumulate in marine sediments and can cause major hazards for aquatic life in marine environments, particularly in areas close to anthropogenic source [8]. The increase in concentration of POPs pollutants in marine environment can pose higher risks to plants and animals including fish at alarming rate [9] [10].

Numerous cases of POPs and their byproducts pollution have been reported in different marine areas of the world [11]. The uses of these chemicals have also led to environmental pollution to non-target organisms through sub lethal exposure and chronic toxicity [12]. In 1970s, these compounds were prohibited in many developing countries although they are still used [13].

Zanzibar coastal areas as a part of the world have been reported to have a number of cases related to organic pollutants originated from different sources, such as sea transportation, fuel-based power generation, the tourism industry and accidental oil spills along the coast. The potential source of PAHs in coastal environment of Pemba Island can be associated with the use of diesel mixture (with heavy oil) for generation of electric power from Wesha power station located few meters from the sea. The station has been working for almost three decades. The oil residues and other wastes were disposed by delivering to sea from oil tanks through pipes. Wesha station is the only port that handles about 2000 tons of different types of oil that is transported to Pemba Island including petrol, diesel, kerosene and heavy oil (industrial diesel oil). Poor management and technological facilities to deal with oil pollution emergency like an accidental spill or leakage during operational activities characterize both depots. Various studies have indicated the presence of POPs in Zanzibar marine waters and their residues were detected in sediments and crabs [14] and fishes [15] as well as in marine seafood [16]. The study aims were to measure levels and vertical distribution of PAHs in marine sediments at Wesha Bay in Pemba Island. **Figure 1**



**Figure 1.** The image of affected mangroves after oil spill at Wesha coastline.

shows the affected mangroves after oil spill in Wesha during the time of this study.

## 2. Materials and Methods

### 2.1. Study Area

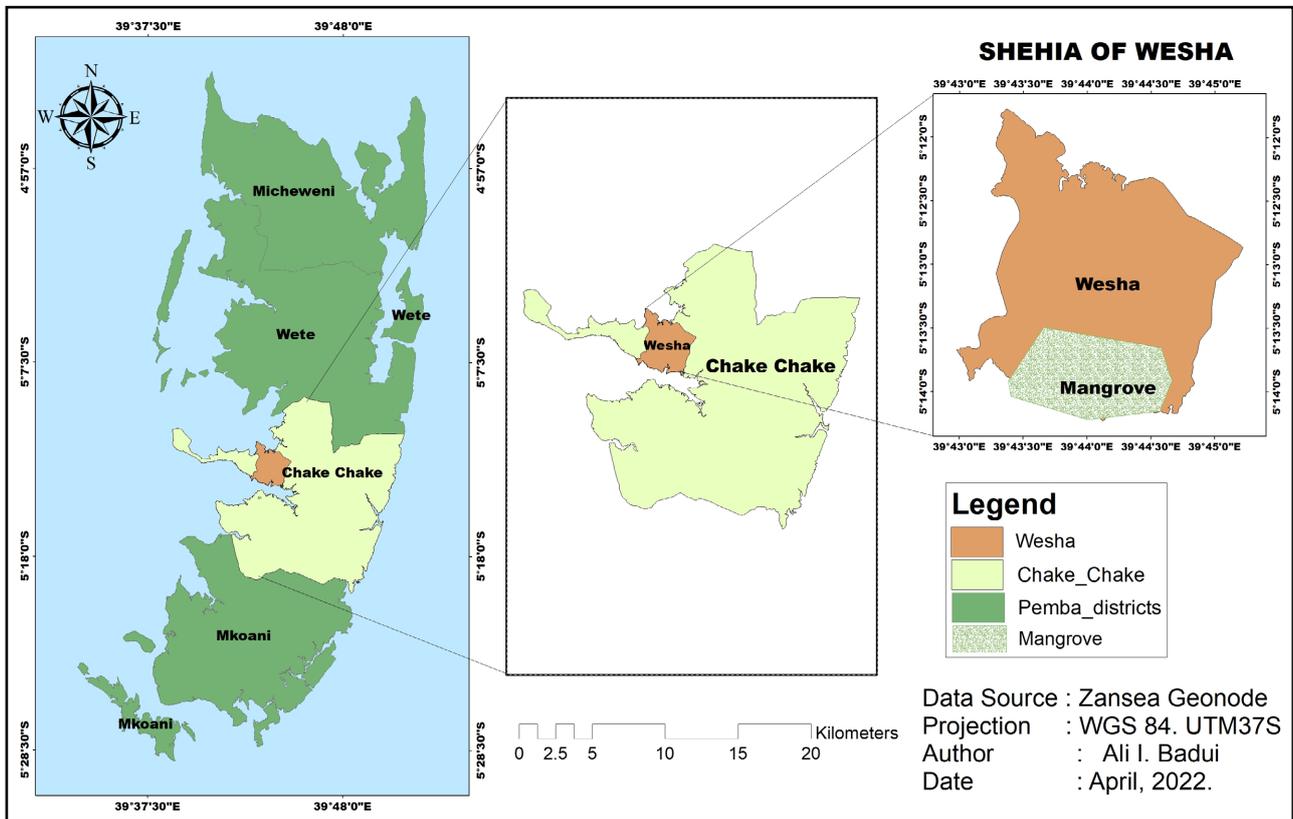
This study was conducted at Wesha, a small village located within Chake-Chake District (Coordinates: 5° 15'S, 39° 7'E), Southern region of Pemba Island in Zanzibar (**Figure 2**). The village is about 5.2 km from Chake-Chake, the central town of Pemba Island. The study area is the eastern part of coastal environment of semi-enclosed bay, which opens to the Indian Ocean channel. Wesha bay is widely known in Pemba for providing very important socio-economical activities not only for the nearby community but also for the islands at large. The bay houses an oil depot through which all types of fuel oils used in Pemba are shipped and temporary stored before being distributed to selling points. The coast also hosted the former power plant, which was the only source of electricity on Pemba-Island. This study, reports the distribution levels of PAHs in marine sediments in Wesha Eastern Peninsular across the sea after 10<sup>th</sup> July 2017 oil leak.

### 2.2. Sampling

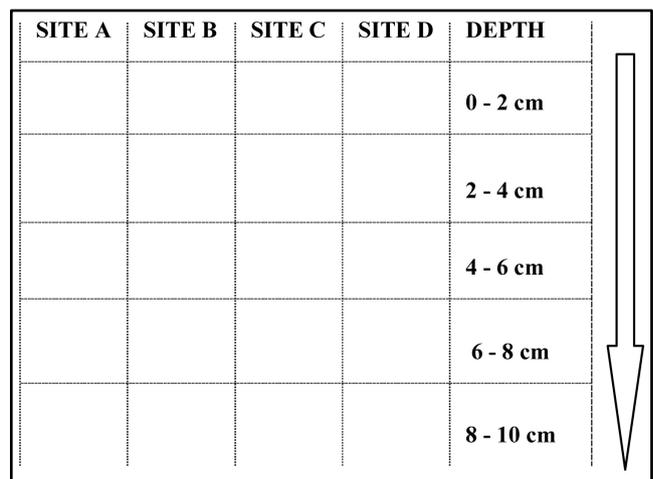
The sampling exercise was done on 22<sup>nd</sup> July 2017 using sampling core technique. The selected site for the collection of organic contaminants in marine sediment covered the area of (180 × 15) m<sup>2</sup>. The sampling area was divided into four points. Each core of 10 cm depth was sliced at interval of 2 cm depth apart from top making five samples per core and total of twenty samples in all area (**Figure 3**). Each sample was then separately sealed with aluminum foil. The samples were then transported to the laboratory in icebox at 4°C and stored at -20°C in the deep freezer until analysis.

### 2.3. Extraction

The samples were defrosted and thoroughly mixed by using stainless metal spatula to give homogenous samples. 50 g of sediment samples were freeze-dried (labconco freezezone 2.5 L) for 24 hrs. A mass of 15 g of individual freeze-dried



**Figure 2.** Map of Pemba Island, Tanzania shows sampling location.



**Figure 3.** Schematic diagram showing depth intervals of sediments in sampling area.

sediment was put in a thimble and then covered with cotton wool at the top. The thimble with samples was then extracted by Soxhlet extraction technique using dichloromethane according to the method described by [17] [18].

### 2.4. Clean-Up

The extracts were cleaned by column chromatograph with pre-backed silica gel

using cyclohexane. The extract was then eluted using 20 mL of cyclohexane/acetone 9:1 mixture. The eluted volume was reduced to 1.00 ml and quantitatively transferred into a GC [19] [20].

### 2.5. The GC-ECD Analysis

The extracted samples were analyzed using gas Chromatography-Mass Spectrometry (GC MS—2010 Shimadzu instrument operating in EI mode—MS) at 70 eV, and ECD detector for GC. The injection temperature was 250°C with splitless injection mode. A Restek-5MS column (30 m × 0.25 mm × 0.25 μm) was used. The oven temperature program was 90°C to 280°C held at 90°C for two minutes before being raised at a rate 6°C per minute. Helium was used as carrier gas at a flow rate of 1.21 mL·min<sup>-1</sup>. The ion source temperature and interface temperature in MS were 230°C and 300°C respectively.

### 2.6. Blanks Determination and Recovery

Each batch of processed sample was followed by blank and recovery samples that were subjected to the same protocol as explained by [21]. Blanks taken from remote area with reference to sampling area aimed at evaluating and monitoring potential introduction of contaminants into samples during processing or determining background levels of the measured properties, while recovery tests were conducted to reflect ability of the used laboratory methods to fully recover surrogate compounds in the samples matrix at the beginning of the procedure. Blank experiments were carried out by running the same amount of pre-cleaned sediments as the samples along the same analytical procedure and did not show any contamination.

### 2.7. Analytical Quality Assurance

All glassware and other equipment used for sampling and preparation were checked for PAHs before use. GC-MS analysis showed that procedural blanks prepared from sediments during sample preparation protocol were free from contamination. The recovery test of the target analytes (PAHs) gave percentage recovery ranging from 91% to 112%. The blanks and the method recovery were within the acceptable range of 5% - 120% [21].

## 3. Results and Discussion

The chromatographic analysis revealed the presence of a total of eight polycyclic aromatic hydrocarbons (PAHs) among the 16 priorities PAH pollutants of concern. The detected PAHs were Acenaphthalene (ACE), Phenanthrene (PHE), Anthracene (ANT), Pyrene (PYR), Chrysene (CHRY), Benzo(a)pyrene (BaP), Fluorine (FLR) and Benz(a)anthracene (BaA). The concentration of the investigated total PAHs in sediment samples varied from site to site (Table 1).

One-way ANOVA was used for the statistical analysis. The differences in contaminants concentrations in sediment samples from the four locations were

**Table 1.** Concentrations of PAHs in sediments (ng/g dw).

SITE	SAMPLE	ACE	PHE	ANT	PYR	CHRY	FLR	BaA	BaP
A	A1	ND	1.65	ND	ND	ND	ND	ND	ND
	A2	ND	53.42	ND	ND	ND	ND	ND	ND
	A3	ND	3.10	0.66	ND	ND	ND	ND	ND
	A4	ND	20.33	ND	ND	ND	ND	ND	ND
	A5	1.78	16.77	ND	7.93	ND	ND	ND	0.69
B	B1	ND	1.72	0.45	0.54	ND	ND	ND	ND
	B2	0.67	0.50	ND	2.24	ND	1.39	ND	ND
	B3	0.56	5.42	ND	2.15	ND	ND	ND	ND
	B4	ND	1.44	0.68	2.80	ND	ND	1.55	ND
	B5	ND	2.55	0.57	ND	ND	ND	ND	ND
C	C1	ND	0.53	ND	0.14	ND	ND	ND	ND
	C2	0.74	0.49	ND	6.32	1.16	ND	ND	ND
	C3	ND	0.58	ND	ND	ND	ND	ND	ND
	C4	ND	3.14	ND	ND	ND	ND	ND	ND
	C5	ND	ND	ND	ND	ND	ND	ND	ND
D	D1	0.09	2.39	0.68	ND	ND	ND	ND	ND
	D2	ND	ND	0.59	ND	ND	ND	ND	ND
	D3	ND	0.97	ND	ND	ND	ND	ND	ND
	D4	ND	3.14	ND	ND	ND	ND	ND	ND
	D5	0.49	3.86	ND	1.51	ND	ND	ND	ND

ND = Not Detected.

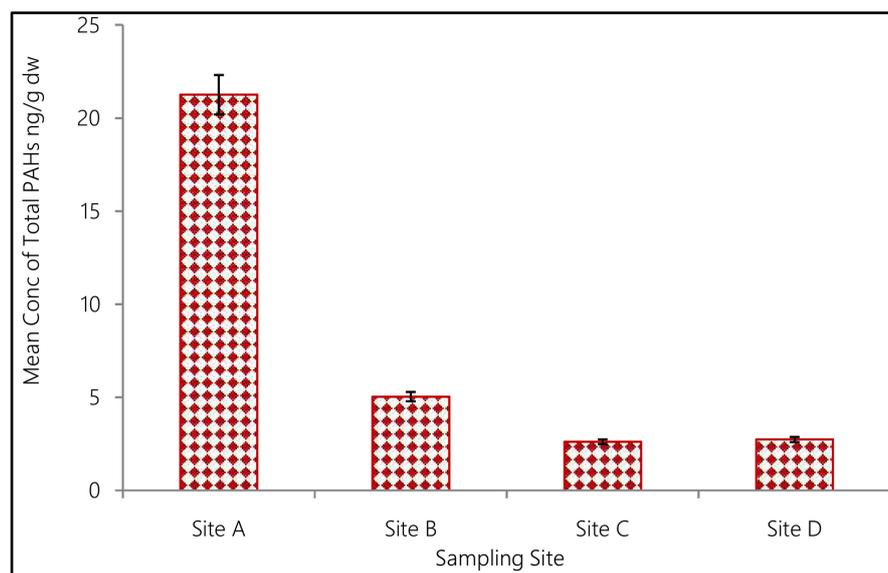
compared using analysis of total means (one way-ANOVA). Normally, the distributed data were evaluated via one-way ANOVA, with an alpha level of 0.05. The differences were determined to be significant, where  $p < 0.05$ . Since the p-value (Table 2) shows that  $p > 0.05$ , thus the results from this study are not significant.

The highest total PAHs was detected in site A where concentration ranged from 1.65 ng/g - 53.42 ng/g dw with mean total concentration of 21.27 ng/g followed by site B (2.71 - 8.13 ng/g dw) with mean total concentration 5.05 ng/g. Site D was third position with total concentration ranged from 0.59 - 5.86 ng/g dw whereas mean total concentration was 3.28 ng/g dw. The lowest mean total concentration was detected in site C (2.62 ng/g dw) with total concentration ranged from ND to 8.71 ng/g dw. The distribution of mean total PAHs concentrations is presented in Figure 4.

Generally, the total concentration of total PAHs showed the following trend  $A > B > D > C$ . On the other hand the composition of PAHs site-wise showed that all sites were dominated by LMW-PAHs except in site C where HMW-PAHs

**Table 2.** P-value of  $\Sigma$ total sampling sites of the present study.

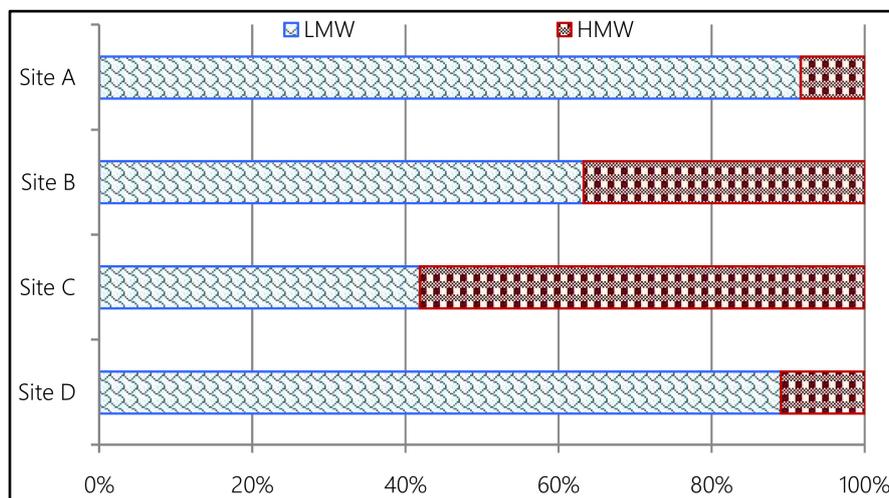
	Sum of Squares	Df	Mean Square	F	Sig.
Between Groups	24.237	17	1.426	2.851	0.438
Within Groups	0.500	1	0.500		
<b>Total</b>	<b>24.737</b>	<b>18</b>			

**Figure 4.** Comparison of total mean concentration of PAHs found in sediments (ng/g).

were relatively higher in LMWPAHs. The dominance of LMWPAHs is more pronounced in site A and D. The LMWPAHs concentrations contributed 91.6% and 89% of the total PAHs at site A and D respectively. At site B the LMWPAHs accounted for 63% of total PAHs. In contrary, the HMWPAHs had higher contribution (63%) in total PAHs compared to LMWPAHs (37%). **Figure 5** shows overall spatial compositions of the measured PAHs.

Higher contribution of LMWPAHs in total PAHs is an indication of recent release and the PAHs are originated from petrogenic sources [22]. The dominance of HMWPAHs particularly to areas polluted by oil pollution is an evidence of aged release. As the source of PAHs at Weshia is obviously petrogenic, very high dominance of LMWPAHs is clear indication that eastern part of Weshia Peninsular was largely affected by new release of oil [23]. The high contribution of HMWPAHs observed at site C indicates that the area was also affected by previous pollution. Furthermore, higher contribution of LMWPAHs was attributed phenanthrene that was predominantly detected. The PAH phenanthrene is known to have low to intermediate aqueous solubility and hydrophobicity and it is reported to have significantly sorbed to soil particles [24].

Fraction of HMWPAHs in this study was mainly attributed by detection of



**Figure 5.** Comparison of percentage concentration of  $\Sigma 8$  PAHs found in sediments (ng/g) collected at the Weshia eastern Peninsular.

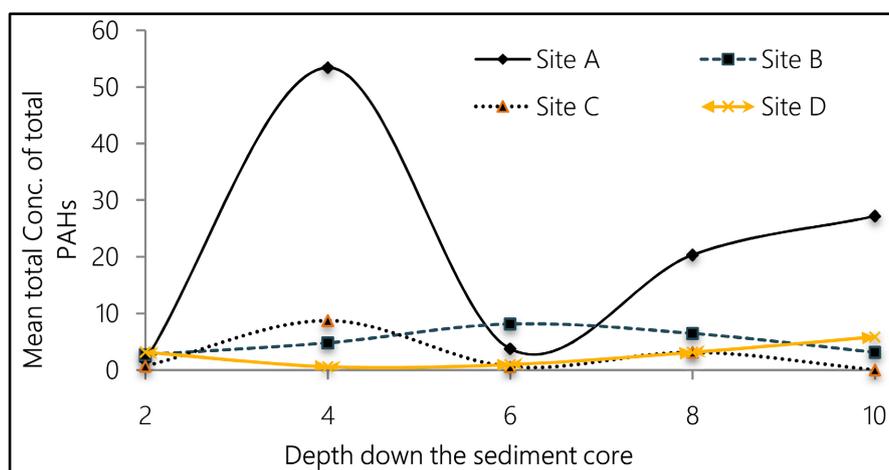
Pyrene. This compound has lower water solubility, higher Log Kow, higher hydrophobicity, non-volatile, and strongly adsorbed into sediment compared to Phenanthrene [25]. These make pyrene ability to resist to degradation such that can remain in the sediment even 12 years after oil spills.

On the context of vertical distribution, PAHs indicated an increase in mean total concentrations from layer 1 (0 - 2 cm) and layer 2 (2 - 4 cm) at site A and B while there were slight decrease in mean total PAHs from the layer 1 and layer 2 (Figure 6). With the exception of site B, all remaining sites showed increase of mean total PAHs concentration from layer 3 (6 - 8 cm) down to layer 5 (8 - 10 cm).

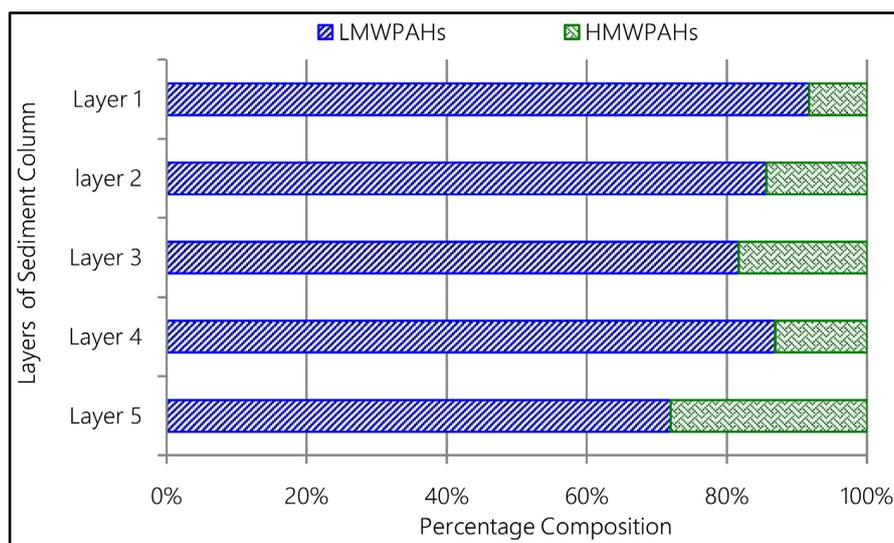
Overall distribution of mean total concentrations indicated that layer 2 accumulated highest concentration compared to other layers. The layer contributed to 42.63% of all measured pollutants in the area, which was higher than the other layers. The layer 2 was followed by far by layer 5 which accounted for 22.82% of the total overall residues. Other layers 1, 3 and 4 contributed 5.17%, 8.49% and 20.89% of the total PAHs, respectively. Thus the vertical general trends of the residues is layer 2 > layer 5 > layer 4 > layer 3 > layer 1.

In this study the surface sediment from layer 1 (0 - 2 cm) had the lowest concentrations likely because the layer is constantly washed by tidal water. The highest concentration in sediments of layer two is an indication that the pollutants are the result of recent release and are in their way down the sediment.

Despite the variation, all layers were dominated by LMWPAHs. The dominance of LMWPAHs is attributed with the detection of high levels of phenanthrene in all sediment layers except one (lower most sampled layer at site C). Moreover other LMWPAHs were also measured in most sediment layer. In contrary, HMWPAHs such as Benzo(a)pyrene, Chrysene and Benzo(a)pyrene were only detected at low concentration in 45% (n = 9) of the sampled layers (Figure 7).



**Figure 6.** Vertical profile of Mean total concentration of PAHs (ng/g).



**Figure 7.** Percentage layers composition of PAHs.

In average composition, PAHs varied with no clear definite trend down the sediment column. Contribution of LMWPAHs gave the following trend layer 1 > layer 4 > layer 2 > layer 3 > layer 5 (Figure 6). Although there was no defined trend but the variations gave indication that surface top layer is highly dominated with LMWPAHs (91.70%) and their proportion decreased as you go down the column to 71.98% at down most layer 5. This findings support the idea that composition of HMWPAHs increases with depth [26]. On the other side, PAHs levels were reported to contribute considerably to the accumulation of PAHs in marine sediments. For example variations of PAHs have been observed in different environments.

Also, the levels of PAHs from this study were lower than those reported in east coast of Malaysia [27] but relatively comparable to those measured in sediments from marine sediment in Thailand [28]. Moreover, the concentrations of total PAHs residues measured in sediments in this study were on average, lower

than those found in sediments from Singapore [29]. Additionally, the level of PAHs from this study were comparable to elsewhere around the world as the study reported in sediment of Day bay, China [30] and lower than those reported in Shenzhen river [31].

In terms of molecular weight and ring size PAHs, the results have shown significant difference between two groups, of the LMW to HMW-PAHs. Phenanthrene was dominant as a LMW and 3-ringed PAHs. The findings of this study correspond to study of PAHs conducted at Estero de Urias that found the higher levels of Phenanthrene followed by pyrene in sediments [32]. The influence of 3 and 4-rings PAHs was also observed in sediment samples collected from Sanerancisco Bay [33] and in sediment from Mumbai harbor in India [34]. Although direct comparison of data on levels of PAHs on sediments between different studies is difficult because of differences in laboratories and methods of analysis, the comparison can at least give an indication of level of contamination of the study areas.

The PAHs levels in this study are also relatively comparable to marine coastal areas of Zanzibar Municipality whereby there are more diffuse sources such as runoff and washings, and boat activities. But the levels in this study are lower than those reported in crabs from different coastal areas of Zanzibar. Generally, the results of this study show that the total maximum concentrations of the PAHs are comparatively lower than those reported for sediments from other countries. For instance, Langat Estuary, Malaysia reported the PAHs up to 2480 ng/g [20] while Egypt Mediterranean Sea reported the maximum of 22,600 ng/g [35]. However, Singapore, reported the maximum PAHs of 82,410 n/g [29]. Other maximum values reported from different coastal areas are indicated in **Table 3**.

**Table 3.** Maximum total PAHs (ng·g<sup>-1</sup> dw.) in sediments from various marine sites in the world.

Locations	Concentration	Reference
Langat Estuary—Malaysia	2480	[20]
East coast of Malaysia	590	[27]
Singapore	82,410	[29]
Egypt Mediterranean sea	22,600	[35]
Naples harbor, southern Italy	31,774	[36]
Gulf of Fos area, France, Mediterranean sea	2700	[37]
Italian marine protected areas	1550	[38]
Xiamen Sea, China	1177	[39]
Jakarta Bay, Indonesia	77,700	[40]
Wesha coastal area, Pemba Island Tanzania	158.38	<i>This study</i>

## 4. Conclusions and Recommendations

The 8 PAHs out of the 16 United State priority pollutants were measured in sediment with total concentration of 158.38 ng/g (dw). In general, LMWPAHs dominated the HMWPAHs signaling the possibility of petrogenic sources. The presence of 2-ring and 3-ring PAHs in the surface soils can suggest that the more PAHs come from recent deposition events. However, the presence of HMPAHs also indicated the long-term contamination within the study area. Interns of sediment depth, PAHs pollution trend was random (rise and fall) may be due to periodic input of oil in the area. Phenanthrene residues (lower molecular weight) were dominant followed by pyrene (higher molecular weight PAHs) accounting for 90% and 40% detection frequencies, respectively. The results indicate that Weshu coastal area is contaminated with PAHs. Generally, the coastal area under this study is less polluted with respect to PAHs pollutants compared to the similar coastal areas reported in other countries. However, there is possibility of posing adverse effect on marine organisms and health effect of Weshu village dwellers and surrounding areas depending on marine foods as the part of their daily necessities. Based on the critical situation experienced in Weshu coastal environment, the study comes with the following conclusions and recommendations:

- 1) This study provides important data set on PAHs levels in the sediments along Weshu eastern peninsula, Pemba.
- 2) Levels of PAHs in sediment were several higher than the other related study reports from various countries.
- 3) PAH distribution profile indicated potential source dependence, as the levels were generally higher in the vicinity of emission inputs.
- 4) The results show that lower molecular weight PAHs were dominant.
- 5) Close monitoring of PAHs contamination around this area remains important.
- 6) Further work is needed not only to assess the spatial distribution of PAHs in sediment but also to examine temporal variation with more intensive sampling.
- 7) Remediation of contaminated area should be introduced by planting new mangroves.
- 8) Establish an extensive awareness campaign on the adverse effect of pollution on the environment and the populace.

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## Declaration of Competing Interest

The authors declare no conflicts of interest.

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