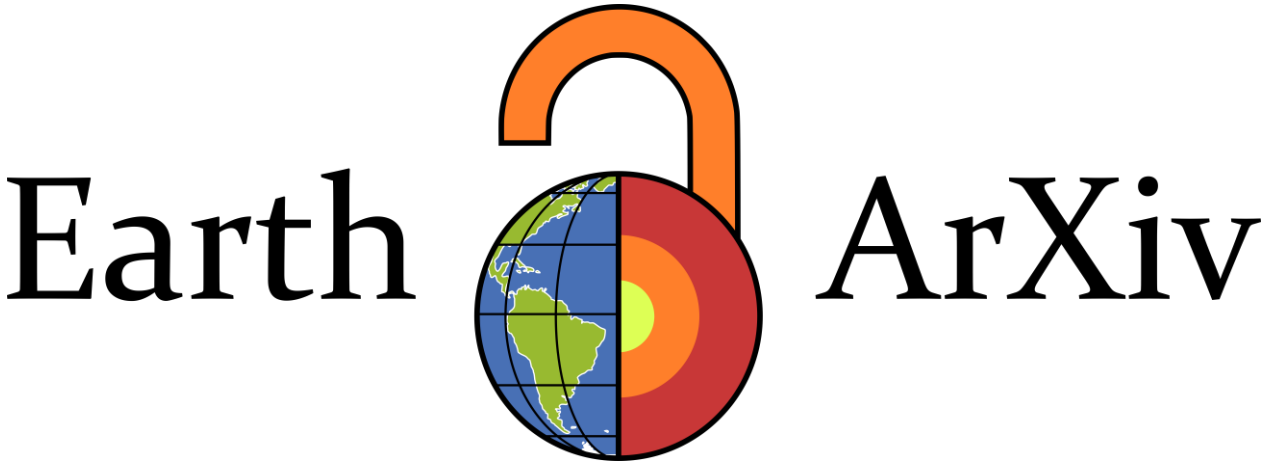

Assessment of heavy metal contamination in seawater, sediments, and fish tissues from Msimbazi Bay, Dar es Salaam, Tanzania.

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Abstract

Heavy metal pollution in marine environments is a growing concern globally due to its potential threats to aquatic ecosystems and human health. Major industrial zones in Tanzania, including Dar es Salaam city, significantly contribute to heavy metal pollution in aquatic ecosystems.

The samples for heavy metal analysis in seawater, sediments, and fish tissues were collected, prepared, and digested accordingly, and then analysed using an Atomic Absorption Spectrophotometer. The obtained results showed that marine sediments had higher levels of zinc (105.16 ± 11.83 mg/kg) at the sampling station in the Mangrove creek compared to other sampling stations in the bay, and their concentration levels were found to be in the order of Zn > Pb > Cr > Cd. In the seawater, Pb and Zn were detected within the range considered as natural in the ocean and their concentration were 1.84 and 19.35 µg/l, respectively. The liver of the fish was found to accumulate the highest concentrations of Pb (0.682 ± 0.25) µg g^{-1} and Cr (0.2 ± 0.09) µg g^{-1} compared to the kidney, gills, and muscle parts ($p > 0.05$). Overall, the concentrations in all compartments were below the permissible levels proposed by USEPA, ANZECC and ARMCANZ, FAO and WHO. Despite the study findings on metal levels, there is still a need to implement mechanisms to monitor the contamination of heavy metals in the Msimbazi catchment area and its Bay

Keywords: Heavy metal contamination, Bioconcentration, Marine pollution, Biomagnification, Msimbazi Bay.

INTRODUCTION

Heavy metal pollution has become a worldwide challenge that can arise from natural processes, such as the chemical and mechanical weathering of rocks, and from anthropogenic inputs, including industrial activities **(Robinson *et al*., 2001)**. In Tanzania, the waters of the Msimbazi River are reported to be contaminated with heavy metals derived from industrial effluents, agricultural chemicals, domestic waste disposal, and other types of discharges **(Ak'habuhaya and Lodenius, 1988)**. Studies by **De Wolf *et al.* (2001)** and **Ak'habuhaya and Lodenius (1988)** reported that most of the industrial and other wastes, supposedly containing heavy metal contaminants, discharged into the Msimbazi River are retained in mangrove stands found near the mouth of the river prior to the bay. Moreover, studies conducted on coastal sediments from Dar es Salaam harbour revealed the presence of high levels of heavy metals **(Machiwa, 1992, Muzuka, 2008, Mremi and Machiwa, 2003, Mohammed, 2000)** thus raising the possibility of heavy metal contamination in marine fish. In natural environments, the concentrations and distribution of heavy metals depend on organic content, sediment texture, and water parameters such as pH, salinity, and water temperature **(Mounier *et al*., 2001)**. These parameters control the availability of metals for uptake by living organisms and may also influence the toxicity of metals through processes such as the methylation of elements **(Mason *et al*., 1994)**.

Msimbazi Bay is well-known for the huge amounts of pollutants discharged into it from the Msimbazi River. To date, most studies including those of **De Wolf *et al.* (2001)**, **Ak'habuhaya and Lodenius (1988), Machiwa (1992), Rumisha *et al.,* (2012) and Mrutu *et al*. (2013)** have studied heavy metal pollution in the mangrove creek sediments, plants and potential bio-indicators like crustaceans and bivalves around Msimbazi mangrove creek. No studies have been conducted on this subject in the receiving bay's ecosystem, specifically regarding the levels of heavy metal contamination in seawater and marine fish species, despite the increased levels of heavy metals on the site. Additionally, there is a lack of clear information on the extent and distribution patterns of these pollutants within the Bay. Therefore, this study aimed to investigate the concentration levels of cadmium (Cd), lead (Pb), zinc (Zn), and chromium (Cr) in marine sediments, seawater, and fish (*Lethrinus harak*) in Msimbazi Bay, on the Dar es Salaam coast.

MATERIALS AND METHODS

Study Area

This study was conducted in Msimbazi Bay, located in the Western Indian Ocean, along the coast of Dar es Salaam, Tanzania (Figure 1). Samples of seawater, sediment and *Lethrinus harak* were collected for three months from November 2014 to January 2015 around Msimbazi Bay.

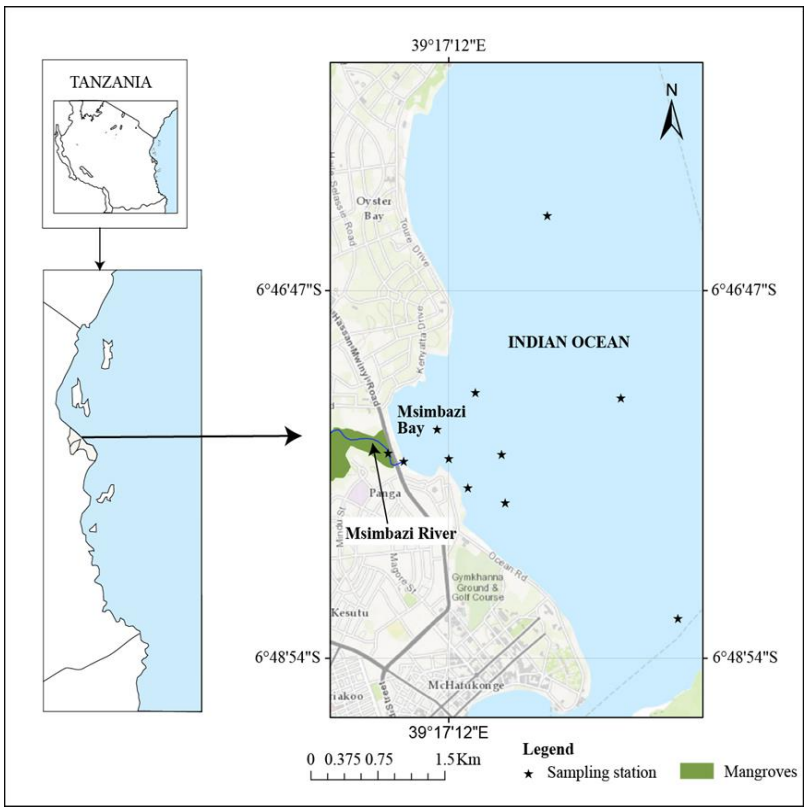

Figure 1: *A Map Showing the Msimbazi Bay (Source: Institute of Resource Assessment, GIS Laboratory – University of Dar es Salaam)*

Heavy Metals Analysis in Sediment Samples

Homogenized samples about 0.5 g each were put in pre-acid-washed beakers, and about 10 mL aqua regia (1:3, HNO3: HCl) were added. Then, the mixture was heated, and 1 mL of perchloric acid ($HClO_4$) was added to the solution. The samples were then heated until they became clear. Afterwards, the solutions were diluted to 50 mL with deionised water, allowed to stand overnight, and then filtered through a 0.45 µm pore size Whatman No. 41 filter paper. The filtrates were analyzed for heavy metals using an atomic absorption spectrophotometer (Thermo Scientific iCE 3000, C113300183, v 1.30) **(Cunniff and Washington, 1995)**.

Heavy Metal Analysis in Seawater Samples

Seawater samples (500 mL) were filtered through 0.45 µm membrane filters and digested according to the standard method of the Association of Official Analytical Chemists, AOAC **(Cunniff and Washington, 1995)**. 10 mL of nitric acid (HNO_3) was added to a 100 mL of seawater sample in a beaker and heated at 150 °C for 30 minutes. HNO_3 (5 mL) was then added to each beaker containing the solution, and the mixture was heated for 30 minutes at 200 °C. After heating, it was left to cool. To the mixture, 2 mL of hydrogen peroxide (H_2O_2) was added, and the mixture was heated for 30

minutes. It was then diluted to 50 mL with distilled water **(Cunniff and Washington, 1995)**. The solutions were then analysed for heavy metals by atomic absorption spectrophotometer.

Heavy Metals Analysis in Fish Tissue Samples

A total of 43 fish samples of the species *Lethrinus harak* (Forsskål, 1775) were collected on site from fishermen fishing at Msimbazi Bay. In the laboratory, biometric measurements were taken, and then the fish was dissected to obtain its organs, including kidneys, livers, gills, and muscle tissues. The samples were oven-dried at a temperature of 105°C for 24 hours. After drying, the fish organs were homogenized and dried samples of 0.5 g were mixed with 10 mL of aqua regia (1:3, HNO_3: HCl) prior to heating. Then, 1 mL of perchloric acid ($HClO_4$) was added to the mixture before dilution to 50 mL with deionized water. The sample solutions were then allowed to stand overnight and subsequently filtered through a 0.45 µm pore size Whatman No. 41 filter paper **(Cunniff, 1995).** The filtrates were analysed for heavy metals by atomic absorption spectrophotometer.

Plate 1: *Sample Preservation at the Msimbazi Bay, Dar es Salaam (A), Field Sampling and Parameter Measurements (B), Preparation (C, D, and E), and Digestion of Samples (F).*

Determination of Bioaccumulation Factor (BAF) of Heavy Metals in Fish

The following equations described by **Barron (1995)** and **Demina *et al.* (2009)** were used for the determination of the bioaccumulation factor (BAF) with respect to sediments and seawater as media for metals bioaccumulation **(Barron, 1995, Demina *et al.*, 2009)**:

$$BCF = \frac{M_{Tissue}}{M_{Water}} \quad \text{.....(ii) and} \quad BSF = \frac{M_{Tissue}}{M_{Sediment}} \quad \text{.....(iii)}$$

Where M_{Tissue} is the metal concentration in tissues of fish, $M_{Sediment}$ is the metal concentration in sediment, BCF is the Bio-Concentration Factor and BSF is the Bio-Sedimentation Factor

Validation of Analytical Methodology

Blank solutions were prepared for each set of digestion processes, and recovery tests were conducted to determine the accuracy and validate the analytical method **(Al-Weher, 2008)**. The percentage concentration of the metal recovered was then calculated using the formula provided by **Al-weher (2008)**.

Statistical Analysis of Data

Descriptive statistics and a one-way analysis of variance (ANOVA) were employed to compare the differences in heavy metal concentrations between transects and sampling stations in fish tissues, sediments, and seawater. Friedman test and Bivariate correlation analysis were also used to determine the degree of relationships between variables.

RESULTS

Validation of Analytical Method

The recovery of metal concentrations in the blank samples for the analytical method used in this study ranges were Pb: 91 - 101%, Cd: 85 - 87%, Zn: 91 - 96% and Cr: 88 - 89%, while correlation coefficients in all cases were found to be above 0.9.

Concentration of Heavy Metals in Sediment

The concentrations of heavy metals (Pb, Cr, Cd, and Zn) in sediment samples from Msimbazi Bay are summarized in Table 1. Statistically, the concentrations of the heavy metals in sediments between transects at the Bay, indicated no significant difference ($p > 0.05$) in all metals; Zn (ANOVA, $df = 2$, $F = 0.552$, $p = 0.603$); Pb (ANOVA, $df = 2$, $F = 2.988$, $p = 0.126$) and Cr (ANOVA, $df = 2$, $F = 0.527$, $p = 0.615$). On the other hand, the concentration levels of heavy metals (Zn, Cd, Pb, and Cr) in sediments between all sampling stations at the mangrove creek (Sampling points OM and OS) and those at the Bay were significantly different (ANOVA, $p < 0.05$).

Table 1: *Concentrations (Mean ± Standard Deviation) of Heavy Metals (mg/kg dry weight) in Sediment Samples (n = 33).*

Transect	Sampling points	Heavy Metals (mg/kg-dry weight)			
		Zn	**Pb**	**Cr**	**Cd**
Source	OM	105.16 ± 11.83	43.29 ± 4.18	23.41 ± 5.83	0.93 ± 0.12
	OS	57.61 ± 19.98	26.15 ± 5.21	7.8 ± 5.41	0.63 ± 0.11
Southeasterly direction (SE)	A1	45.53 ± 16.59	12.17 ± 4.54	1.56 ± 0.39	BDL
	A2	30.16 ± 8.81	11.06 ± 6.75	0.91 ± 0.55	BDL
	A3	35.21 ± 7.25	4.62 ± 1.54	0.53 ± 0.63	BDL
Central (Easterly) direction (E)	B1	38.25 ± 10.12	8.16 ± 1.3	1.41 ± 0.16	BDL
	B2	29.06 ± 2.72	9.64 ± 3.14	1.00 ± 0.21	BDL
	B3	26.74 ± 1.42	6.84 ± 2.61	BDL	BDL
Northeasterly direction (NE)	C1	41.11 ± 7.25	14.74 ± 0.99	1.32 ± 0.7	BDL
	C2	25.17 ± 7.46	10.64 ± 4.62	BDL	BDL
	C3	27.72 ± 2.28	21.29 ± 2.44	BDL	BDL

Note: BDL means Below Detection Limit

3.2.1 Variation of Heavy Metal Concentrations in Sediment with Distance from the River Mouth

Comparison of the distribution of heavy metals between transects and with increasing distance from the river mouth (100, 500 to 1500 m) is shown in figure 2.

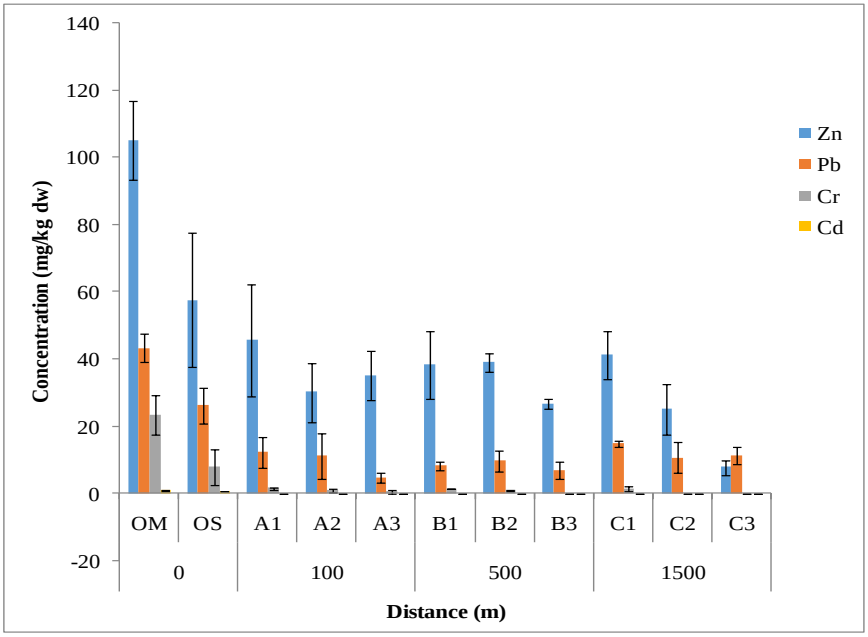

Figure 2: *Variation of Heavy Metal Concentration with Increasing Distance from the River Mouth*

Concentrations of Heavy Metals in Seawater

Zinc (Zn) was found in all sampling stations at different concentrations while Cr and Cd detection were below limits. Statistically, the mean concentrations of metals in seawater at the Bay between transects were significantly different $p < 0.05$; Zn (ANOVA, df = 2, F = 10.682, p = 0.011) and Pb (ANOVA, df = 2, F = 8.522, p = 0.018). On the other hand, concentration levels of Zn and Pb between sampling stations at the Bay and those at the mangrove creek were significantly different $p < 0.05$; Zn (ANOVA, df = 3, F = 28.181, p > 0.001) and Pb (ANOVA, df = 3, F = 13.355, p = 0.003).

Concentrations of Heavy Metals in Fish Tissues

The concentrations of Pb, Cr, Cd, and Zn in the muscle tissues, kidneys, livers, and gills of the analyzed fish species are presented in Figure 3. However, concentrations of Zn (0.79 µg g^{-1}), Cr (0.1 µg g^{-1}), Pb (0.18 µg g^{-1}) and Cd (0.05 µg g^{-1}) in the fish specimens were lower than the permissible levels recommended by FAO and WHO **(FAO, 1983, Organization, 2023)**.

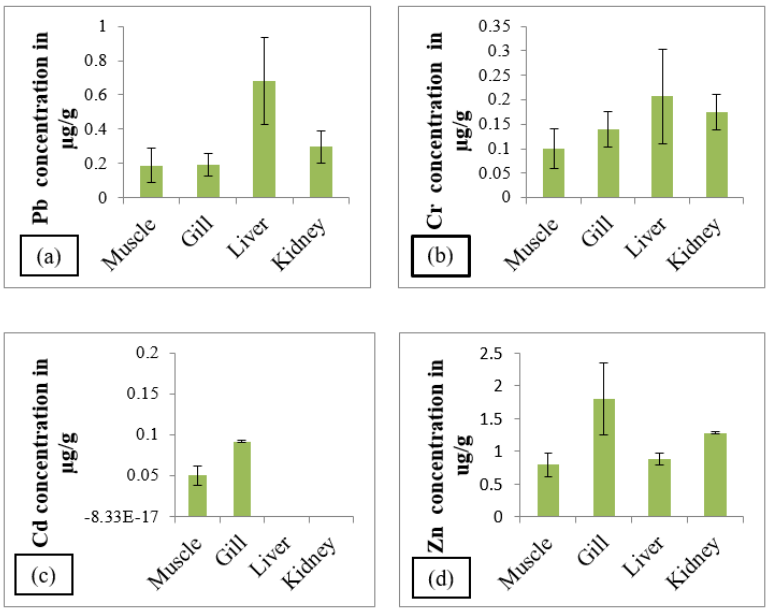

Figure 3: *Concentration (Mean ± SD) of (a) Pb (b) Cr (c) Cd and (d) Zn (µg g^{-1}) in Fish tissues*

Bioaccumulation Factor of Heavy Metals in Fish Tissues

The highest bio-accumulation factors for metal Zn (BCF = 0.35) was found from gills - water and was greater than that from sediments (BSF = 0.04). Lead (Pb) exhibited the highest bioaccumulation factor in the liver (BCF = 0.75), which was greater than that from sediments (BSF = 0.04). The highest bio-sedimentation factor for metal Cd (BSF = 0.7) was found in the muscle part, while metal Cr (BSF = 0.06) was found in the liver.

DISCUSSION

According to the Interim Sediment Quality Guidelines (ISQGs) of Australia and New Zealand Sediment Quality guidelines **(ANZECC and ARMCANZ, 2000)** the levels of heavy metals in this study area were within range of acceptable limit for environmental values of a healthy ecosystem except for Pb (43.29 ± 4.19 mg/kg) at station OM (mangrove creek) which had higher values than those set by ISQGs. The higher value of Pb in the sediment of station OM probably exposes marine organisms feeding on bottom sediments to high levels of Pb **(De Wolf *et al*., 2001, Sany *et al*., 2011)**, which they can potentially transfer to higher trophic levels. Generally, the concentration levels of Zn, Pb and Cr showed a decreasing trend from the river mouth to offshore areas. This could be due to several factors, such as differences in the chemical composition of sediments, organic matter content, and sediment texture along the distance to the offshore area **(Kishe and Machiwa, 2003, Nyandwi, 2001)**. Higher concentrations of Pb at Station OM are likely due to the discharge of waste containing Pb into the bay, which originates from activities such as the use of leaded petrol and car batteries, as well as other industries that utilize this metal and similar reasons were given by **Ak'habuhaya and Lodenius (1988)** and **Mohamed (2002)**. Chromium concentration was significantly correlated with Zn ($r = 0.977$) at 95% confidence level. This association of Cr with Zn suggests a common source to the receiving Bay of Msimbazi **(Preda and Cox, 2005, Muzuka, 2008)**.

The concentrations of heavy metals in seawater at the receiving Bay may be attributed to various sources found in the Msimbazi River Basin, which is characterized by four small and seasonal tributaries that collect anthropogenic inputs, including those from industries, domestic, and agricultural sources **(Machiwa, 2010)**. **Ak'habuhaya and Lodenius (1988)** reported that power stations, breweries, textile mills, dump sites, and the central hospital were significant sources of metal pollution in the basin, from which the effluents are eventually released into the seawater.

Generally, the concentration of heavy metals studied followed the trend Zn > Pb > Cr = Cd from the mangrove creek to offshore stations. Although concentrations of Pb and Zn in this study were the highest but they were lower than USEPA seawater guideline levels of 8.1 $\mu g\ l^{-1}$ for Pb and 81.0 $\mu g\ l^{-1}$ for Zn and also lower than the Malaysia Marine Water Quality Criteria (IMWQS) of 8.5 $\mu g\ l^{-1}$ and 50 $\mu g\ l^{-1}$ respectively **(BOBLME, 2011)**. The two sampling stations (OM and OS) had the highest concentrations of all the metals examined in the study, probably due to the presence of mangrove creek at the river mouth, which acts as a filter for most pollutants **(Mrutu *et al*., 2013)**. Additionally, the creek receives wastewater from the Msimbazi River, whose tributaries include the Msimbazi, Sinza, Luhanga, and Ubungo Rivers. These tributaries collect waste discharges from different sources, including industrial, domestic and agricultural sources **(Rumisha *et al*., 2012, Ak'habuhaya and Lodenius, 1988). Muzuka and Shaghude (2000)** and **Nyandwi (2001)** indicated that the coastal

currents in the study area flow in a northward direction; thus, the above-mentioned currents could be responsible for the distribution of these metals around the Msimbazi Bay.

It is well known that fish muscles are not active sites for the biotransformation and accumulation of heavy metals **(Elnabris *et al*., 2013)**. However, in coastal areas with increasing pollution rates over time, such as Msimbazi Bay, bioaccumulation levels in fish muscles may exceed the permissible limits for human consumption. When comparing the results from this study with those of others, the concentration values in muscle tissues were significantly lower **(Oze *et al*., 2006, Saeed and Shaker, 2008)**. These variations may be attributed to the amount of pollutants that enter the receiving bodies and differences between fish habitats. The difference in the accumulation of heavy metals in liver tissues compared to muscle tissues could be attributed to differences in their physiological roles. This observation is supported by other studies that reported that muscles have low metabolic activities and are not active organs in accumulating various heavy metals **(Uluturhan and Kucuksezgin, 2007)**. The gills accumulated a high content of Zn and Cd compared to other organs of the fish studied in this work, which could be attributed to the presence of metallothionein in the gills, influencing the bioaccumulation and uptake of these metals **(Saeed, 2000)**. Moreover, the accumulation of heavy metals in fish organs differs as some metals are organ-specific, and their distribution within the organism changes due to the different affinities of various metals to tissues **(Jezierska and Witeska, 2006)**.

CONCLUSION

Levels of heavy metals in both sediments and seawater exhibited a decreasing concentration trend with increasing distance from the mangrove creek towards the offshore area, following the order of Zn > Pb > Cr > Cd. Comparatively, the concentrations of heavy metals in sediments were higher than those in seawater, implying that sediments act as a sink for heavy metals in the Msimbazi Bay. Generally, the concentrations of heavy metals in both compartments (sediment, seawater and fish) were below the permissible levels proposed by the ANZECC and ARMCANZ, as well as the USEPA, FAO, and WHO. Therefore, it can be inferred from this study that, over time, the levels of heavy metals may elevate, making Msimbazi Bay less safe for human activities. Hence this environmental contamination necessitates continuous monitoring of pollution and engage control measures in the Dar es Salaam city.

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