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Research Article

Lead and Cadmium Levels in Water, Surficial Sediments, and Edible Biota of Urban Rivers in Dar es Salaam, Tanzania, During Two Seasons

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Abstract: This study evaluated levels and seasonal patterns of lead (Pb) and cadmium (Cd) in sediment, water, and mudskippers (*Periophthalmus argentilineatus*) from urban rivers in Dar es Salaam, Tanzania. The samples were seasonally collected from 15 locations, treated as appropriate, and analyzed instrumentally. The study intended to establish contamination levels, identify hotspots, and assess ecological and human health risks. The results indicated that Cd concentrations were lower than Pb in all the sample matrices. In water samples, Pb levels (0.7 to 24.0 mg/L, n = 30, mean = 7.2 ± 6.5) were higher than both the World Health Organization (WHO) and the Tanzanian Bureau of Standards limits, whereas Cd was below the detection limit. In sediment samples, both the Pb concentrations (9.0 to 159.3 mg/kg, n = 30, mean = 44.7 ± 36.6) and the Cd concentrations (0.03 to 0.82 mg/kg, n = 30, mean = 0.24 ± 0.14) exceeded sediment quality guidelines for the protection of aquatic life. The mudskipper samples contained levels below the Food and Agriculture Organization (FAO) and WHO maximum permissible limits. This indicated that they were safe for human consumption. Metal concentrations in water and sediments were lower during the dry season than in the wet season, probably owing to contaminated runoff from land-use practices during the wet season, whereas the mudskipper samples showed the opposite pattern. Determination of the potential contamination index (PCI) in samples enabled the identification of two hotspots that may need intervention. Evaluation of chronic daily intake (CDI), hazard quotient (HQ), and hazard index (HI) implied minimal health risks. Contamination of the rivers by these toxic metals, be it in low concentrations, is an issue of concern.

***Keywords*:** metal pollution, lead, cadmium, risk assessment, urban rivers, Tanzanian coast

1. Introduction

Lead (Pb) and cadmium (Cd) are among the groups of elements that have no recognized biological function in living organisms and are established to be toxic even at low levels [1]. These metals are broadly spread in the aquatic environment, typically from human activities, although as components of the earth's crust, they may also occur naturally at very low concentrations [2]. Anthropogenic activities such as transportation, solid waste disposal, wastewater discharge, mining, metal smelting, and agriculture are generally believed to be the main contributors to environmental contamination by these metals [3, 4]. For example, Pb is easily smelted and has been used by humans for thousands of years; as a result, it is regarded as a long-standing environmental contaminant [5]. Until recently, leaded gasoline

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was a major source of Pb in the environment, but this is now greatly reduced due to the ban of tetraethyl-Pb as a fuel additive for almost two decades now [6]. Other uses of Pb include storage batteries, casting materials, and various alloys. Many environmentally significant compounds of Pb, such as phosphates, halides, sulfates, and hydroxides, are insoluble in water and therefore have relatively low toxicity in aquatic systems [5]. Pb behavior in natural ecosystems has been extensively studied and a broad range of ecotoxicological data exists [7-11]. The compounds of Cd have many uses in a variety of industrial operations and products, causing a high possibility of being directly discharged into the environment. It is used in electroplating to provide a bright appearance and resistance to corrosion in different products. It is also applied to ceramics, plastics, pigments, paints, and batteries in different types of consumer items [12]. The application of phosphate fertilizers is also one of the most important sources of Cd in aquatic systems [13]. When present at high enough levels, Pb and Cd may induce changes in the nutrient dynamics of ecosystems and subsequently cause toxic effects on sensitive aquatic species [14, 15]. Due to their high degree of toxicity, Pb and Cd are included in the list of priority metals with significance to public health [16].

Metal pollution in aquatic environments is a well-researched field in Tanzania, although many locations remain to be investigated. Over a period of more than two decades, authors have used various environmental and biological matrices, including sediments, water, soil, plants, and biota, to measure concentrations of metals in various locations in the country [17-22]. Because of their toxicity, persistence, and non-degradability, the elements Pb, copper (Cu), Cd, chromium (Cr), mercury (Hg), arsenic (As), cobalt (Co), nickel (Ni), and zinc (Zn) were the most commonly researched. Some of the studies reported large amounts of Pb and Cd in various locations [23-26]. This has increasingly become a serious environmental concern, particularly due to industrialization and urbanization prospects. Dar es Salaam City is a dynamic commercial and tourist location where each year hundreds of new businesses are opened and thousands of people are visiting. Studies on levels and dispersions of metal pollutants in surface waters around the city have been reported in the past. Nevertheless, there is a need to regularly monitor the environmental quality and get up-to-date insights into the contamination status of the rivers in this area. The objective of the present study is therefore to meet this information need by evaluating contamination levels, seasonal patterns, and health risks of the two metals in rivers flowing through the urban locations of Dar es Salaam City. To achieve this, concentrations of the two trace metals were quantified in surface waters, surficial sediments, and mudskipper samples in two distinct seasons. The data set was subjected to various statistical analyses, environmental quality guidelines, contamination indices, and hazard quotients to establish the quality of the river ecosystems.

2. Materials and methods

2.1 *Study area*

Tanzania has abundant surface water resources, including hundreds of rivers and streams, some in urban and some in rural locations. Situated in East Africa (1.8 - 12.8º S and 29.8 - 41.8º E), the country is bordered by Mozambique to the south and Kenya to the north. About 94% of the total area (886,000 km^2) is dry land, including the coastal islands of Zanzibar, Pemba, and Mafia. The country is estimated to have a coastline of about 800 km long and surface water that occupies an area of about 59,000 km^2. The study area is Dar es Salaam, the business capital and major city in the country. The city, located at 6° 48′ S and 39° 17′ E, occupies approximately 1,800 km^2 of area, with about 1,400 km^2 being landmass [27]. The current population of Tanzania is 61,741,120, whereas Dar es Salaam City has a population of 5,383,728 [28]. The port of Dar es Salaam is a significant harbor in the region. The city experiences climatic conditions typical of the tropics throughout the year, characterized by humid and hot weather. The months of July and August have relatively low precipitation (up to an average of 47 mm), whereas December and January precipitation reach 195 mm, making an average of 1,100 mm [29]. The area experiences heavy rains from March to May and dry weather in the remaining months. Precipitation rates from the previous period usually control the river flow around Dar es Salaam, and they are usually as low as a baseline flow of 1 m^3/s or lower during the dry season and 15 m^3/s or higher during the rainy season [29].

Three rivers, the Mbezi, Kizinga, and Msimbazi, were involved in the study. These rivers were selected because of the various anthropogenic activities taking place around them, which include unplanned settlements, urban vegetable farming, and domestic activities. The Mbezi River (approximately 24 km long) flows into the city through the relatively less densely populated locations of Mbezi Beach and Kawe and later drains into the Indian Ocean. Through its course, this river passes a lot of human settlements and fewer industries that generate a considerable amount of waste. The Kizinga River (approximately 30 km long) passes along the relatively urbanized localities of Mtongani, Keko, Mbagala, Chang'ombe, Buza, and Kurasini before passing the mangroves of Mtoni and discharging into the ocean. Throughout its course, it is presumed to carry a lot of waste from residential, industrial, and agricultural activities [30]. The Msimbazi River (approximately 36 km long) flows from the Kisarawe hills into the Indian Ocean after collecting residential and industrial (building materials, dye, detergents and soap, textiles, and breweries) wastes from densely populated and industrialized areas [31].

2.2 *Sample collection and preparation*

Samples for Pb and Cd analysis were collected during the wet season (March to April 2018) and during the dry season (August to September 2018) in 15 locations (Table 1). Water, sediment, and biota samples were also collected simultaneously from these locations, five on each river. During both seasons, sampling was done in the same locations, which were marked by a handheld global positioning system (GPS). Water samples were collected in clean, pre-labeled, Teflon-capped, one-liter plastic bottles. The bottles were vigorously rinsed with river water twice at each station before being filled with the samples, which were then acidified (pH < 2.0) using concentrated nitric acid (HNO_3; 5 mL/L). The sample bottles were kept in an icebox at < 10 °C and then transported to the Analytical Chemistry Laboratory, Chemistry Department, University of Dar es Salaam, for laboratory analysis.

Sediment samples were collected using a hand corer, as described by Mihale et al. [21]. Duplicate sediment samples (approximately 1 kg) were collected, appropriately packed, labeled, and stored in an icebox. The samples were frozen to -20 °C before being transported to the Department of Analytical, Environmental and Geo-Chemistry Laboratory, Vrije Universiteit Brussel (VUB), Belgium. Before analysis, lyophilization, crushing, grinding and sieving of the samples were performed, and the samples were later kept in clean vials.

The biota used for this study was mudskippers (*Periophthalmus argentilineatus*). These edible fish species are abundant along the Dar es Salaam coast and are found in fresh, marine, and brackish waters. The fish were trapped using fishing nets with the aid of local fishermen at the locations. The samples were then rinsed with water at the station, wrapped in aluminum foil, and packaged in labeled and sealed plastic bags in an icebox at < 10 °C. The sediment samples were then deep-frozen and transported to the VUB for further processing and instrumental analyses.

Table 1. Locations of the sampling sites

River	Site code	GPS coordinate		Elevation (meter)
Kizinga	Kz_1	06° 53.060' S	039° 15.853' E	8
	Kz_2	06° 52.987' S	039° 16.176' E	6
	Kz_3	06° 52.903' S	039° 16.521' E	8
	Kz_4	06° 52.439' S	039° 17.054' E	4
	Kz_5	06° 52.107' S	039° 17.485' E	4
Msimbazi	Ms_1	06° 49.125' S	039° 15.318' E	10
	Ms_2	06° 48.943' S	039° 15.764' E	12
	Ms_3	06° 48.592' S	039° 15.921' E	7
	Ms_4	06° 48.236' S	039° 16.171' E	11
	Ms_5	06° 47.775' S	039° 16.849' E	3
Mbezi	Mz_1	06° 4.098' S	039° 1.020' E	25
	Mz_2	06° 42.728' S	039° 13.689' E	8
	Mz_3	06° 42.715' S	039° 13.901' E	6
	Mz_4	06°42.696' S	039° 13.967' E	6
	Mz_5	06° 42.592' S	039° 13.984' E	7

2.3 *Sample analysis*

Water samples were analyzed by flame atomic absorption spectrometry (FAAS; iCE 3000 Series, Thermo Fisher Scientific) using standard method 3111A [32]. Instrumental calibration was done using working standards of the two elements at varying concentrations, prepared in 5% (v/v) hydrochloric acid (HCl). The calibration curves that aided in the estimation of the concentration of the intended analyte were made for each element. Pb was determined at 216.9 nm wavelength, whereas for Cd it was 228.7 nm. The sediment samples were digested in a microwave (Multiwave Go Plus Microwave Digestion System, Anton Paar) at 180 °C for 25 minutes using aqua regia (6 mL of distilled HCl and 2 mL of distilled HNO_3). Later, the sample (0.5 mL) was diluted with milli-Q water (4.5 mL) to make the final 5 mL solution for analysis. The analysis was performed using a high-resolution inductively coupled plasma mass spectrometer (HR-ICP-MS; Thermo Finnigan Element II). Standard solutions of each metal were prepared from stock standard solutions obtained from the ICP multielement standard solution (Merck) by serial dilution. The frozen fish samples were dried through the lyophilization method and ground with a mortar and pestle. Sub-samples weighing approximately 0.2 g were then processed with 6 mL of a concentrated distilled mixture of HNO_3 and H_2O_2 (5:1) in the same microwave digestor that was used for the sediment samples. The digested fish samples were analyzed instrumentally in the same manner as the sediment samples.

2.4 *Quality control*

The quality control of the analytical procedure was ensured throughout the study, from sample collection, transportation, preparation, instrumental analysis, and finally the data checks. All tools were carefully washed and rinsed with distilled water, and the instruments were calibrated using prescribed protocols by the instruments' manufacturers. Distilled water was used as blank water samples, whereas distilled water spiked with standard solutions of Pb and Cd at their method detection limits (BDL) was used for the water recovery tests. The river clay sediment (sample number 0261, batch 001, Government Chemist, United Kingdom) was used as the certified reference material (CRM) for the sediment samples. Similarly, a sample of fish protein (DORM-4, National Research Council of Canada) was used as the CRM for fish samples. All reference samples were processed and analyzed in the same manner as ordinary samples. Indium was used as the internal standard at 1 μg/L. Standard working concentrations were prepared at 1, 5, 10, and 20 ppm and analyzed before and after each batch that constituted ten samples. The analyses were carried out in duplicate and run twice. The percentage recoveries of both Pb and Cd were > 90%; therefore, the final results were not corrected. Before use in the digestion step, the aqua regia was analyzed as a reagent blank. The blanks had no traces of the analytes; therefore, no blank correction was made. The procedural blanks were used to obtain the average signal from which the standard deviation of the blank was computed. The standard deviation multiplied by three was used as the limit of detection (LOD) [33].

2.5 *Data analysis*

Microsoft Excel was used to summarize and present the results using descriptive statistics (means, ranges, and standard deviations). The Shapiro-Wilk test was used to examine the normality of the datasets, in which a variable was accepted as normally distributed if $p > 0.05$. Spearman's rank test was used to correlate the various datasets to evaluate the magnitude and direction of their associations. The Friedman test was used to compare the levels of the measured parameters in the three rivers. The Wilcoxon signed ranks test and the paired samples t-test were used to evaluate the spatial and seasonal variation trends of the metal concentrations. The test results were considered significant at the 95% confidence level (CL) and $p > 0.01$. All the statistical analyses were done using the Statistical Package for the Social Sciences (SPSS) version 23.0 from the International Business Machines Corporation (IBM).

2.6 *Ecological risk assessment*

The ecological risk of contamination by the two metals was further scrutinized by the evaluation of the potential contamination index (PCI). The PCI was determined using Equation 1 [34].

$$\mathrm{PCI_i} = \frac{c_{i(max)}}{c_{(Background)}} \tag{1}$$

where $C_{i(max)}$ is the maximum concentration of metal in the sediment at a particular location and $C_{(Background)}$ is the background concentration of the metal.

In this study, the general abundances of metals in the upper crust of the earth [2] were used as estimations of the background concentrations of the two metals. This was because the specific background values for the locations have yet to be established. The interpretation of the PCI values according to the method is such that PCI < 1 is indicative of low contamination; 1 < PCI < 3 is indicative of moderate contamination; whereas PCI > 3 is indicative of severe contamination [34]. Evaluation of contamination status using sediment quality indices has been used by other researchers to evaluate the ecological risks of metals in various settings [35-38].

2.7 *Health risk assessment*

The evaluations of the health risks of both metals in the analyzed sample matrices were done using the model described by the United States Environmental Protection Agency (USEPA) [39], where chronic, non-carcinogenic, or carcinogenic risks were determined. The chronic daily exposure to metals through dermal absorption as well as ingestion was computed according to the description by De Miguel et al. [40] using Equations 2 and 3, respectively.

$$\text{Ingestion intake (IngI) per day (mg/kg)} = \frac{\mathrm{C \times CF \times EF \times ED \times FI \times IR}}{\mathrm{AT \times BW}} \tag{2}$$

$$\text{Dermal intake (DI) per day (mg/kg)} = \frac{\mathrm{C \times CF \times AF \times EF \times ED \times ABS \times SA}}{\mathrm{AT \times BW}} \tag{3}$$

Table 2 shows the abbreviation and values employed for the determination of the chronic daily intake (CDI).

Table 2. Exposure values used in the determination of CDI [41-43]

Exposure factor	Description	Value
C	Metal concentration (mg/kg)	-
CF	Conversion factor (kg/mg)	$\times 10^{-6}$
ED	Exposure duration (years)	30
EF	Exposure frequency (events or days/year)	350
FI	Fraction of ingested amount	0.05
IR	Ingestion rate (mg/day)	100
AF	Adherence factor (mg/cm^2)	0.07
ABS	Absorption factor	0.001
SA	Contact surface area (cm^2/event)	400
AT	Averaged exposure time of the metal (days)	25,550
BW	Body weight (kg)	70

The hazard quotient (HQ) of a given metal was computed as the ratio of CDI to the metal's chronic reference doses (CRD) to give the non-carcinogenic risk using Equation 4 [40].

$$HQ = \frac{CDI}{CRD} \tag{4}$$

Table 3 shows the CRD values for the two metals used in the calculation as adopted by De Miguel et al. [40].

Table 3. CRD of the analyzed metals

Metal	Reference dose (mg kg^{-1} day^{-1})	
	Ingestion	Dermal
Pb	3.5 x 10^{-3}	5.25 x 10^{-4}
Cd	1.0 x 10^{-3}	1.0 x 10^{-5}

The summation of all the HQ of every metal in the sample gave the non-carcinogenic hazard index (HI) as shown in Equation 5.

$$HI = \sum HQ = HQ_{Pb} + HQ_{Cd} \tag{5}$$

3. Results and discussion

3.1 *Occurrences and seasonal variations of metals in the water samples*

The results from analyses of the water samples revealed that only Pb was present in concentrations ranging between 0.7 and 24.0 µg/L during the wet season (mean = 9.0 ± 7.9 µg/L) and between 0.8 and 14.6 µg/L during the dry season (mean = 5.4 ± 4.2 µg/L), as illustrated in Figure 1. Cd concentrations were lower than the BDL in all of the water samples.

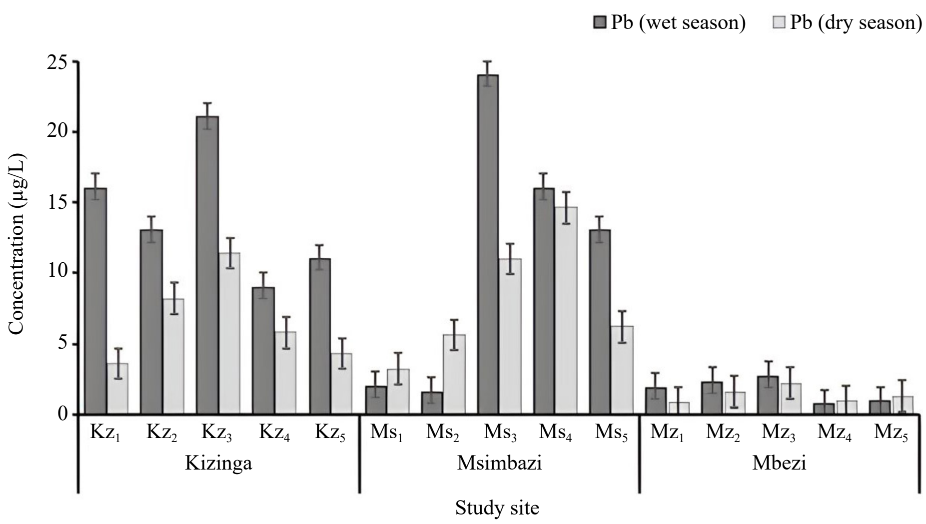

Figure 1. Concentrations of Pb in water samples from the 15 sites during the two seasons

Figure 1 shows that the highest level of Pb was detected at Ms_3, and this was during the wet season. This site is within the vicinity of a highly industrialized and populated locality along the Ilala Municipality. Other locations with relatively high concentrations of Pb during both seasons were Kz_3, Ms_4, and Kz_1. Visual observation of Figure 1 also shows that of the three rivers, the Mbezi River generally had the lowest Pb levels. This was statistically confirmed by the Friedman test. The test further revealed that the Kizinga River generally had the highest levels, followed by the Msimbazi River. The maximum permissible limit of Pb in potable water provided by the Tanzania Bureau of Standards (TBS) [44] and by the World Health Organization (WHO) [45] is 10.0 mg/L. This limit was exceeded at six of the 15 sampling locations, including three in the Msimbazi River and three in the Kizinga River. Low levels of Pb are expected in river waters due to natural sources such as the dissolution of rocks and soils. However, levels exceeding the stipulated limit are considered contamination from anthropogenic sources. Some of these sources may include the production and use of Pb-containing products [45]. Tanzania is implementing legislation restricting the use of leaded fuels [6], therefore its importance as one of the largest sources of Pb contamination is diminishing. A high concentration of Pb, such as those found in some stations in this study, is considered a health risk since Pb is toxic and has no health benefits. The levels of Pb and Cd contamination found in this study were typically lower than those that Othman [17] discovered 20 years ago in some urban areas of Dar es Salaam City. However, the study by Othman [17] investigated levels of Cd and Pb, among other metals, in water samples from different locations of the Msimbazi River catchment than those sampled in the current study. In the study, concentrations of both metals exceeded the limits set by the TBS and the WHO for potable water. The study indicated high impacts from contaminated runoff from industrial, domestic, and vegetable farming activities.

The data on the Pb concentrations were further subjected to the paired samples t-test to study their seasonal variation trends. The test output revealed that Pb concentrations in the water samples for the two seasons were strongly positively correlated, with a Spearman rank correlation coefficient ($r_s = 0.813$, at $p = 0.01$). It was also observed that there was a significant difference between the wet season and the dry season concentrations ($t_{14} = 2.756$, $p = 0.017$). t_{14} stands for the statistical test value (t-value) that compares the two groups of samples. The wet season mean was higher than the dry season mean by 3.62 mg/L (95% CL [0.80 to 6.45]). This is indicative of the influence of human activities as well as weather conditions. Agricultural runoffs, domestic discharges, and industrial effluents are higher during the rainy season compared to the dry season when the flow is reduced, which therefore increases metal inputs into the river systems. The same trend was also observed by Kihampa et al. [31] in a similar environment.

3.2 *Occurrences and seasonal variations of metals in the sediment samples*

The levels of both Pb and Cd in sediments from all 15 sampling locations during the two seasons are presented in Figure 2. The data indicate that concentrations of Pb ranged from 10.7 to 159.3 mg/kg in the wet season (mean = 46.2 ± 35.9 mg/kg) and from 9.0 to 149.3 mg/kg in the dry season (mean = 43.1 ± 38.6 mg/kg). The Cd concentrations were from 0.10 to 0.45 mg/kg during the wet season (mean = 0.26 ± 0.17 mg/kg) and from 0.19 to 0.82 mg/kg during the dry season (mean = 0.22 ± 0.10 mg/kg). These concentrations reflect the general abundances of both metals in the upper Earth crust, where Pb is 20 mg/kg and Cd is 0.009 mg/kg [2].

The data in Figure 2 shows that the sampling location Kz_1 recorded the highest concentrations of both Pb (159.3 mg/kg) and Cd (0.45 mg/kg) during the wet season, whereas the lowest concentrations of both metals during both seasons were recorded at Mz_1. Another important observation was the conspicuously high concentrations of both Pb (149.1 mg/kg) and Cd (0.82 mg/kg) at Ms_4 during the dry season, which differed significantly from the rest of the dry season values as well as the wet season values for the rest of the locations. This might be an indication of point-source contamination in particular locations.

The correlation of the Pb and Cd concentrations in the sediment samples using Spearman's rank test revealed that at the 0.01 level (2-tailed), the two metals were strongly correlated ($r_s = 0.796$). This might be an indication that they are derived from the same source, most likely stormwater drainage of wastewater discharges. It also indicates that they have similar transport characteristics. A recent study by Mihale [46] in some coastal rivers in Dar es Salaam observed comparable results, where Cd and Pb levels in sediments showed a strong correlation.

The Pb and Cd levels in sediment samples were also compared to the sediment quality guidelines (SQGs). Tanzania lacks such guidelines for sediments; therefore, the Canadian Council of Ministers of the Environment (CCME) [47] and the Consensus-Based Sediment Quality (CBSQ) guidelines [48] were used for comparison purposes in this study. The CCME defines the level of probable effect (PEL) and interim guideline on freshwater sediment quality (ISQG), while the CBSQ guideline establishes the concentration of probable effect (PEC) level and the concentration of threshold effect (TEC) level.

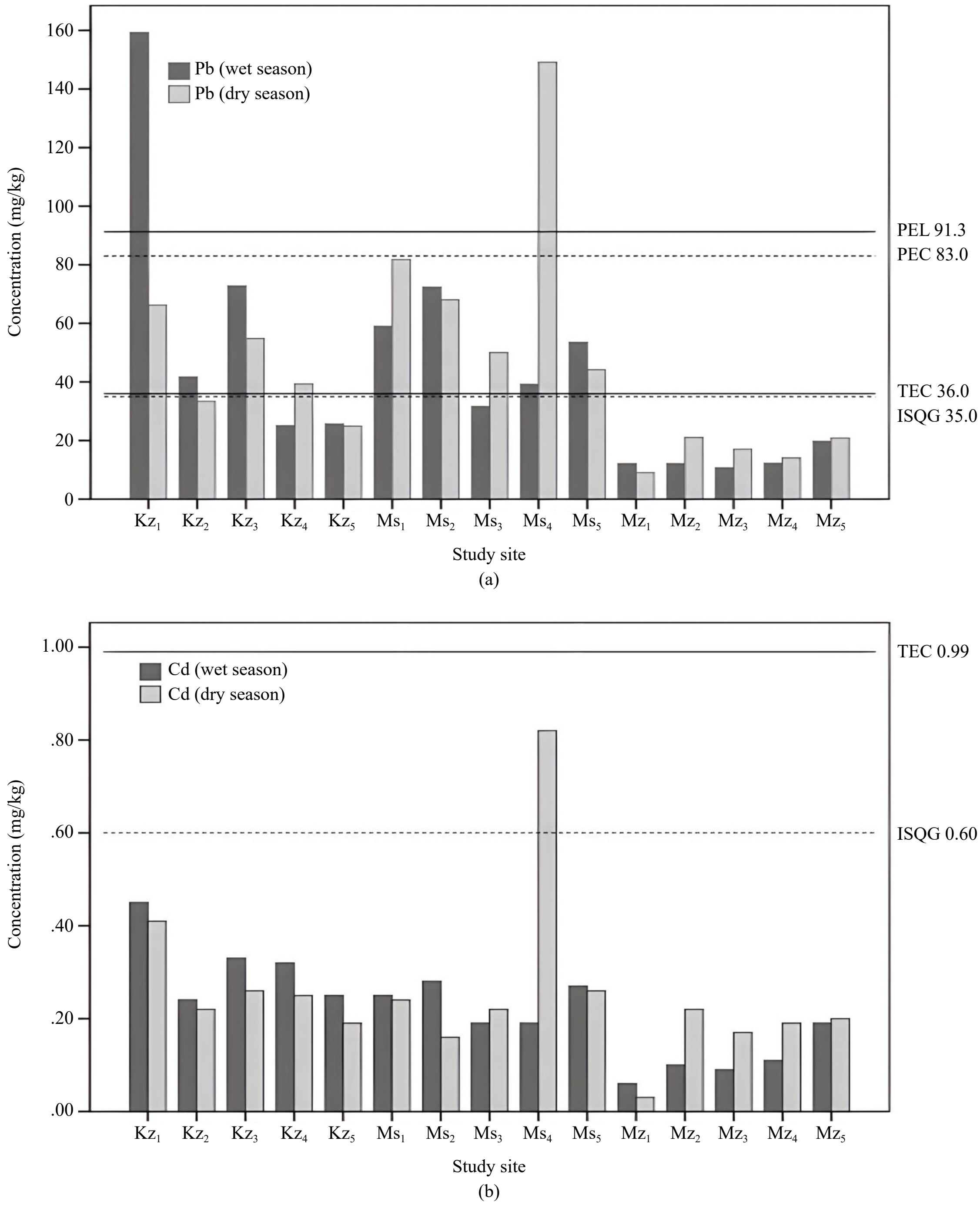

Figure 2. Concentrations of (a) Pb and (b) Cd in sediment samples from the 15 sites during the two seasons

The information in Figure 2 shows that the Pb levels in sediment exceeded both the TEC and the ISQG guidelines in nine out of the 15 locations, which included all of the Msimbazi River locations and four of the Kizinga River locations. All stations in the Mbezi River (n = 5) were below the referred SQGs. The Pb levels in locations Kz_1 and Ms_4 exceeded both the PEC and PEL guidelines by far, indicating that sediments at these locations pose a risk to the organisms dwelling underwater. The concentrations of Cd exceeded only the ISQG, and this was at one location. Although it may be considered less severe, the fact that this was the same location that had high Pb levels may raise concerns.

Determination of the ecological risk by PCI evaluation (Equation 1) revealed that the highest PCI values for Pb were at location Kz_1 (PCI = 18.39), followed by Ms_4 (PCI = 17.22), indicating that sediments at these sites were severely contaminated by Pb. It was also observed that the PCI values for the Msimbazi River, which ranged from 5.78 to 17.22, were relatively higher than those for the Mbezi and Kizinga. The determined PCI value of Cd also revealed severe contamination at Kz_1 (PCI = 3.21) and Ms_4 (PCI = 5.86), similar to Pb. Low to moderate contamination by Cd was observed in all the remaining stations (PCI = 0.43 to 2.36).

A comparison of the concentrations of both elements using the Friedman test revealed that the average levels of both Cd and Pb were significantly different at 95% CL, where Mbezi River had the lowest concentrations (n = 10, mean [M] = 0.14), followed by Msimbazi River (n = 10, M = 0.28), and Kizinga River with the highest mean concentrations (n = 10, M = 0.29), $\chi^2(2) = 8.769$, $p = 0.012$. The Cd and Pb contamination levels at Kz_1 and Ms_4 indicate that the two locations can be considered hotspots. Generally, the Pb and Cd levels observed in sediment in the area are similar to those recorded in other locations on the Tanzanian coast, particularly those that are relatively less anthropogenically influenced [20-22, 49]. The apparent variance can be attributed to the differences in the characteristics of the specific study locations, periods, and analytical methods used; some researchers analyzed the metals in the bulk samples, while others, such as Bungala et al. [22], Ngassapa et al. [50], and Sawe et al. [51], used metal speciation.

When concentrations of the two metals were statistically compared between the two seasons, it was revealed that concentrations of both metals were significantly higher during the wet season compared to the dry season. The test results showed that for Pb, the two datasets were strongly correlated ($r_s = 0.428$, $p = 0.112$), whereas the wet season mean was higher than the dry season mean by 3.14 mg/kg (95% CL [18.95 to 25.24]). Similarly, for the Cd, the two datasets were positively correlated ($r_s = 0.378$, $p = 0.117$), and the wet season mean was higher than the dry season mean by 0.04 mg/kg (95% CL [0.06 to 0.13]). A possible reason for this variation might be increased reception of the contaminated runoff from various sources such as vegetable farming, urban stormwater, and domestic effluents during the wet season.

3.3 *Occurrences and seasonal variations of metals in the biota samples*

The levels of Cd and Pb in the tissues of the mudskippers from the studied locations during the two seasons are summarized in Figures 3(a) and (b). These figures depict arithmetic means and standard errors. From the figures, it is shown that Pb was detected in ten out of the 15 locations. In the remaining five locations (Ms_3, Mz_1, Mz_3, Mz_4, and Mz_5), the Pb concentrations were below the BDL. The concentrations of Pb in the wet season ranged from BDL to 0.136 µg/g of dry weight (d.w; mean = 0.037 ± 0.041), and for the dry season, it was from BDL to 0.320 µg/g (d.w; mean = 0.055 ± 0.097). The highest level of Pb was found at Kz_1 during the dry season, followed by Kz_2. Figure 3(a) also shows that samples from the Kizinga River were generally the most contaminated with Pb, whereas those from the Mbezi River were the least contaminated, with Pb detected in only one of its locations (Mz_2).

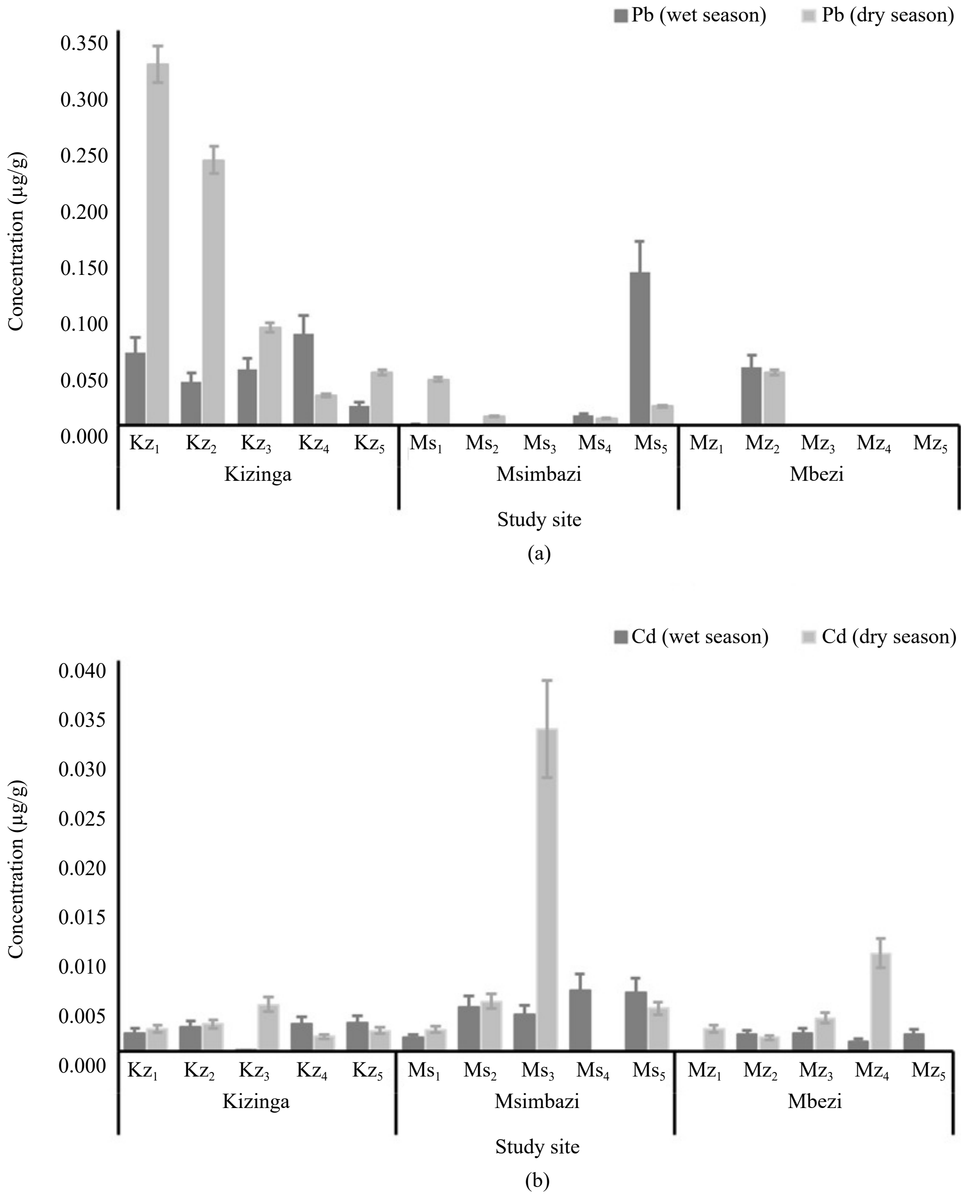

Figure 3. Concentrations of (a) Pb and (b) Cd in mudskipper tissue samples from the 15 sites during the two seasons

Figure 3(b) shows that Cd was detected in all locations with varying concentrations, ranging from BDL to 0.006 mg/g (d.w) during the wet season (mean = 0.003 ± 0.002) and from BDL to 0.033 mg/g (d.w) during the dry season (mean = 0.005 ± 0.008). The highest concentration of Cd was recorded at Ms_3 during the dry season. The Pb and Cd levels found in this study are higher than those found in fish from several markets in Dar es Salaam City and the Coast Region by Koleleni and Mosha [52], in which Pb was not detected in the fish samples. However, Koleleni and Mosha [52] sampled sardines (*Sardina pilchardus*), while this study sampled mudskippers. Shilla and Sawe [53] conducted a comparable study and discovered higher concentrations of Pb ranging from 0.57 to 0.80 mg/g in finfish and shellfish.

This study further assessed the safety of the fish samples from the study locations for human consumption. This was done based on comparisons with international food standards. The concentrations of Pb and Cd in the fish samples obtained in this study were lower than the general standards for contaminants and toxins in food and feed recommended by the Codex Alimentarius of the Food and Agriculture Organization (FAO) and WHO [54]. The concentrations of Pb and Cd in the fish samples were also positively correlated ($r_s = 0.675$), as was observed in the sediment samples. This indicates that they came from the same origin with similar transportation characteristics, as suggested for the sediments.

The datasets were further subjected to the paired samples t-test to assess the significance of their seasonal variations. The test results indicated that the wet and dry seasons concentrations of both Pb and Cd were strongly positively correlated, and their levels were significantly different during the two seasons. For Pb, the wet season mean was lower than the dry season mean by 0.018 mg/g (t_{14} = -0.746, 95% CL [-0.068 to 0.033]). Likewise, for Cd, the wet season mean was lower than the dry season mean by 0.002 g/g (t_{14} = -1.096, 95% CL [-0.006 to 0.002]). This was the opposite of what was observed for the sediment and water samples, which had higher concentrations during the wet season

3.4 *Estimated daily intake*

The CDI of the selected metals was used to estimate the daily intake. The CDI for Pb metal ranged from 9.0 x 10^{-5} at Mz_1 to 9.6 x 10^{-4} at Kz_1 in sediment, 7.0 x 10^{-5} at Mz_4 to 1.38 x 10^{-4} at Kz_3 in water, and 9.8 x 10^{-7} at Mz_1 to 4.3 x 10^{-6} at Kz_3 in biota. The CDI for Cd ranged from 1.5 x 10^{-6} at Mz_1 to 1.5 x 10^{-5} at Kz_1 in sediment, and from 0.0 at Ms_3, Mz_3, Mz_4 and Mz_5 to 6.5 x 10^{-6} at Kz_1 in biota (Tables 4, 5, and 6). The determined CDIs in water and biota were lower than those in sediment. Similarly, the determined CDI values were lower in the Mzinga River stations and higher in the Kizinga River stations. The determined CDI levels were lower than the maximum tolerable daily intake (MTDI), which implies that there is minimal health risk [35].

Table 4. CDI, HQ, HI and total risk for the two metals in sediment samples

Site code	CDI =∑CD		HQ		HI = ∑HQ (× 10^{-3})	Total risk	
	Pb (× 10^{-4})	Cd (× 10^{-5})	Pb (× 10^{-3})	Cd (× 10^{-4})		Pb (× 10^{-2})	Cd (× 10^{-3})
Kz_1	9.55	1.45	23.88	3.63	24.24	2.39	0.36
Kz_2	3.19	0.78	7.98	1.94	8.18	0.80	0.19
Kz_3	5.43	1.00	13.57	2.49	13.82	1.36	0.25
Kz_4	2.74	0.96	6.85	2.41	7.09	0.69	0.24
Kz_5	2.15	0.74	5.38	1.86	5.56	0.54	0.19
Ms_1	5.99	0.83	14.98	2.07	15.18	1.50	0.21
Ms_2	5.97	0.74	14.94	1.86	15.12	1.49	0.19
Ms_3	3.47	0.69	8.68	1.73	8.86	0.87	0.17
Ms_4	8.01	1.70	20.04	4.26	20.46	2.00	0.43
Ms_5	4.16	0.89	10.40	2.24	10.62	1.04	0.22
Mz_1	0.90	0.15	2.25	0.38	2.28	0.22	0.04
Mz_2	1.41	0.54	3.53	1.35	3.66	0.35	0.14
Mz_3	1.18	0.44	2.95	1.10	3.06	0.29	0.11
Mz_4	1.12	0.51	2.80	1.27	2.92	0.28	0.13
Mz_5	1.72	0.66	4.31	1.65	4.48	0.43	0.16

Table 5. CDI, HQ, HI and total risk for the two metals in water samples

Site code	CDI =∑CD		HQ		HI = ∑HQ (× 10^{-3})	Total risk	
	Pb (× 10^{-4})	Cd	Pb (× 10^{-3})	Cd		Pb (× 10^{-3})	Cd
Kz_1	0.83	0.00	2.09	0.00	2.09	2.09	0.00
Kz_2	0.90	0.00	2.26	0.00	2.26	2.26	0.00
Kz_3	1.38	0.00	3.45	0.00	3.45	3.45	0.00
Kz_4	0.63	0.00	1.57	0.00	1.57	1.57	0.00
Kz_5	0.65	0.00	1.63	0.00	1.63	1.63	0.00
Ms_1	0.22	0.00	0.55	0.00	0.55	0.55	0.00
Ms_2	0.31	0.00	0.77	0.00	0.77	0.77	0.00
Ms_3	1.49	0.00	3.72	0.00	3.72	3.72	0.00
Ms_4	1.30	0.00	3.26	0.00	3.26	3.26	0.00
Ms_5	0.82	0.00	2.04	0.00	2.04	2.04	0.00
Mz_1	0.11	0.00	0.29	0.00	0.29	0.29	0.00
Mz_2	0.17	0.00	0.41	0.00	0.41	0.41	0.00
Mz_3	0.21	0.00	0.52	0.00	0.52	0.52	0.00
Mz_4	0.07	0.00	0.17	0.00	0.17	0.17	0.00
Mz_5	0.09	0.00	0.23	0.00	0.23	0.23	0.00

Table 6. CDI, HQ, HI and total risk for the two metals in biota samples

Site code	CDI =∑CD		HQ		HI = ∑HQ (× 10^{-3})	Total risk	
	Pb (× 10^{-8})	Cd (× 10^{-6})	Pb (× 10^{-7})	Cd (× 10^{-4})		Pb (× 10^{-7})	Cd (× 10^{-4})
Kz_1	1.79	6.50	4.47	16.25	16.29	4.47	1.63
Kz_2	2.26	4.62	5.64	11.55	11.61	5.64	1.16
Kz_3	427.68	2.30	1069.20	5.76	16.45	1069.35	0.58
Kz_4	1.83	1.82	4.58	4.55	4.60	4.58	0.46
Kz_5	2.13	1.08	5.32	2.69	2.75	5.32	0.27
Ms_1	1.53	0.71	3.83	1.77	1.81	3.83	0.18
Ms_2	33.88	0.14	84.70	0.34	1.18	84.71	0.03
Ms_3	15.66	0.00	39.16	0.00	0.39	39.16	0.00
Ms_4	2.68	0.23	6.70	0.57	0.63	6.70	0.06
Ms_5	4.55	2.58	11.39	6.46	6.57	11.39	0.65
Mz_1	0.98	0.29	2.45	0.73	0.75	2.45	0.07
Mz_2	1.32	1.66	3.30	4.16	4.19	3.30	0.42
Mz_3	2.26	0.00	5.64	0.00	0.06	5.64	0.00
Mz_4	4.68	0.00	11.70	0.00	0.12	11.71	0.00
Mz_5	7.58	0.00	18.94	0.00	0.19	18.94	0.00

3.5 *Carcinogenic potential in sediment, water and biota*

The HQ was determined to evaluate health risks in sediment, water and biota. HQ values of < 1 are indicative of low risk, while HQ values of > 1 indicate high exposure risk [43]. The results in Table 2 indicate that concentrations of Pb and Cd in the water, sediment and biota at the study location pose no significant health risk through dermal contact or ingestion. The non-carcinogenic HI revealed that the analyzed samples were relatively safe. Similarly, the carcinogenic risk revealed that the samples are relatively safe because the values obtained were lower than the acceptable limits for the two metals. The carcinogenic risk is acceptable within the range of 1×10^{-6} and 1×10^{-4} [55]. Though the ranges of Pb and Cd levels are acceptable, the availability of these trace metals is a concern because of their cumulative health effects.

4. Conclusion

This study reports concentrations, distributions and seasonal trends of Pb and Cd in water, surficial sediments, and mudskippers from three urban rivers (Msimbazi, Kizinga, and Mbezi) along the Dar es Salaam coast in Tanzania. The findings obtained revealed that all three sample matrices were contaminated to varying degrees. While no traces of Cd were observed in the water samples, Pb levels exceeded the limits set by the WHO and the TBS for potable water at some locations, particularly in the Kizinga and Msimbazi Rivers. The concentrations of Pb also exceeded the Canadian Sediment Quality and the CBSQ guidelines for the protection of underwater dwelling organisms in the Kizinga and Msimbazi Rivers. Evaluation of the PCI in the sediment samples indicated impacts from human activities. Two sampling locations were found to have significantly high levels of Pb, which deemed them hotspots. These were sites Kz_1 in the Kizinga River and Ms_4 in the Msimbazi River. The concentrations of the two metals in the mudskipper samples were low and within the recommended limits for human consumption. Definite seasonal trends were observed in all three sample matrices. Metal concentrations in water and sediment samples were lower during the dry season than in the wet season, while in the biota samples, it was the opposite. Generally, the environmental and biota samples indicated relatively low potential health and carcinogenic risks. However, the presence of these metals, even in low concentrations, is a concern as they pose a potential health threat.

Conflict of interest

All authors declare no conflicts of interest in this paper.

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