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ORIGINAL ARTICLE

Evaluating the potential health risks of heavy metal pollution in sediment and selected benthic fauna of Benin River, Southern Nigeria

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Abstract

The potential ecological and human health risk of heavy metal pollution in sediment and Benthic Fauna (*Chrysichthys auratus* and *Tympanotonus fuscatus*) of Benin River, Southern Nigeria, was evaluated. Three sampling sites associated with heavy anthropogenic activities along the course of the river were sampled. Heavy metals concentrations were determined in the samples using atomic absorption spectrophotometer (Model 210 VGP, Buck Scientific). In all sediment samples, only Pb exceeded the threshold/probable effect level (TEL). Very high contamination degrees (CD > 24) 181.74, 50.11, and 101.96) for stations 1, 2, and 3, respectively, were observed indicating serious anthropogenic pollution. Geoaccumulation index (i_{geo}) showed slight pollution with Pb and Cd and severely to extremely polluted with Fe across the stations. Cd exhibited moderate individual potential risk (E_{r}^{i}), and the other heavy metals showed low E_{r}^{i}. Potential ecological risk index (RI) showed low risk of contamination for heavy metals in sediment. Human health risk assessment for Co, Cd, Cu, Zn, Mn, Fe, and Ni in *C. auratus* and Co, Zn, Mn, Fe, and Ni in *T. fuscatus* indicated no obvious health risk from these heavy metals over a lifetime of exposure. However, hazard quotient (HQ) values for Pb in *C. auratus* and Cd, Cu, and Pb in *T. fuscatus* indicated significant health risk. The hazard index (HI) values for both *C. auratus* and *T. fuscatus* were > 1 indicating significant adverse health risk of non-carcinogenic effect. Therefore, the consumption of these contaminated fish and shellfish by the people of Koko portends risks of the health of the public. The industries operating in this community should adopt more sustainable and eco-innovative management options in order to attenuate potential ecological and human health risk of metal pollution.

Keywords Metal pollution · Health risk · Sediment · Fish · Periwinkle

Introduction

Anthropogenic activities are major sources of heavy metals pollution in aquatic systems wordwide (Valavanidis and Vlachogianni 2010). Heavy metals refer to metals with a specific gravity greater than 5. They are toxic and accumulate within organisms in the natural environment. Heavy metals can be discharged into the aquatic environment via several routes including effluent/waste discharge, runoffs, leachates, shipping activities, and atmospheric depositions, especially from industrial and urban areas (Maanan 2008). In aquatic ecosystems, sediments play important roles in the growth, evolvement, and establishment of aquatic organisms. They are also a sink for pollutants. The ability of sediment to act as a sink for pollutants arises from a combination of processes, which include river hydrodynamics, biogeochemical processes, and environmental conditions. Consequently,

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heavy metals in sediments are useful markers of environmental changes in the aquatic ecosystem and give an indication of the ability natural mechanism to eliminate them while in this compartment (Arnason and Fletcher 2003). Within the aquatic food chain, the presence of heavy metals can lead to a wide range of effects ranging from molecular alterations to deaths in local fish populations (Massaquoi et al. 2015). Furthermore, the presence of toxic levels of heavy metals in benthic organisms has led to a sharp reduction in the diversity, growth, and reproduction rates of these organisms. The potential health consequences of heavy metal toxicity on humans through food transfer by consumption are an issue of serious concern.

In response to the growing public concern about heavy metal pollution, there has been an increase in the monitoring of heavy metal concentrations in aquatic systems globally. In Nigeria, several studies have determined the presence of heavy metals in various environmental compartments and biota especially in the Nigeria Delta region. This area is noted for its massive oil production activities that have led to increased rate of heavy metal contamination and pollution. At present, assessment and monitoring of heavy metal contamination in the Niger Delta mainly the Benin River has been predominately based on chemical analysis of environmental compartments of water, sediment, and soil alone. This step is fundamentally insufficient in deriving the potential toxicity of contaminated environmental samples. Moreover, chemical analysis on abiotic compartments cannot directly assess the antagonistic or synergistic effects as well as the bioavailability of toxicants to organisms because the magnitude of contamination does not necessarily reflect a similar level of ecotoxicological effect (Barhoumi et al. 2016). Consequently, scientists have developed many indices in evaluating the potential risk of heavy metal in sediments based on the total content, bioavailability, and toxicity to associated and exposed fauna (Yang et al. 2009).

There are several methods to assess sediment quality with the aim of describing the adverse effects of contamination (Ridgway and Shimmield 2002). Some of these methods include; Sediment Quality Guidelines (SQGs), contamination factor (CF), contamination degree (CD), pollution load index (PLI), geoaccumulation index (I_{geo}), and potential ecological risk index (RI). They were employed in this study to assess the degree of contamination of the sediments, anthropogenic influence on the sediment quality and describe the sensitivity of the biota to the toxic heavy metals.

Sediment quality guidelines (SQGs) are essential tools for identifying contaminated sediment hotspots and also for assessing possible effects of contaminated sediments on benthic organisms (Harikumar et al. 2009; Luo et al. 2010). Sediment contamination is estimated by comparing sediment contaminant concentration with the corresponding quality guideline (MacDonald et al. 2000). These guidelines are also designed to assist in the interpretation of sediment quality. For freshwater ecosystems, two guidelines have been developed: the effects range low/effects range median (ERL/ERM) and threshold/probable effect level (TEL/PEL). The low range values (ERL or TEL) have been reported as the concentration of contaminants with a relatively low effect on biological communities; below this concentration, there will be a rare occurrence of adverse effects upon sediment dwelling fauna. On the other hand, ERM and PEL values represent contaminant concentrations above which adverse effects are likely to occur (Long and MacDonald 1998; MacDonald et al. 1996). These SQGs were developed based on sediment toxicity information collected for freshwater and saltwater sediments throughout the USA and were developed in a manner consistent with the TELs and PELs for freshwater sediments by Smith et al. (1996). Human health risk assessment of potentially toxic heavy metals provides an indication of the risk level due to pollutant exposure, and it is based on the characterization or quantification of the risk level either as carcinogenic or a non-carcinogenic risk (Cherfi et al. 2016).

Therefore, the aim of the current study was to assess the distribution of heavy metals in the sediments of the Benin River and evaluate the ecological and human health risks posed by contaminated sediments and ingested benthic fauna (fish: *C. auratus* and periwinkle: *T. fuscatus*) from Benin River using indices based on the total heavy metal content.

Materials and methods

Description of the study area

The Benin River is in the western Niger River Delta region of Nigeria. It flows through Koko town, in Delta State, Southern Nigeria. The river acts as a drain to various oil-processing outfits along its stretch of the river (Iwegbue et al. 2008). The town is reportedly known as a petroleum prospecting and processing area, and it is home to various petroleum products depot and local illegal refineries lubricating oil factories and oil distribution outfits (Akporido and Asagba 2013). The river is also the hub of various commercial activities including a collection point for palm oil and kernels and timber. Other activities around the river and its port include fishing which also comprises of the harvesting of crayfish and shrimps. The river is also used for transportation since it is wide and deep, while the adjoining land is used for cultivation of arable and commercial crops (Akporido and Ipeaiyeda 2014). The proximity of these activities to the Benin River makes the river a suitable drain for effluents containing toxic heavy metals.

Sampling sites

The sampling sites were selected based on proximity to areas of anthropogenic activities. Station 1 is beside a mangrove forest, immediately after Ebialegbe, a rural community (5°, 58.264′ N and 5°, 29.433′E) located close to the River. Station 2 is located in a zone that is in the vicinity of a waste management facility, bulk oil storage facilities and watercraft maintenance workshop (5°, 59.830′ N and 5°, 27.859′E). Station 3 is located adjacent a bitumen Blending Plant, which regularly discharges effluents from its operations directly into the river (Fig. 1).

Sample collection

Surface sediment samples were collected using an Ekman grab. Sampling was conducted between December 2014 to May 2015, covering part of dry and rainy seasons. Fifty-four sediment samples were collected from the core of the grab to prevent contamination from the wall of the grab using a rubber spatula. After collection, samples were wrapped in an aluminum foil and transported below −4 °C to the laboratory for analysis. All sediment samples were freeze-dried and sieved using a 2-mm mesh sieve to remove debris (US EPA 2007). Samples of *C. auratus* and *T. fuscatus* were collected from several points along the stretch of the river. They were then transported to the laboratory and preserved in the refrigerator within 24 h after collection until analysis.

Sample analysis

Analysis of heavy metals in sediment

Digestion of sediment samples was performed following procedures described by USEPA, 2007 and AOAC, 1990. In summary, a 10:4:1 mixture of nitric acid (HNO_3), perchloric acid ($HClO_4$), and sulfuric acid (H_2SO_4) was added to 0.25 g of each sediment sample. The samples were heated at 70 °C for 1 h, and then 10 mL of deionized water was added to the solution. The final suspended mixture was cooled and filtered through a 0.45-μm membrane filter. The same procedure was performed with a blank and a standard reference material in each batch of digestion. Solutions were kept in vials for further analysis using an atomic absorption spectrophotometer (Model 210 VGP, Buck Scientific).

Analysis of heavy metals in biota

Biota samples were processed within 4 h after collection. Samples were rinsed thoroughly in distilled water and stored at −4 °C before analysis. Edible muscles of *C. auratus* were used for this analysis, while soft tissues of deshelled *T. fuscatus* were used for analysis.

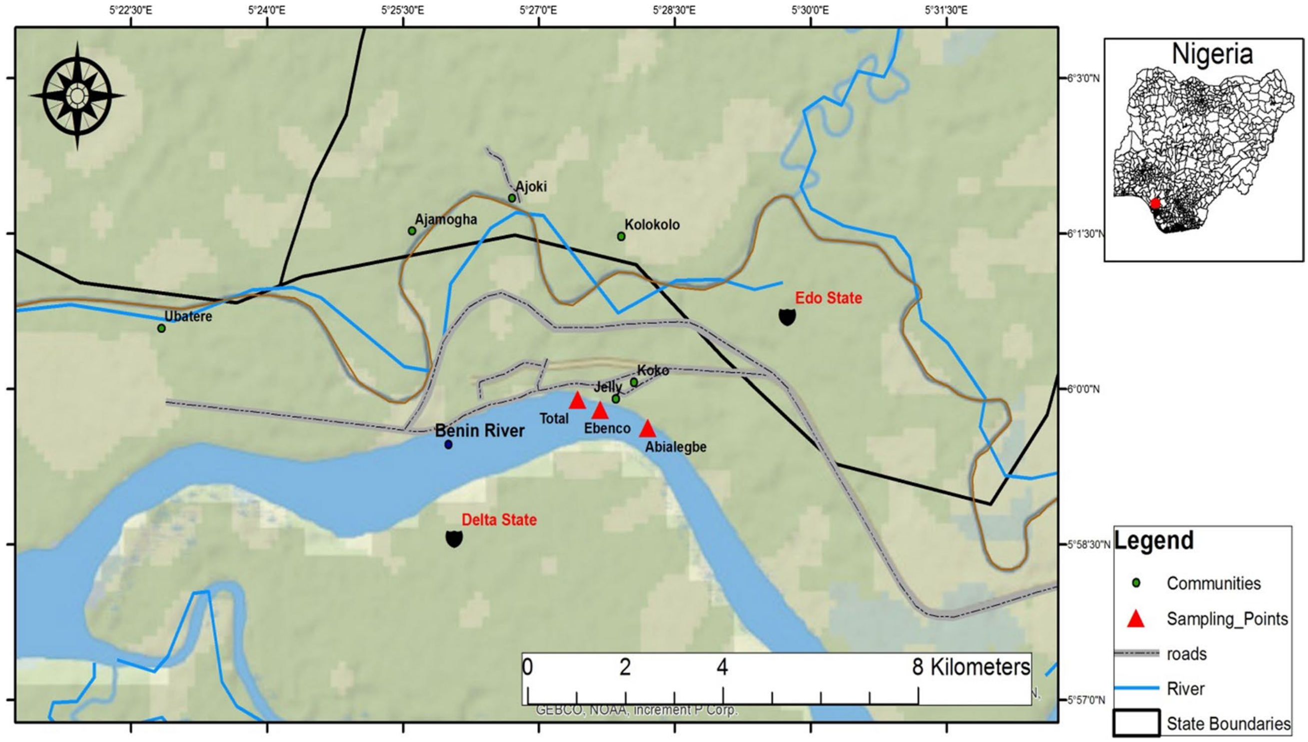

Fig. 1 Map showing study area

Samples were freeze-dried and homogenized using a porcelain mortar and pestle, sieved with a 150-µm nylon mesh sieve, and stored in appropriate glass bottles at −20 °C until metal analysis. Then 0.5 g of each homogenized sample was digested with a 10:4:1 mixture of nitric acid (HNO_3), perchloric acid ($HClO_4$) and sulfuric acid (H_2SO_4) in 100-ml Kjeldahl flask. The samples were heated at 70 °C for 1 h, allowed to cool, and then 10 mL of deionized water was added to the solution. Subsequently, 40 ml of deionized water was added to each vessel, and the resulting mixture transferred to 50-ml vials. The same procedure was performed with a blank and a standard reference material in each batch of digestion. Solutions were kept in vials for further analysis using an atomic absorption spectrophotometer (Model 210 VGP, Buck Scientific).

Quality assurance and control

The equipment was calibrated using buck-certified atomic absorption standards for the several heavy metals to obtain a calibration curve. Reagent blank was first run at intervals of every 10 samples analysis to eliminate equipment drift. Recoveries ranged from 82 to 110%. Metal concentrations in sediments and biota samples were analyzed by atomic absorption spectrophotometry (Model 210 VGP, Buck Scientific). The AAS detection limits (mg/kg) were 0.1 (Pb), 0.2 (Cr), 0.1 (Ni), 0.05 (Cu), and 0.6 (Zn). All samples were run in duplicates, and the mean values were reported.

Ecological risk assessment of heavy metals in sediment

Pollution load index (PLI)

Pollution load index (PLI) represents the number of times by which the metal content in the sediment exceeds the background concentration. It provides comprehensive information about the metal toxicity in a particular sample (Yang et al. 2011). The pollution load index (PLI) is defined as the nth root of the multiplications of the concentrations. The PLI value of > 1 indicates polluted, whereas < 1 indicates no pollution (Barakat et al. 2012). PLI was evaluated using the following formula proposed by Tomilson et al. (1980).

$$\mathrm{PLI} = (\mathrm{CF}_1 \times \mathrm{CF}_2 \times \mathrm{CF}_3 \cdots \times \mathrm{CF}_n)^{1/n}$$

where n is the number of metals (eight in the present study) and CF is the contamination factor.

The contamination factor can be calculated from the following relation:

$$\mathrm{CF} = \frac{\text{Metal concentration in sediment}}{\text{Background value of metal}}.$$

According to Håkanson (1980), CF < 1 indicates low degree of contamination, 1 < CF < 3 indicates moderate degree of contamination, 3 < CF < 6 indicates considerable degree of contamination, and CF > 6 indicates very high degree of contamination.

Contamination degree (CD)

This parameter refers to the sum of all contamination factors. It gives an indication of the degree of overall contamination in sediments from a sampling site. It expressed as:

$$\mathrm{CD} = \sum_{i=1}^{n} \mathrm{CF}.$$

Håkanson (1980) proposed the classification Cd < 6 is low degree of contamination, 6 ≤ Cd < 12 is indicative of moderate degree of contamination, 12 ≤ Cd < 24 indicates considerable degree of contamination, and Cd ≥ 24 represents very high degree of contamination.

Geoaccumulation index (I_{geo})

The geoaccumulation index (I_{geo}) introduced by Muller (1969) is widely used to quantify the level of heavy metal contamination in sediment. This index is used to determine metals contamination in sediments, by comparing current concentrations with pre-industrial levels.I_{geo} is mathematically expressed as:

$$I_{\mathrm{geo}} = \log_2 \left[\frac{C_n}{1.5B_n} \right]$$

where C_n is the concentration of element 'n' and B_n is the geochemical background value or each metal. World surface rock average was used as background values (Turekian and Wedepohl 1961; Tang et al. 2016). The factor 1.5 is incorporated in the relationship to account for possible variation in background data due to lithogenic effect (Wang et al. 2016).

Muller (1969) classification of I_{geo} grouped it into seven grades: $I_{\mathrm{geo}} \leq 0$ (grade 0), unpolluted; $0 < \leq 1$ (grade 1), slightly polluted; $1 < I_{\mathrm{geo}} \leq 2$ (grade 2), moderately polluted; $2 < I_{\mathrm{geo}} \leq 3$ (grade 3), moderately severely polluted; $3 < I_{\mathrm{geo}} \leq 4$ (grade 4), severely polluted; $4 < I_{\mathrm{geo}} \leq 5$ (grade 5), severely to extremely polluted; and $I_{\mathrm{geo}} > 5$ (grade 6), extremely polluted.

Potential ecological risk index (RI)

The potential ecological risk could be used to evaluate the ecological risk of heavy metals in sediments by considering the toxicity of the metal and a comparison between the concentration of the metal and the background value. *RI* was used in this study to quantify the potential ecological hazard of contaminated sediment to biota. Håkanson (1980) provided a formula to estimate *RI*.

Firstly,

$$E_r^i = T_r^i \times \mathrm{CF},$$

where T_r^i is the toxic response factor for a given substance and CF is the contamination factor.

The toxic response factor assigned to the following heavy metals Co, Cd, Cu, Zn, Mn, Pb, and Ni used in the calculation of potential ecological risk index (RI) are 5, 30, 5, 1, 1, 5, and 5, respectively (Jiao et al. 2015; Soliman et al. 2015).

The sum of the individual potential risks (E_r^i) is the potential ecological risk index (RI) for the water body. It is presented as:

$$\mathrm{RI} = \sum_{i=1}^{n} T_r^i \times \mathrm{CF}.$$

For the classification of individual potential risks (E_r^i) in sediments, $E_r^i \leq 40$ indicates low ecological risk, $40 < E_r^i \leq 80$ indicates moderate ecological risk, $80 < E_r^i \leq 160$ indicates considerable ecological risk, $160 < E_r^i \leq 320$ indicates high ecological risk, $E_r^i > 320$ indicates very high ecological risk. Furthermore, classification of potential ecological risk index (RI) is as follows:

$\mathrm{RI} \leq 150$ = low ecological risk,
$150 < \mathrm{RI} \leq 300$ = moderate ecological risk,
$300 < \mathrm{RI} \leq 600$ = considerable ecological risk,
$\mathrm{RI} > 600$ = very high ecological risk.

Sediment-to-benthic transfer assessment

Sediment-to-benthic fauna metal transfer was computed as transfer factor (*TF*) which is defined by the equation.

$$\mathrm{TF} = \frac{C_{\mathrm{fauna}}}{C_{\mathrm{sediment}}}$$

where C_{fauna} is the concentration of heavy metals in *C. auratus* and *T. fuscatus*, respectively, and C_{sediment} is the concentration of heavy metals in sediment.

Human health risk assessment of heavy metals in sediment and biota

Exposure assessment

Exposure to toxic heavy metals could also be of significant concern to humans living close to contaminated aquatic ecosystems. There are three primary pathways of exposure to heavy metals in sediments when dealing with human health risk assessment. They are ingestion, dermal contact, and inhalation. The exposures through ingestion, inhalation, and dermal contact were, respectively, calculated using equations below

$$\mathrm{EXP\ (ingestion)} = \frac{C \times \mathrm{IRs} \times \mathrm{ED} \times \mathrm{EF}}{\mathrm{BW} \times \mathrm{AT}},$$

$$\mathrm{EXP\ (dermal)} = \frac{C \times \mathrm{CF} \times \mathrm{SA} \times \mathrm{AF} \times \mathrm{ABS} \times \mathrm{EF} \times \mathrm{ED}}{\mathrm{BW} \times \mathrm{AT}},$$

$$\mathrm{EXP\ (inhalation)} = \frac{C \times \mathrm{IR(inh)} \times \mathrm{EF} \times \mathrm{ED}}{\mathrm{PEF} \times \mathrm{BW} \times \mathrm{AT}},$$

where *C* is the concentration of heavy metals in the sediment; IRs is the ingestion rate (114 mg/day); CF is the unit conversion factor (10^{-6} kg/mg); EF is the exposure frequency (350 days/year); ED is the exposure duration (30 years); BW is the body weight (70 kg); SA is the exposed skin surface area (5700 cm^2); AF is the adherence factor from sediment to skin (0.07 $\mathrm{mg/cm}^2$); and ABS is the dermal absorption from sediment (0.001) (unitless); SL is the skin adherence factor (0.2 $\mathrm{mg\ cm^{-2}\ h^{-1}}$) for children and (0.2 $\mathrm{mg\ cm^{-2}\ h^{-1}}$)for adults; PEF is the particle emission factor ($1.316 \times 10^{-9}\ \mathrm{m^3\ kg^{-1}}$); AT is the average time. For non-carcinogens, it is ED × 365 days. For carcinogens, it is $70 \times 365 = 25{,}550$ days.

Similarly, dietary intake of contaminated food has been implicated as a primary source of human exposure to toxic chemicals including heavy metals. The exposures through ingestion of contaminated *C. auratus* and *T. fuscatus*, respectively, were calculated using equation below

$$\mathrm{EXP(diet)} = \frac{\mathrm{C} \times \mathrm{IR(biota)} \times \mathrm{ED} \times \mathrm{EF}}{\mathrm{BW} \times \mathrm{AT}},$$

where *C* is the concentration of the per mass of the medium (ppm), IR is the ingestion rate of the medium (g/day), ED is the exposure duration (years), EF is the exposure frequency (days/year), BW is the body weight (kg) and AT is the averaging time (years).

Risk characterization

Non-cancer risk

The potential non-cancer risk of heavy metal concentrations in sediments and biota is characterized using a hazard quotient (HQ). Hazard quotient (HQ) assumes that there is a level of exposure known as the reference dose (RfD). It is estimated that a daily oral intake of the heavy metal at the reference dose will pose no reasonable risk even to sensitive populations, over a 70-year lifetime (Afrifa et al. 2013). USEPA, 2010, defines hazard quotient (HQ) as the ratio of the average daily intake or dose (ADD) (mg/(kg/day)) to the reference dose (RfD, mg/(kg/day)). It was estimated using the formula:

$$\mathrm{HQ} = \frac{\mathrm{EXP}}{\mathrm{RfD}}$$

where HQ = hazard quotient (unitless), ADD = average daily dose (mg/kg$^{-\mathrm{day}}$), RfD = Reference dose (mg/kg$^{-\mathrm{day}}$). For n number of heavy metals, the non-carcinogenic effect to the population is as a result of the summation of all the HQs due to individual heavy metals.

$$\mathrm{HI} = \mathrm{HQ}_1 + \mathrm{HQ}_2 \cdots + \mathrm{HQ}_n$$

If the HI is less than 1.0, it is highly unlikely that significant additive or toxic interactions would occur, so no further evaluation is necessary. When the HI exceeds 1.0, there may be a concern for potential non-cancer health effect.

Cancer risk

The potential cancer risk of heavy metals in sediment and biota were estimated using the incremental or excess individual lifetime cancer risk. Risks are estimated as the incremental probability of an individual developing cancer over a lifetime as a result of exposure to the potential carcinogen. For all matrices, the cancer risk was estimated using the following formula:

$$\text{Cancer Risk} = \sum_{k=1}^{n} \mathrm{EXPk} \times \mathrm{CSFk}$$

where risk is a unitless probability of an individual developing cancer over a lifetime. EXP$_k$ (mg/kg/day) is the average daily intake while CSF$_k$ is the cancer slope factor (mg/kg/day)$^{-1}$ for the kth heavy metal, for n number of heavy metals. The slope factor converts estimated daily intakes averaged over a lifetime of exposure directly to the incremental risk of an individual developing cancer.

Statistical analysis

One-way analysis of variance (ANOVA) was used to determine the differences in heavy metals concentrations in sediment and benthic fauna between wet and dry seasons at a significant level of 0.05. Standard errors were also estimated. All statistics were run on the computer using Microsoft Excel 2010 and IBM SPSS Statistics 20.

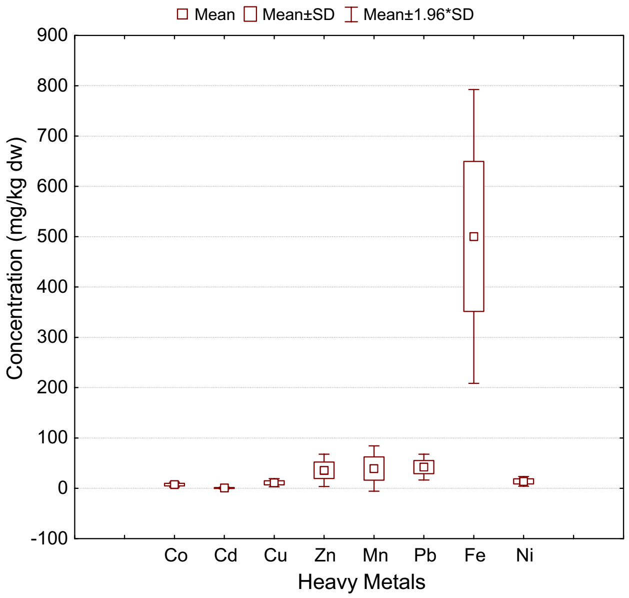

Fig. 2 Box and whisker plot showing variation in heavy metals concentrations in sediment

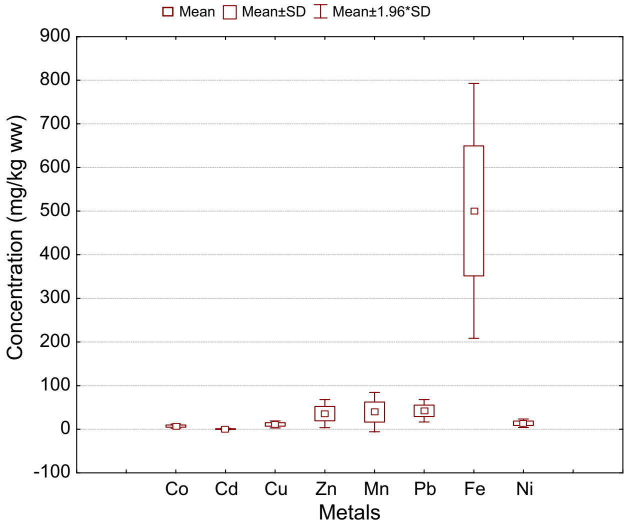

Fig. 3 Box and whisker plot showing variation heavy metals concentrations in fish

Results and discussion

The mean variations of heavy metals in sediments, fish, and periwinkle are depicted in Figs. 2, 3, and 4, respectively. Fe showed the widest variation in concentration for sediment, fish, and shrimp in all the stations sampled, while Cd showed the least variation. The profile of concentrations of heavy metals in all samples was: Fe > Pb > Mn > Zn > Ni > Cu > Co > Cd.

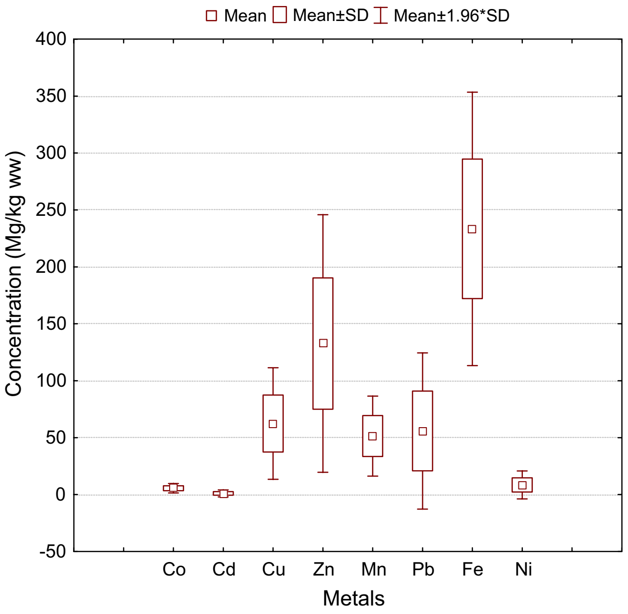

Fig. 4 Box and whisker plot showing variation heavy metals concentrations in *T. fuscatus*

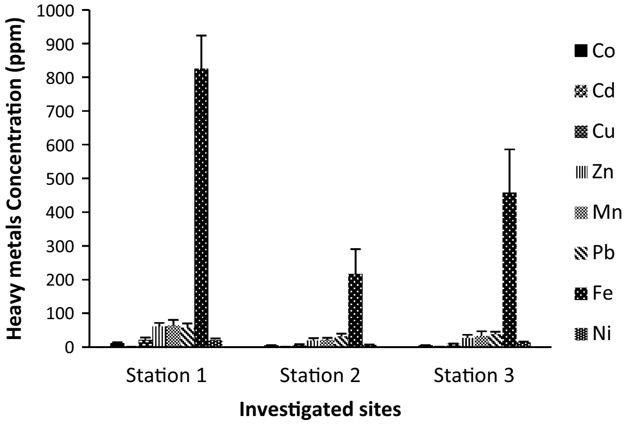

Fig. 5 Spatial distribution of heavy metals concentrations across investigated stations

Variation in metal concentrations in sediment, fish, and periwinkle for all study areas

Figure 5 shows variation of Co, Cd, Cu, Zn, Mn, Pb, Fe, and Ni in sediments of Benin River with respect to different locations investigated.

Fe and Cd were highest and lowest, respectively, in all the three stations. Station 1 had the highest mean concentrations of all the investigated heavy metals except Cd which had the highest mean concentration in stations 2 and 3. This may be related to particle size of sediment and the nearby mangrove forest in station 1. Quite a few researchers reported that sediment particle size is a significant parameter which is able to control heavy metal concentration because fine particles have high ability to adsorb soluble heavy metals and deposit them at the bottom sediment (Lijklema et al. 1993; Abrahim et al. 2007; Nobi et al. 2010). In the same way, several studies have shown that mangrove forests can increase the suspended solid deposition by decreasing the water dynamic energy and providing enough time for fine grain size to sink and deposit (Woodroffe 1992; Wolanski et al. 1992; Kathiresan 2003; Cunha-Lignon et al. 2009). Many other studies have shown that mangrove sediments act as a trap for chemical contaminants because such sediments contain high percentage of silt and clay that cause an increase in the metals adsorption (Lacerda 1998; Shriadah 1999; Ranjan et al. 2008; Vallejuelo et al. 2010). Generally, the mean concentrations of heavy metals in station 2 were relatively lower. This may be due to the sandy nature (coarse grain particles), the land-based runoff and the various pollution control services of Ebenco Global Link Limited that has waste management and storage facilities on the bank. However, the concentration of Cd peaked at stations 2 and 3 probably because of industrial waste from watercraft maintenance workshop near station 2 and industrial outlets in station 3.

Table 1 shows that the concentrations of Fe, Pb, and Cd, in all the three stations exceeded their natural background levels suggesting high enrichment of sediments with these heavy metals. The mean heavy metal concentrations in sediments collected from the studied stations in Benin River were also compared with US Environmental Protection Agency (USEPA) Sediment Quality Guidelines (SQGs). Taking USEPA SQGs into consideration, station 1 sediment were moderately polluted with Cu, Pb and Ni and non-polluted with Zn, Mn and Fe. Stations 2 and 3 are non- polluted with Cu, Zn, Mn, Pb, Fe and Ni. Hence, station 1 ranges from non-polluted to moderately polluted with the investigated heavy metals. While SQGs may be appropriate in some situations, scientists generally acknowledge there are several limitations and uncertainties associated with different SQG approaches that have the potential to cause confusion and concern among sediment assessment and management practitioners (Wenning and Ingersoll 2002).

Furthermore, the heavy metal concentrations in the sediment were compared with the threshold-effects level (TEL) and probable-effects level (PEL) values. In all sediment samples, only Pb exceeded the TEL value, although Cu and Ni exceeded the TEL values only in station 1, and Pb in all the three stations. The exceedance of the TEL values with respect to Cu, Pb, and Ni in station 1 suggests that the station is moderately toxic, while the exceedance of TEL by only Pb in stations 2 and 3 is less toxic. This implies that the occasional toxic effects are expected for Cu, Pb, and Ni in station 1 and rare toxic effect probably occur for Pb in stations 2

Table 1 Comparison of geochemical background values, SQG by USEPA and TEL/PEL guideline values with the mean heavy metals concentration (ppm) of sediment samples from Benin River

	Co	Cd	Cu	Zn	Mn	Pb	Fe	Ni
Station 1								
Mean ± SE	11.69 ± 2.44	0.55 ± 0.35	20.84 ± 7.18	60.72 ± 10.90	64.06 ± 16.76	57.19 ± 12.75	825.68 ± 98.25	21.71 ± 3.96
Minimum	4.01	0	4.23	9.12	15.54	15.87	342.33	4.65
Maximum	19.95	1.72	54.67	83.56	103.43	103.88	975.9	31.65
Station 2								
Mean ± SE	4.86 ± 1.37	0.56 ± 0.35	6.88 ± 1.71	18.89 ± 7.84	20.78 ± 6.42	31.49 ± 7.54	216.87 ±73.38	6.43 ± 1.31
Minimum	1.21	0	2.8	2.66	5.68	9.42	26.1	2.24
Maximum	10.69	1.77	14.67	49.96	45.31	54.61	439.93	10.56
Station 3								
Mean ± SE	5.29 ± 0.82	0.56 ± 0.36	9.07 ± 0.94	27.59 ± 8.90	33.04 ± 13.67	38.16 ± 7.55	458.63 ± 127.70	13.05 ± 3.02
Minimum	2.93	0	6.39	5.17	9.97	17.97	59.4	4.3
Maximum	8.26	1.7	12.07	63.95	96.97	61.69	862.5	26.26
All samples								
Mean ± SE	7.28 ± 2.21	0.56 ± 0.00	12.26 ± 4.34	35.73 ± 12.74	39.29 ± 12.88	42.28 ± 7.70	500.39 ± 176.98	13.73 ± 4.42
Minimum	4.86	0.55	6.88	18.89	20.78	31.49	216.87	6.43
Maximum	11.69	0.56	20.84	60.72	64.06	57.19	825.68	21.71
World surface rock average[a] SQG	19	0.3	45	95	850	20	4.72	68
Non-polluted[b]	–	–	< 25	< 90	< 300	< 40	<17,000	< 20
Moderately polluted[b]	–	–	25–50	90–200	300–500	40–60	17,000–25,000	20–50
Heavily polluted[b]	–	> 6	50	> 200	> 500	> 60	>25,000	> 50
TEL[c]	–	0.68	18.7	124	–	30.2	–	15.9
PEL[c]	–	4.21	108	271	–	112	–	42.8

– values unavailable

[a] Geochemical background value taken is that given by Turekian and Wedepohl (1961)

[b] USEPA SQG given by Perin et al. 1997

[c] TEL/PEL guidelines developed by MacDonald et al. (1996)

and 3. However, exceedance of SQG values does not firmly guarantee the occurrence of deleterious ecological effects, unless they are also coherent with regional background levels (Soliman et al. 2015). Hence, occasional toxic effects are expected for Pb in the three stations.

Contamination degree and pollution load index

Table 2 shows the average CD and PLI values for different heavy metals in the sediments collected from Benin River. For all stations along the Benin River, the CF value for Fe was > 6, while that of Pb and Cd exceeded 1 but > 3. The rest of the heavy metals had CF values > 1. Also, very high contamination degrees (CD > 24) were estimated. However, on the basis of the mean values of CD, the pollution levels for the stations in the following order: 1 > 3>2.

Table 2 Calculated contamination degree (CD) and Pollution Load Index (PLI) of sediment samples from Benin River

Metals	Station 1	Station 2	Station 3	All samples
Co	0.62	0.26	0.28	0.38
Cd	1.84	1.86	1.88	1.86
Cu	0.46	0.15	0.20	0.27
Zn	0.64	0.20	0.29	0.38
Mn	0.08	0.02	0.04	0.05
Pb	2.86	1.57	1.91	2.11
Fe	174.93	45.95	97.17	106.02
Ni	0.32	0.09	0.19	0.20
CD	181.74	50.11	101.96	111.27
PLI	1.19	0.47	0.67	0.78

Table 3 Geoaccumulation index values for sediment samples from the Benin River

Metals	Station 1	Station 2	Station 3	All samples
Co	− 1.29	− 2.55	− 2.43	− 1.97
Cd	0.29	0.31	0.33	0.31
Cu	− 1.70	− 3.29	− 2.90	− 2.46
Zn	− 1.23	− 2.92	− 2.37	− 2.00
Mn	− 4.32	− 5.94	− 5.27	− 5.02
Pb	0.93	0.07	0.35	0.49
Fe	6.87	4.94	6.02	6.14
Ni	− 2.23	− 3.99	− 2.97	− 2.89

Table 4 Individual potential risks (E_r^i) and potential ecological risk (RI)

Metals	Station 1	Station 2	Station 3	All samples
Co	3.08	1.28	1.39	1.92
Cd	55.11	55.78	56.44	55.78
Cu	2.32	0.76	1.01	1.36
Zn	0.64	0.20	0.29	0.38
Mn	0.08	0.02	0.04	0.05
Pb	14.30	7.87	9.54	10.57
Ni	1.60	0.47	0.96	1.01
RI	77.11	66.39	69.67	71.06

The value of PLI ranged from 1.19 in station 1–0.47 in station 2 (Table 6). PLIs for the heavy metals in the sediments were less than 1 in stations 2 and 3 except station 1. Stations 2 and 3 had the lowest and the highest PLI values, respectively. Higher PLI values (PLI > 1) in sediments demonstrated substantial anthropogenic impacts on the sediment quality whereas lower PLI values (PLI < 1) pointed to no considerable anthropogenic activities.

According to CF values (Table 4), all the three stations indicated that Benin River is highly contaminated with Fe, although Pb and Cd displayed moderate contamination. The other heavy metals exhibited low contamination in general. Very high contamination degrees (CD > 24) 181.74, 50.11, and 101.96 for stations 1, 2, and 3, respectively, were observed indicating serious anthropogenic pollution. The pollution levels for the stations in the following order: 1 > 3 > 2, suggested that the station located in the upstream is more seriously polluted by heavy metals than the two other stations. The PLI value (1.19) suggests that station 1 is polluted and also indicates anthropogenic impacts, while Stations 2 and 3 with PLI values 0.47 and 0.67 are designated as no to low pollution and also point to no considerable anthropogenic activities. Hence, the results in this study indicate that heavy metal contamination in sediments of Benin River was as a result of both natural and anthropogenic sources.

Geoaccumulation index

The results of the calculated I_{geo} in Table 3 shows that stations 1, 2, and 3 had values < 0 for Co, Cu, Mn, Zn, and Ni, > 0 for Cd and Pb. For Fe, the I_{geo} is > 4 in station 2, and > 6 in stations 1 and 3. According to the calculated I_{geo}, all the three stations are slightly polluted with respect to Cd and Pd. Station 2 is severely to extremely polluted with Fe, though stations 1 and 3 are extremely polluted. The calculated I_{geo} for the other investigated heavy metals fell into grade 0, unpolluted.

Ecological risk assessment

Table 4 summarizes the individual potential risks (E_r^i) of different heavy metals and their contributions to the potential ecological risk index (RI) of the sediments from the three different investigated stations in the Benin River. Cd had the highest E_r^i (greater than 40), in all the three stations despite the fact the other heavy metals had much lower E_r^i of less than 40. Worthy to note is that the highest value for E_r^i for Cd occurred in station 3. In all the stations, the RI values were much lower than 150. The E_r^i of seven heavy metals in the sediments of the Benin River were in the order: Cd > Pb > Co > Cu > Ni > Zn > Mn. Similar to the model of *PLI*, the RI decreased in the order of: station 1 > station 3 > station 2. The potential ecological risk index (RI) values for stations 1, 2, and 3 were 77.11, 66.39, and 69.67, respectively. The E_r^i of Cd, which belongs to moderate ecological risk, highlights the adverse effect it poses to benthic fauna and ecosystem in general. This implies that all investigated stations have low ecological risk for the individual heavy metals except Cd. Station 3 had the highest single ecological risk for Cd. All the three stations along Benin River have low RI due to heavy metal contamination the as values were lower than 150. The individual potential risks of Cd, which belongs to moderate ecological risk highlight the adverse effect it pose to benthic fauna and ecosystem in general.

In a recent study, the spatial and temporal investigation by Manoj and Padhy (2014) showed Cd as the contaminant of chief concern. Cd concentration was noted above its geochemical background value throughout the studied area in both study periods. Its concentration was significantly higher at sites characterized by dominant anthropogenic activities. The authors highlighted that the contamination of sediments of freshwater systems with Cd is increasingly becoming a major problem in developing countries worldwide.

Table 5 Heavy metals concentration (ppm) of fish and periwinkle samples of Benin River

	Co	Cd	Cu	Zn	Mn	Pb	Fe	Ni
C. auratus								
Mean ± SE	2.85 ± 0.63	0.73 ± 0.46	5.46 ± 1.38	79.47 ± 12.54	22.50 ± 3.69	39.46 ± 8.06	98.90 ± 12.29	3.08 ± 1.48
Minimum	1.10	0.00	1.21	30.98	12.25	10.24	81.80	0.02
Maximum	5.54	2.25	10.77	112.75	33.21	66.84	159.73	8.93
T. fuscatus								
Mean ± SE	5.62 ± 0.87	1.00 ± 0.64	62.42 ± 10.21	132.67 ± 23.55	51.41 ± 7.32	55.88 ± 14.28	233.31 ± 25.01	8.49 ± 2.54
Minimum	2.76	0.00	30.72	63.47	23.86	5.27	165.39	1.28
Maximum	8.20	3.18	90.17	235.91	70.53	100.58	333.00	16.52

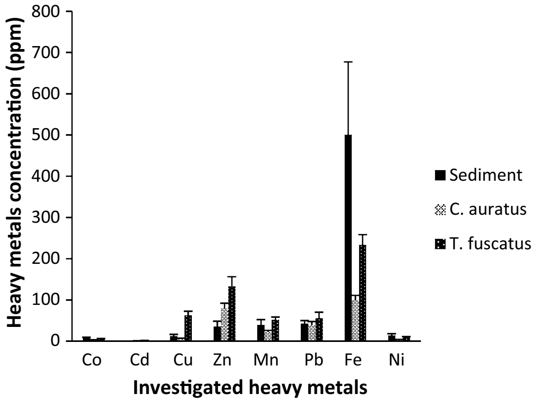

Fig. 6 Mean concentrations of heavy metals in the sediment and benthic fauna

Table 6 Maximum permitted concentrations (mg/kg wet weight) for certain heavy metals in fish

Organization	Cd	Pb	Reference
FAO/WHO limits	0.50	0.50	FAO/WHO (1989)
FAO/WHO limits	–	0.30	JECFA (2011)
European Community	0.05	0.30	EC (2006)

Table 7 Calculated transfer factor metals

	Sediment	*C. auratus*	*T. fuscatus*	Calculated transfer factor	
				C. auratus	*T. fuscatus*
Co	7.28	2.85	5.65	0.39	0.77
Cd	0.56	0.73	1	1.79	1.37
Cu	12.26	5.46	62.42	0.45	5.09
Zn	35.73	79.47	132.67	2.22	3.71
Mn	39.29	22.5	51.41	0.57	1.31
Pb	42.28	39.46	55.88	0.93	1.32
Fe	500.39	98.9	233.31	0.20	0.47
Ni	13.73	3.08	8.49	0.22	0.62

Benthic fauna analysis

Heavy metals in *C. auratus* and *T. fuscatus*

The mean concentration (ppm) of heavy metals in the whole tissue of *C. auratus* and whole soft tissue of *T. fuscatus* in Benin River is presented in Table 5 and Fig. 6. The results showed that *T. fuscatus* had higher mean concentration of heavy metals than *C. auratus*. The heavy metals accumulated by *C. auratus* and *T. fuscatus* were in the order: Fe > Zn > Pb > Mn > Cu > Ni > Co > Cd and Fe > Zn > Cu > Pb > Mn > Ni > Co > Cd, respectively.

It is evident from the analysis of Fig. 6 that *T. fuscatus* (shellfish) accumulated heavy metals than *C. auratus* (finfish) as well as sediment except for Co, Fe, and Ni. Results from Kakulu et al. (1987) also indicated that the levels of Cd, Cu, Zn, Mn, Pb, and Fe were higher in shellfish that in finfish.

The mean concentrations of Cd (0.73 mg/kg) and Pb (98.90 mg/kg) in *C. auratus* observed in this study were compared with FAO/WHO, JECFA and European Community (EC) recommended maximum levels in seafood (Table 6). The result revealed that the concentrations of these heavy metals in *C. auratus* collected from Benin River were higher than the permissible limits for consumption.

Transfer factor

Metal transfer factor from sediment to benthic fauna is viewed as a major pathway of human exposure to heavy metals via food chain. It is an essential tool for investigating the human health risk index (Cui et al. 2004). The calculated transfer factor values (Table 7 and Fig. 7) point out the level of bio-magnification that has occurred in *C. auratus* and *T. fuscatus*, respectively. A transfer factor of 1 and above indicates that the metal is biomagnified (Ibhadon et al. 2014). Except for Cd and Fe, all other transfer factors in *T. fuscatus* were above 1 indicating that there was bio-magnification of the rest heavy metals but in *C. auratus* only Cd and Zn

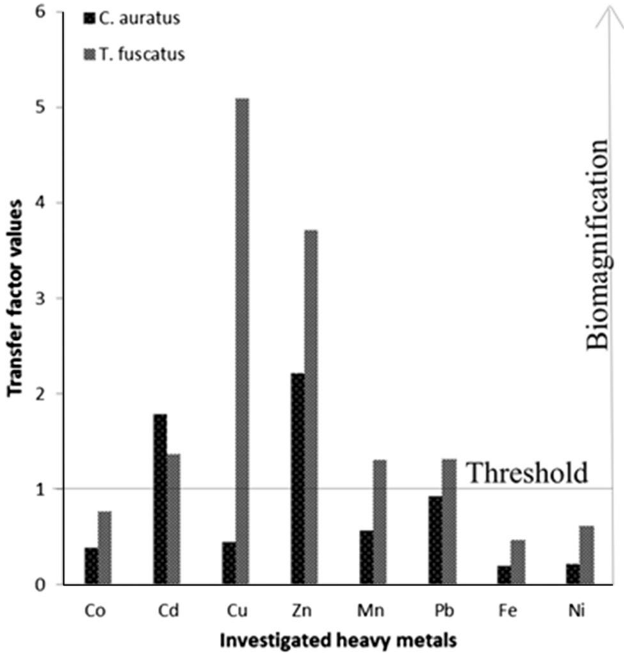

Fig. 7 Transfer factor of heavy metals in benthic fauna

were biomagnified as they were the only investigated heavy metals above 1.

Bioconcentration and magnification could lead to high toxicity of these metals in organisms, even when the exposure level is low. Under such conditions, the toxicity of a moderately toxic metal could be enhanced by synergism and fish population may decline. Apart from destabilizing the ecosystem, the accumulation of these toxic metals in aquatic food web is a threat to public health and thus their potential long-term impact on ecosystem integrity cannot be ignored (Ogoyi et al. 2011).

Health risk assessment

The results of the average daily dose (ADD) and hazard quotient (HQ) for the benthic fauna of Benin River are summarized in Table 8 with corresponding oral reference dose (RfD). The calculated HQ values for the selected heavy metals ranged from 0.1286 to 15.2692 for *C auratus* and from 0.2538 to 21.6274 for *T. fuscatus*.

The human health risk assessment and HQ values for Co, Cd, Cu, Zn, Mn, Fe, and Ni in *C. auratus* and Co, Zn, Mn, Fe and Ni in *T. fuscatus* were less than 1 indicating that there is no obvious health risk from these heavy metals over a lifetime of exposure. However, HQ values for Pb in *C. auratus* and Cd, Cu, and Pb in *T. fuscatus* were above 1 indicating significant health risk for these heavy metals. The hazard index (HI) values for both *C. auratus* and *T. fuscatus* were greater than 1 indicating significant adverse health risk for non-carcinogenic effect. It is important to note that Pb, contributed 87.0% of the non-cancer effects of heavy metals to the HI in the populace in *C. auratus* (Fig. 5) and the other major contributor is Cd (5.7%). Correspondingly, Pb contributed 78.7% of the non-cancer effects of heavy metals to the HI in the populace in *T. fuscatus* (Fig. 6), while other major contributors are Cu and Cd with contributions of 7.7% and 5.0%, respectively.

Table 8 ADD and HQ for studied benthic fauna

Metals	ADD1	ADD2	RfD	HQ1	HQ2
Co	0.0039	0.0076	0.0300	0.1286	0.2538
Cd	0.0010	0.0014	0.0010	0.9911	1.3595
Cu	0.0074	0.0845	0.0400	0.1850	2.1137
Zn	0.1076	0.1797	0.3000	0.3588	0.5990
Mn	0.0305	0.0696	0.1400	0.2177	0.4974
Pb	0.0534	0.0757	0.0035	15.2692	21.6274
Fe	0.1340	0.3160	0.7000	0.1914	0.4515
Ni	0.0042	0.0115	0.0200	0.2084	0.5749
HI	–	–	–	17.5502	27.4772

1 and **2** represent *C. auratus* and *T. fuscatus*, respectively, *ADD* average daily dose, *HQ* hazard quotient, and *HI* hazard index

The human health risk assessment of the present research work was compared with the one reported by Enuneku et al. (2014). Results of HQ and HI were found to be higher than that of Enuneku et al. (2014).

Conclusion

This study was undertaken to investigate heavy metal concentrations in sediments and benthic fauna of Benin River and, most importantly, assess the ecological and human health risk of contaminated sediments and benthic fauna as these delicacies provide relatively cheap source of animal protein to Koko Community inhabitants. The results from this study showed that the contamination of sediment and benthic fauna (*C. auratus* and *T. fuscatus*) of Benin River with heavy metals (Co, Cd, Cu, Zn, Mn, Fe, Pb, and Ni) were largely from anthropogenic sources. In all sediment samples only Pb exceeded the threshold/probable effect level (TEL). The heavy metals under investigation in sediments reflected a low ecological risk to Benin River with an exception for cadmium, which posed a moderate ecological risk to the river. Hence, Cd is considered the most eco-toxic metal in this study. In general, *T. fuscatus* (shellfish) accumulated heavy metals in higher concentrations than *C. auratus* (finfish) as well as sediment suggesting that *T. fuscatus* could be used as bioindicators for heavy metal pollution. Furthermore, the following heavy metals Cd, Cu, Zn, Mn, and Pb in *T. fuscatus*, while Cd and Zn in *C. auratus* were bio-magnification as the calculated transfer factor were above 1. The human health risk assessment showed that HQ values for Pb in *C. auratus* and Cd, Cu, and Pb in *T. fuscatus* indicated significant health risk for these heavy metals. The human health risk assessment showed that HQ values for Pb in *C. auratus* and Cd, Cu, and Pb in *T. fuscatus* indicated significant health risk for these heavy metals. The hazard index (HI) values for both *C. auratus* and *T. fuscatus* were > 1 indicating significant adverse health risk of non-carcinogenic effect. Therefore, the consumption of these contaminated fish and shellfish by the people of Koko portends risks for the health of the public. The industries operating in this community should adopt more sustainable and eco-innovative management options in order to attenuate potential ecological and human health risk of metal pollution.

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