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Metal Contamination and Health Risks in West African Mud Creeper (*Tympanotonos fuscatus* var r*adula*) from Abule-Agele Creek, Nigeria

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Abstract

This study assessed the seasonal distribution of trace metals in soft tissues of *Tympanotonos fuscatus* var *radula*, surrounding water, and sediment of Abule-Agege Creek in Southwest Nigeria. A non-significant variation ($p > 0.05$) in water physicochemical parameters occurred across wet and dry seasons. Metallic contamination (Copper, zinc, mercury, chromium, lead and cadmium) was found to be higher in *T. fuscatus* var *radula* than in water, and the sediment serves as a large depository of these trace metals. In this gastropod, the ability of metal accumulation from water (Bio-water accumulation) was higher than that from sediment (Bio-sediment accumulation). In addition, the linear regression models revealed positive relationship between tissue and sediment concentrations of lead and cadmium for both seasons. The estimated daily intake of investigated metals for both seasons was lower than the oral reference dose, while the target hazard quotient and total hazard index of individual metals were both less than 1, meaning that *T. fuscatus* var *radula* from the study region posed no health risk.

Keywords Bioaccumulation · Food analysis · Food composition · Mollusc · Risk assessment

Mangrove ecosystems consist of diverse communities such as the intertidal zones of tropical and subtropical coastal rivers, estuary habitats etc., that exist in different regions of the world (Marchand et al. 2016). This ecosystem acts as a habitat for resident and migratory animals and plays a part in carbon sequestration and protection against coastal erosion. These functional features make the mangrove unique, which is why mangrove ecosystem pollution has recently gained increased attention from those concerned with conservation issue (Liu et al. 2015). Due to metal distribution, its abundance and persistence in the aquatic ecosystem, the contamination of mangrove ecosystems is on the rise and has become a global problem (Aljahdali and Alhassan 2020). The suspended contaminants in water bodies and those present in land-based effluent, end up in continental shelf and coastal areas. These contaminants have detrimental effects by accumulating in bottom sediments of the mangrove environment and altering its natural status (Bakshi et al. 2018).

Some metals such as Cd, As, Hg, Pb are known as non-essential toxic metals due to their adverse effects on biological organisms, and also recognized as serious environmental pollutants (Heidary et al. 2012). Metallic contaminants are problematic since they are non-biodegradable and may persist in living tissues and food, causing a major threat to both human health and environmental protection (Corrias et al. 2020). Metallic contaminants are low water soluble ions that can cause toxicity and serious side effects on human health. After discharge into water body, sediments normally accumulate trace metals and some of them are biomagnified in tissues of invertebrates (Jitar et al. 2015). There are some benefits of using sedentary herbivorous organisms such as gastropods for trace metal monitoring. Throughout the year, these living organisms are available, simple to gather, sessile, and have enough tissues for analysis (Corrias et al. 2020).

Shellfish such as edible molluscs have become a global delicacy for seafood lovers because of their vital nutrients that are beneficial to human health (Afolayan et al. 2020). *Tympanotonos fuscatus* var *radula* (Linnaeus 1758) is an economically important gastropod mollusc, which serves as a delicacy in many coastal communities of Nigeria. Despite

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the nutritious quality, due to their bottom-feeding strategy, aquatic molluscs have a reputation for being unhealthy. There are two key reasons why metal bioaccumulation in mollusc needs to be constantly studied. One, mollusc is among the widely and preferred eaten seafood and secondly, mollusc has widespread distribution, easy accessibility, and resistance to toxins while playing a significant role in aquatic ecosystem's food chain (Wang and Lu 2017; Ju et al. 2020).

The Southern region of Nigeria has numerous species of shellfish, which are nutritionally important in the supply of protein, the nine essential amino acids and minerals (Moruf et al. 2019). However, due to rapid population growth, this region has experienced typical anthropogenic activities and industrialization at the catchment site. The contamination of aquatic resources with trace metals from untreated households and industrial waste is one of the consequences of such anthropogenic activities, which poses a danger to functionality and raises the ecological risk of the aquatic ecosystem (Ayanda et al. 2019).

Many studies have implicated moluscs as a source of trace metals in dietary intake by the use of Inductively Coupled Plasma Mass Spectrometry (ICP-MS) and Atomic Absorption Spectroscopy (AAS) technique (Davies et al. 2007; Chinda et al. 2009; Afolayan et al. 2020; Etuk et al. 2020; Moruf and Durojaiye 2020). ICP-MS offers extremely lower detection limit compared to AAS. Using ICP-MS technique, this study assessed the concentration of trace metals in water, bottom sediment and gastropod tissues (*T. fuscatus* var *radula*) across Abule-Agege Creek in Lagos, Nigeria, with respect to bioaccumulation and potential health risk to consumers.

Materials and Methods

Abule-Agege Creek is a brackish water creek located between latitudes 6° 26′–37′ N and longitude 3° 23–4° 20′E on the highly populated western axis of the Lagos Lagoon (Moruf et al. 2018). Site selection (Fig. 1) was based on increasing anthropogenic effects from heaps of domestic and solid waste dumps.

Monthly collection of samples was carried out across seasons between May 2018 and April 2019. Water and bottom sediment (at a depth of 2 cm) were collected using bottles with glass stoppers and Van Veen Grab (wt. 25 kg; height—20 cm) respectively. Six representative samples of water and sediment, each from different sampling station were taken. In total, one hundred and sixty seven (167) adult specimens of *T. fuscatus* var *radula* were collected by scooping from the waterbed at low tides. All samples were initially kept in an ice chest and later taken to the laboratory for analysis. The examined gastropod samples ranged in carapace length from 1.4 cm to 12.1 cm and in weight from 1.1 to 23.7 g.

Water temperature and dissolved oxygen were measured in-situ using a mercury-in-glass thermometer and Lutron DO meter (Model: DO 5519) respectively. Separate water samples were collected in 250 mL dissolved oxygen bottles at each station and incubated in the dark for five days for biochemical oxygen demand (BOD) determination as described in APHA (2005), while chemical oxygen demand (COD) was determined using Titrimetric method. At the laboratory, the sediments were defrosted by keeping them at room temperature for about 24 h, then dried in an oven at 40°C (it has been proved by Gilli et al. 2018 that samples drying

Fig. 1 Map showing the study area (Red dots indicating the study site)

at 40°C does not cause evaporation losses of Hg), disaggregated and sieved through a 200 µm sieve. The sieved samples were subsequently homogenized in a porcelain mortar and re-sieved. Approximately 5 g of the samples was put in Teflon tubes, and 5 mL aqua regia (HCl: HNO_3 in a ratio of 3:1) added for digestion, following the ISO 11466 digestion method (Pueyo et al. 2001).

Sub-samples of approximately 1 g tissue were weighed in a precision scale with decimal resolution (0.001 g) and digested in a mixture of 5 mL of concentrated nitric acid (TMA, Hiperpure, PanReac, Spain) and 3 mL of 30% w/v hydrogen peroxide (PanReac, Spain) in a microwave-assisted digestion system (Ethos Plus; Milestone, Sorisole, Italy). Digested samples were transferred to polypropylene sample tubes and diluted to 15.0 mL with ultrapure water. As described by Dussubieux and Van Zelst (2004), the determination of the non-essential elements copper, zinc, chromium mercury, lead and cadmium in all the samples was carried out by ICP-MS. ICP-MS-based multi-element determination was performed in an Agilent 7700 × ICP-MS system (Agilent Technologies, Tokyo, Japan) equipped with collision/reaction cell interference reduction technology. The continuous sample introduction system consisted of an autosampler, a Scott double-pass spray chamber (Agilent Technologies, Tokyo Japan), a glass concentric MicroMist nebuliser (Glass Expansion, West Melbourne, Australia), a quartz torch and nickel cones (Agilent Technologies, Tokyo Japan). Elemental concentrations were quantified using a MassHunter Work Station Software for ICPMS (version A.8.01.01 Agilent Technologies, Inc. 2012, Tokyo, Japan). (Luna et al. 2019). Analytical quality control was applied throughout the study. Blank values were processed alongside samples, and the values obtained were subtracted from sample readings before the final results were calculated. The limits of detection (LOD) were calculated as three times the standard deviation of the reagent blanks and were based on the mean sample weight analysed. In all cases, the LODs obtained were low enough to determine all trace metals at the common levels in the samples analysed (Minervino et al. 2018). The accuracy of determination was evaluated by comparison with the analytical recoveries determined in certified reference materials (fish protein DORM- 3 National Research Council, Ottawa, Ontario, Canada) analysed following exactly the same procedure as for the samples.

Bioaccumulation factor (BAF), which is the ratio of the concentration of metal in organism tissue to the concentration of metal in sediment (Bio-sediment accumulation) and water (Bio-water accumulation), was calculated.

Health risk was estimated based on Environmental Protection Agency guidelines (EPA 2005). To assess the potential health risk via the consumption of the *T. fuscatus* var *radula*, the estimated daily intake (EDI), target hazard quotient (THQ) and target hazard index (THI) were calculated using Eqs. 1, 2, and 3 respectively with the following assumptions:

- The hypothetical body weight for adult Nigerian was 70 kg (Agwu et al. 2018).
- The maximum absorption rate was 100% while the bioavailability factor was also 100% (ATSDR 2005).

$$\textit{Estimated Daily Intake, EDI}(mg/kg/day) = \frac{\mathrm{C} \times \mathrm{CR} \times \mathrm{AF} \times \mathrm{EF}}{BW} \tag{1}$$

where

C = Concentration of the contaminant in the exposure pathway (mg/kg) of *T. fuscatus* var *radula.*

CR = Consumption Rate; Nigeria aquatic mollusc taken/day, 0.0366 kg/day = 13.359 kg/y.

AF = Bioavailability factor (100%).

EF = Exposure Factor = 1.

Bw = Body weight (70 kg) (Wang et al. 2005)

$$\textit{Target Hazard Quotient, THQ} = \frac{\mathrm{EDI}}{\mathrm{RfD}} \tag{2}$$

where

EDI= Estimated Daily Intake,

RfD = the oral reference dose (mg/kg/day), (Wang et al. 2005)

$$THI = \sum_{i=1}^{n} \mathrm{THQ} \tag{3}$$

where THQ_i is the target hazard quotient of an individual trace metal.

The mean, standard deviation, range and correlation factors were determined with Microsoft excel 2010 software. Box plots, one-way ANOVA and PCA were performed with R-studio (version 3.5.2). The statistical criterion for significance was chosen at $p < 0.05$.

Results and Discussion

The water physicochemical values measured at the study stations during the study period are presented in Fig. 2. The results agree with Ibanga et al. (2019) who found that physicochemical factors varied with seasons and locations. Although temperature was higher in the dry season (27.92°C) than in the wet season (24.35°C), there was no significant difference ($p > 0.05$) in the variation. The range of temperatures recorded (25.35–27.92°C) is also considered normal with reference to the creek locations in the Lagos Lagoon (Nwankwo et al. 2013). The decrease or increase in water temperature depends mainly on the climatic conditions, sampling times, sunshine hours and

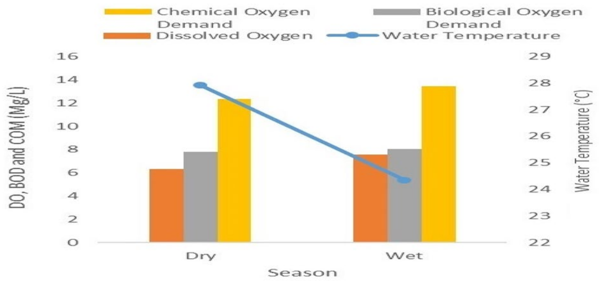

Fig. 2 Physicochemical parameters of Abule-Agege Creek, Nigeria

specific characteristics of water environment such as turbidity, wind force, plant cover and humidity (Ahmed et al. 2017).

The DO, BOD and COD were generally higher during the wet season with the mean values of 7.54 mgL^{-1}, 8.02 mgL^{-1} and 13.45 mgL^{-1} respectively. The reduced concentration of the dry season DO could be due to enormous amount of organic loads that required high levels of oxygen for chemical oxidation, decomposition or break down. In addition, higher DO recorded during the wet season may be due to the low temperature and high run-offs experienced at the study location. According to Moshood (2008), DO is important as a respiratory gas and it acts as a water quality indicator as well as an indicator of health and productivity of a river. The BOD values were lower than the permissible limit of 50 mg/L for coastal water (FMENV 2001). The higher COD values recorded may be due to chemical oxidation of some organic substances. The recorded values are comparable to the results of Lawal-Are et al. (2019) who reported mean DO of 5.3 ± 0.1 mgL^{-1}, BOD of 5.1 ± 0.6 mgL^{-1} and COD of 19.0 ± 3.9 mgL^{-1} for Abule-Eledu Creek.

Figures 3, 4, 5 depict box and whisker plots showing trace metal concentrations in water, sediment and *T. fuscatus* var *radula* in relation to season. In Fig. 3, cadmium and chromium concentrations in water showed a narrow spread while the concentrations of other investigated trace metals were moderately spread across seasons. The result revealed that the trace metal concentrations in water ranged from 0.009 mgL^{-1} in cadmium to 0.142 mgL^{-1} in copper in the dry season while 0.007 mgL^{-1} and 0.110 mgL^{-1} in cadmium and copper respectively, in the wet season.

In Fig. 4, copper, zinc, mercury and chromium distribution in sediment showed a wide spread while lead and cadmium had a narrow spread for both seasons. The trend of trace metals in sediment during the dry season was in the order: zinc > chromium > copper > mercury > lead > cadmium. In the wet season, a similar but inconsistent and relatively lower concentration was also observed in the study area for the metals: zinc > chromium > mercury > copper > lead > cadmium. According to Moruf and Akinjogunla (2019), sediment is the major metal depository, containing more than 99 percent of the total amount of metal found in the aquatic environment in some cases. A deeper insight into the long-term contamination state of the aquatic ecosystem can be calculated when the sediment trace metals are studied (Yau and Gray 2005; Ogbeibu et al. 2014). In general, sediment of Abule-Agege Creek has low enrichment of trace metals, which may be due to elevated tidal flushing in the area.

In Fig. 5, zinc, chromium, lead and cadmium showed a narrow distribution in *T. fuscatus* var *radula* while copper and mercury showed a wider distribution across seasons. Copper (0.54–0.66 mg kg^{-1}) recorded the highest mean concentrations in both seasons. Cadmium (0.03 mg

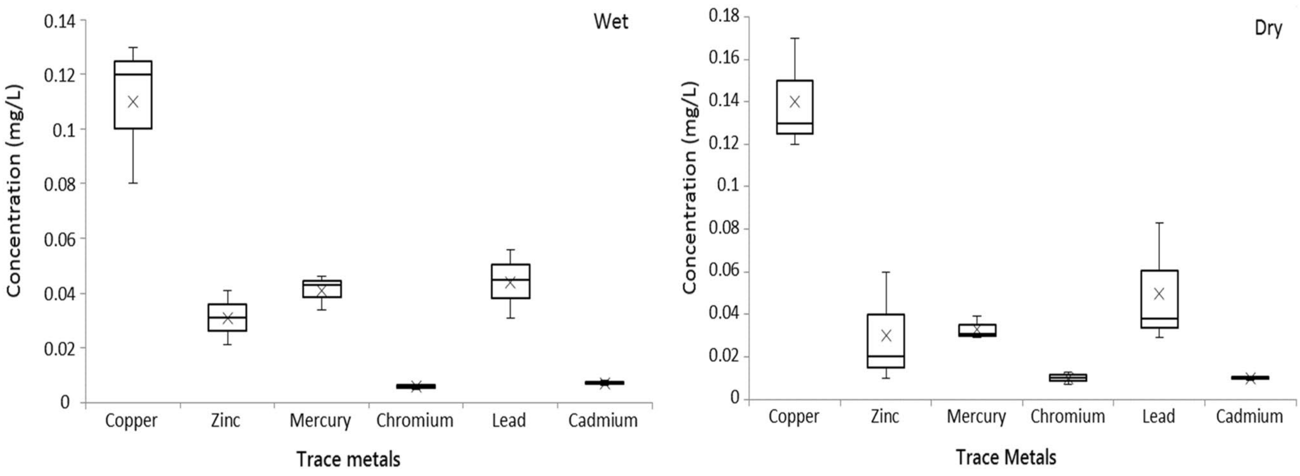

Fig. 3 Box plot for seasonal distribution of water trace metals in Abule-Agege Creek, Nigeria (Lower and upper box boundaries 25th and 75th percentiles, respectively, line inside box median, lower and upper error lines 10th and 90th percentiles, respectively, filled circles data falling outside 10th and 90th percentiles)

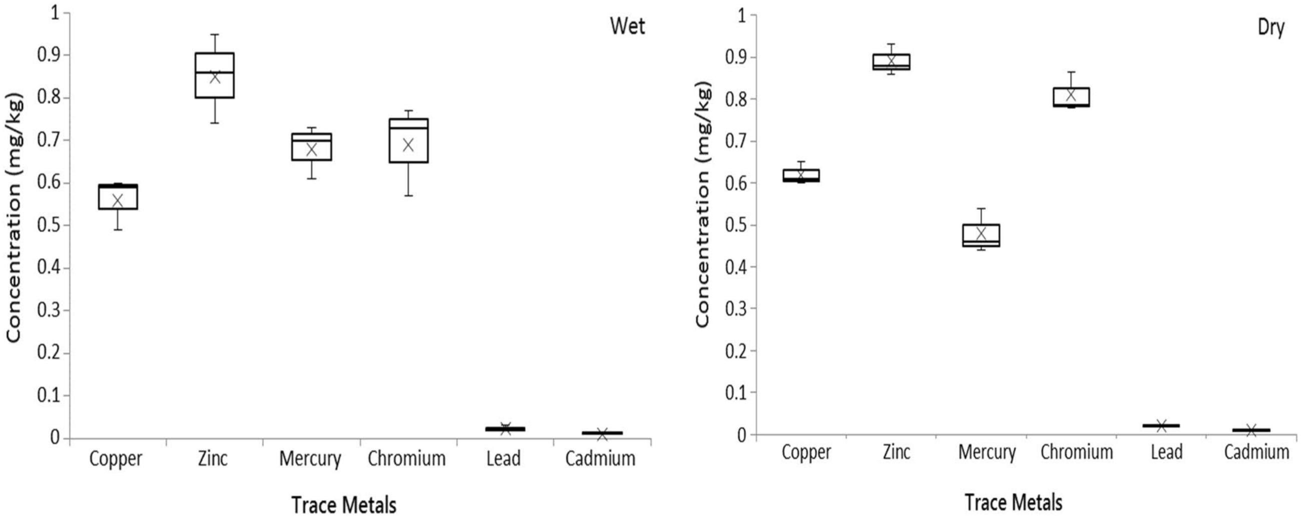

Fig. 4 Box plot for seasonal distribution of sediment trace metals in Abule-Agege Creek, Nigeria (see Fig. 3 caption for explanation of box-plot)

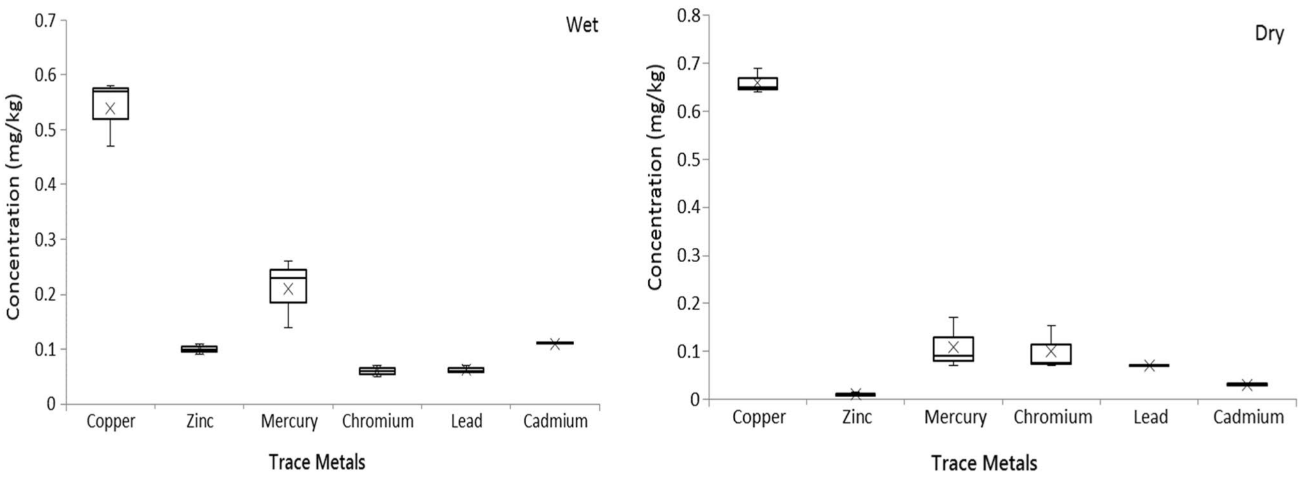

Fig. 5 Box plot for seasonal distribution of trace metals in *T. fuscatus* var *radula* (see Fig. 3 caption for explanation of box-plot)

kg^{-1}) and lead (0.06 mg kg^{-1}) recorded the lowest mean concentrations for dry season and wet season respectively. The higher levels in *T. fuscatus* var *radula* compared with water can be attributed to biological accumulation. Similarly, Etuk et al. (2020) reported high copper concentration (11.096 ± 0.84–33.143 ± 1.09 mg kg^{-1}) in periwinkle from Cross River Estuary. According to Abdel-Mohsien and Mahmoud (2015), elevated trace metal concentrations in aquatic organisms reflect accumulative exposure through water and/or food.

The bioaccumulation factors of trace metals in the sampled gastropods are presented as Bio-water accumulation factor (BWAF) and Bio-sediment accumulation factor (BSAF) in Figs. 6 and 7 respectively. All investigated trace metals were observed to bioaccumulate in measurable concentrations in the organism across seasons. Significant

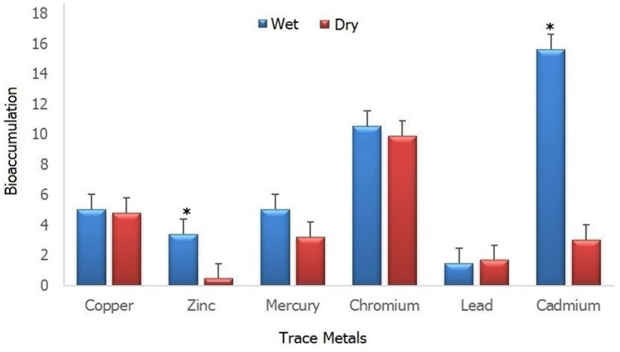

Fig. 6 Bio-water accumulation factor of *T. fuscatus* var *radula* in Abule-Agege Creek

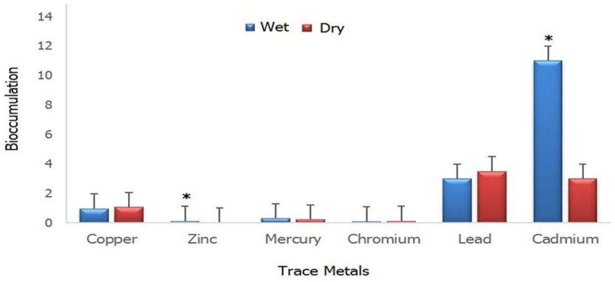

Fig. 7 Bio-sediment accumulation factor of *T. fuscatus* var *radula* in Abule-Agege Creek

Table 2 Regression analysis between trace metal concentrations in *T. fuscatus* var *radula* and sediment during wet season

Metal	Regression equation	Coefficient of determination (R^2)
Copper	$Y = -0.021 + 6.230\ X$	0.731
Zinc	$Y = 0.054 + 0.054\ X$	0.245
Mercury	$Y = -0.470 + 3.421\ X$	0.612
Chromium	$Y = -0.002 + 0.089\ X$	0.675
Lead	$Y = 0.04 + 0.930\ X$	0.508
Cadmium	$Y = 0.1 + 0.661\ X$	0.532

higher BWAF and BSAF were recorded for zinc and cadmium during the wet season. The recorded BSAF values were greater than one (1) for lead (3.0–3.5) and cadmium (3.0–11.0) while other investigated metals were less than one (1). The high BAF values of these two trace metals in spite of their low concentration in sediment reveal their high bio-magnification abilities. The seasonal variation in the values of BWAF and BSAF in this study may be attributed to various factors that affect metal uptake in *T. fuscatus* var *radula* as listed by Chindah et al. (2009) to include age, developmental stages, feeding mode, metabolic activities and reproductive conditions. The result for BSAF in this study corroborated the findings of Davies et al. (2007) who reported that the BAF values decreases with increasing metal concentration in sediments.

The prediction of pollutant concentrations in tissues of *T. fuscatus* var *radula* for dry and wet seasons is shown in Tables 1 and 2 respectively. Higher variation was observed in the dry season compared to the wet season. In the dry season, positive correlations exist between the tissue and sediment concentrations of copper, lead and cadmium with respective regression coefficient (*b*) of 0.723, 0.671 and 0.582. The unit decrease for the concentrations of zinc, mercury and chromium in the tissue attributable to the pollutant in the sediment were 0.112, 0.824 and 0.597 respectively. Also, the percentage variation as revealed by the coefficient of determination (R^2) indicates that 69.8%, 91.1% and 89.7% of the variation in the concentration of zinc, mercury and chromium in *T. fuscatus* var *radula* is attributable to the sediment respective metal loads while the variation for copper, lead and cadmium were 32%, 76% and 87% respectively. In the wet season, positive correlation between the tissue concentration and concentration in the sediment was recorded for zinc, lead, cadmium while copper, mercury and chromium recorded negative correlations. Furthermore, 73.1% of copper, 24.5% of zinc, 61.2% of mercury, 67.5% of chromium, 50.8% of lead and 53.2% of cadmium in *T. fuscatus* var *radula* were predicted by the concentration of the respective metals in the sediment. This result is comparable to the findings of Etuk et al. (2020), who reported a linear regression models with positive relationships between concentrations of metals in sediment and periwinkle tissues from the Cross River Estuary in Niger Delta, Nigeria.

Table 1 Regression analysis between trace metal concentrations in *T. fuscatus* var *radula* and sediment during dry season

Metal	Regression equation	Coefficient of determination (R^2)
Copper	$Y = 0.04 + 0.723\ X$	0.320
Zinc	$Y = -0.079 + 0.112\ X$	0.698
Mercury	$Y = -0.37 + 0.824\ X$	0.911
Chromium	$Y = -0.71 + 0.597\ X$	0.897
Lead	$Y = 0.05 + 0.671\ X$	0.760
Cadmium	$Y = 0.02 + 0.582\ X$	0.870

The EDI of the trace metals via the consumption of *T. fuscatus* var *radula* in both wet and dry seasons is presented in Table 3. The result in milligram per body weight per day revealed that the EDI of the investigated metals for both seasons were lower than their respective oral reference dose (RFD). The result corroborated the findings of Moruf and Durojaiye (2020), who determined the EDI of copper, zinc, chromium and mercury for the edible molluscs from Nigeria. In the present study, the EDI values were within the recommended range of FAO/WHO (2004).

Both the THQ and the THI of individual metal are presented in Table 4. The highest THQ value was recorded for mercury (0.115–0.219) while the least value was recorded for zinc (0.00001–0.00002) across seasons. The trend of the THQ for the wet season followed the pattern of mercury > cadmium > lead > copper > zinc > chromium, while that of the dry season was mercury > cadmium > lead > copper > chromium > zinc. Korkmaz et al. (2019) reported higher THQ for copper and zinc in edible mollusc species marketed in Mersin, Turkey. In the present study, the THQ for all the investigated metals and the THI in both dry and wet seasons were less than one (1), suggesting no considerable health hazard via the consumption of *T. fuscatus* var *radula* from the study area.

Table 3 Estimated daily intake (mg/kg/day) of trace metals in *T. fuscatus* var *radula*

Metal	Wet			Dry			Oral reference dose(RfD)
	Min	Max	Mean + SE	Min	Max	Mean + SE	
Copper	0.00025	0.00030	0.00028 ± 0.00	0.00034	0.00036	0.00035 ± 0.00	0.04
Zinc	0.00005	0.00006	0.00005 ± 0.00	0.00001	0.00001	0.00001 ± 0.00	0.30
Mercury	0.00007	0.00014	0.00011 ± 0.00	0.00004	0.00009	0.00006 ± 0.00	0.0005
Chromium	0.00003	0.00004	0.00003 ± 0.00	0.00004	0.00008	0.00005 ± 0.00	1.50
Lead	0.00003	0.00004	0.00003 ± 0.00	0.00004	0.00004	0.00004 ± 0.00	0.004
Cadmium	0.00006	0.00006	0.00006 ± 0.00	0.00002	0.00002	0.00002 ± 0.00	0.001

Table 4 Target hazard quotient (THQ) and total hazard index (THI) of trace metals via the consumption of *T. fuscatus* var *radula* from Abule-Agege Creek in Lagos, Nigeria

	Wet season	Dry season
Copper	0.00706	0.00863
Zinc	0.00001	0.00002
Mercury	0.2194	0.11503
Chromium	0.00002	0.00003
Lead	0.00815	0.00915
Cadmium	0.05769	0.01569
Total hazard index	0.29269	0.14855

In conclusion, the physicochemical variables of Abule-Agege Creek differ with season, although not to a significant level. The study has shown various concentrations of trace metals in Abule-Agege water/sediment and the level of accumulation varied in *T. fuscatus* var *radula* for both seasons. The higher trace metal concentrations in *T. fuscatus* var *radula* compared with water can be attributed to biological accumulation while the sediment acts as a major depository of the metals. All examined trace metals were observed to bioaccumulate in measurable concentrations in the organism across seasons. In this gastropod, the ability of metal accumulation from water (Bio-water accumulation) was higher than that from sediment (Bio-sediment accumulation). In addition, the linear regression models revealed positive relationships between the tissue and sediment concentrations of lead and cadmium for both seasons. Estimated Daily Intake of the investigated metals for both seasons were lower than the oral reference dose while the target hazard quotient and the total hazard index of individual metal via the consumption of *T. fuscatus* var *radula* by average Nigeria adults were less than 1, suggesting no considerable health hazard in consuming of *T. fuscatus* var *radula* from the study area.

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