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Trophodynamics and biomagnification of trace metals in aquatic food webs: The case of Rufiji estuary in Tanzania

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ABSTRACT

Transfer of trace metals into fish of commercial value poses a public health concern. Therefore, sediments, invertebrates, and fish from three sampling sites in the Rufiji estuary were analyzed for trace metals to evaluate their concentrations and trophic transfer within estuarine food web. Stable isotopes of carbon and nitrogen were also used to study the trophic relationship between different organisms. Biomagnification of trace metals in organisms from different trophic levels was quantified and evaluated by calculating the bioaccumulation factor and biomagnification factor. Trophic magnification factor for different trace metals was determined from the slope of the regression line between trace metal concentration and the trophic level of functional groups in sampled organisms. The results indicated that As and Zn displayed trophic level-dependent accumulation in the Rufiji food webs. As and Zn increased with the trophic level, whereas Ag, As, Co, Cr, Cu, Mn, Ni, and Pb depicted an opposite trend. Food web magnification factors varied from −0.57 for Ni to 0.39 for Zn, whereas trophic magnification factor varied from 0.27 for Ni to 2.47 for Zn. Zn and Ni bioaccumulate in the food webs as indicated by a slope greater than zero, whereas the remaining trace metals are eliminated from food webs or their trophic transfer is interrupted.

1. Introduction

Trace metal pollution in the environment has resulted in global concerns in the scientific community. In marine environments, trace metal pollution can result from point and/non-point sources (Birch et al., 1996), atmospheric deposition (Berg and Steinnes, 1997), and continental erosion (Duman et al., 2007). These metals are ingested and they accumulate in animal tissues or transported to higher trophic levels through the food chain (Xie et al., 2010; Chen et al., 2011; Zhang and Wang, 2012). Consequently, organisms in marine environments can be exposed to higher levels of these contaminants causing adverse health effects and various complications (Bird et al., 2008; Cai et al., 2009; Volpe et al., 2009; Coeurdassier et al., 2010).

Traditional analyses such as direct field observations and gut content can be used to evaluate trophic relationships (Pinnegar and Polunin, 2000), but they show lot of variability. Hence, stable isotope analysis, which reflects the whole diet history of a species, is commonly used to quantify the relative trophic positions for different species (Bucci et al., 2007; McIntyre and Beauchamp, 2007; Bond, 2010). Naturally occurring tracers of stable isotope ratios of carbon ($\delta^{13}C$) and nitrogen ($\delta^{15}N$) have been recommended to investigate the trophic relationships and biomagnification of different contaminants (Bucci et al., 2007). For example, $\delta^{15}N$ values are effective in quantifying the trophic position because enrichment of ^{15}N occurs incrementally across trophic levels at a constant rate (3–4‰; McCutchan et al., 2003). Although enrichment of ^{13}C is not obvious, it is considered a valuable marker for identifying the different sources of primary production, contributing towards ~1‰ shift between the trophic levels (Hobson et al., 2002; Hoekstra et al., 2003). Hence, $\delta^{15}N$ and $\delta^{13}C$ isotope trends have been extensively used to elucidate trophodynamics and biomagnification of various contaminants in aquatic food chains (Borga et al., 2001; McIntyre and Beauchamp, 2007; Monferran et al., 2016).

Mangroves located in intertidal areas of the coastal region are exposed to trace metal pollution due to various human activities like mining, smelting and processing of metal ores, burning of fossil fuels, pesticides, and domestic and industrial sewage inputs (Anouti, 2014). Mangrove sediments have a high capacity for binding trace metals because they have an anaerobic nature with rich sulfide content that can sorb various metals and increase their concentrations in mangrove sediments. Although mangrove forests have the ability to tolerate high levels of trace metals (MacFarlane et al., 2007), availability of these metals lead to bioaccumulation in plant tissues (Lotfinasabasl and

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Gunale, 2012). In particular, trace metal pollution can damage the biodiversity in marine ecosystems because of their tendency to bioaccumulate in the marine food chain (Chakraborty et al., 2014). Thus, trace metal bioaccumulation is related to their concentration in the surrounding environment and its exposure to the biota. In addition, other factors like salinity, pH, hardness, and temperature are also responsible for accumulation of trace metals, which affects its distribution in coastal areas.

The Rufiji mangrove ecosystem is a dynamic environment of key socio-economic importance and comprises of biologically diverse flora and fauna sensitive to regional climate change, hydrology, and anthropogenic activities. This mangrove ecosystem has partly been impacted by urban development and agricultural activities in the catchment, and pollution in upstream sections of the Rufiji River (Minu et al., 2018). The Rufiji River is one of the largest rivers in East Africa and drains ca. 20% of mainland Tanzania (Erftemeijert and Hamerlynck, 2005). The river has significant potential for transporting diverse pollutants from its catchment into the Rufiji delta, which supports the largest mangrove ecosystem in East Africa (Richmond et al., 2002). The mangrove sediments are clayey silts and silty clays with high organic matter content. This makes mangrove sediments both a good sink and/or source for various metal-bound particles actively participating in different biogeochemical reactions (Marchant and Hooghiemstra, 2004, 2007). The livelihood of many riparian communities depends on fishery and other interdependent activities within the Rufiji delta. Hence, investigations on the status of trace metal pollution within the ecosystem, trophodynamics, and biomagnification of trace metals in the food chain are imperative in order to implement habitat protection and appropriate conservation measures. While recent studies have investigated the distribution of trace metals in the Rufiji mangrove forests (Minu et al., 2018), there is little information on trophodynamics and biomagnification of trace metals. Hence, the objectives of this study are to 1) study the distribution of various priority trace metals (Ag, As, Cd, Co, Cr, Cu, Pb, Mn, Ni, and Zn) in the Rufiji estuary, and 2) evaluate and quantify presence of trace metals within a telescoping aquatic food web including sediments, seaweed, polychaetes, gastropods (*Terebralia* sp., *C. cucullata*), crab (*Uca* sp.), penaeid shrimps (Penaeus *monodon)*, teleost (*Hilsa kelee*, *Trichiurus lepturus, Arius thalassinus, Valamugil buchanani)* by calculating the bioaccumulation factor (BAF) and biomagnification factor (BMF). The new data generated will contribute towards identification of potential biomonitors and provide useful information for guiding suitable management plans as part of environmental remediation initiatives in the Rufiji estuary.

2. Materials and methods

2.1. Study sites

The study sites are located in the Rufiji estuary, 100 km south of Dar es Salaam city, in the Rufiji River delta (Fig. 1). This river has a catchment area of 177,400 km^2 and is over 640 km long (UNEP, 2001). About 30 km from the coast, the lower Rufiji River branches out into a series of channels forming a large delta, covering approximately 1200 km^2 of which 530 km^2 is covered by dense mangrove forests (Sørensen, 1998). The Rufiji estuary is an integral part of the largely undisturbed saline swamps, tidal marshes, woodland, and mangrove forests. It is by far the largest delta in eastern Africa and contains the largest contiguous block of mangrove forests on the eastern seaboard of the African continent. The four northern tributaries carry the bulk of sediments discharged from the catchment into the Rufiji River. The southern part of the delta is sheltered, and receives little freshwater input. Water in front of this part of the delta is clear and saline and features seagrass meadows and scattered reefs. Tidal influence in the delta is considerable (REMP, 2003). In the upstream part, some small towns are located in the catchment and people depend on agriculture and small industries as means of livelihood. The downstream part of the river is free of industries. The soil type in the delta is mostly black loamy soil, which is fertile and suitable for agriculture (Minu et al., 2018). The mangroves have been partially logged for firewood and agriculture. In particular, various pesticides are used for rice cultivation in the mangroves (Taylor et al., 2003).

Eight mangrove species occur in the Rufiji forests, of which, *Rhizophora mucronata*, *Avicennia marina*, and Heritiera littoralis are the dominant varieties. Other species include Bruguiera gymnorrhiza, Ceriops tagal, Lumnitzera racemosa, Sonneratia alba, and Xylocarpus granatum (Semesi, 2002). Other dominant habitats in the estuary consist of tidal marshes, seagrass meadows, and mud flats. Several omnivorous crustaceans of commercial importance spend part of their life cycle in these mangroves feeding on foliar detritus. In addition, live benthic microalgae, and occasionally small animals are also found in the mangroves (Taylor et al., 2003).

The present study was carried out in three locations (Muhoro – station 1, Msomeni – station 2, and Kalale – station 3), representing the south, middle, and northern parts of the Rufiji delta (Fig. 1). These locations were chosen because they are representative of the entire estuary and include a variety of adjacent habitats (i.e. mangrove stands, mud flats, and seagrass meadows) with sufficient diversity and density of consumer organisms.

2.2. Sample collection

Ethical clearance was sought before beginning the study because it involved both flora and fauna that were collected and analyzed for various assays. Clearance for experiments involving animals was obtained from the Directorate of Research and Consultancy at the University of Dar es Salaam. The animals were euthanized prior to laboratory testing according to pre-approved protocol. All flora, fauna, and sediment samples were collected from three locations at the Rufiji estuary in June and July 2014 as part of a reconnaissance survey. Three replicates of each sample were collected. The main primary producers (*Ulva* sp. and *Padina* sp.) and seagrass blades (*T. hemprichii*), and a range of animals including polychaetes, gastropods (*Terebralia* sp.), clams (*Crassostrea cucullata*), crabs (*Uca* sp.), and Penaeid shrimps (*Penaeus monodon*) (see Table 2) were collected.

The consumers under investigation are the dominant species at respective stations within the Rufiji Delta (REMP, 2003). Ten to twenty individuals of gastropods, bivalves, crabs, and shrimps were dissected to remove muscle tissues, and used for preparing the dry tissue pool. Six individuals of teleost (*H. kelee, T. lepturus, A. thalassinus,* and *A. Pectoralis*) species were randomly selected from the total catch of each sample and they were euthanized in an ice slurry. Muscle tissues (after removing the skin) were collected from both sides of each fish, for biochemical analyses within 24 h of sampling. Sample details for primary producers, invertebrate species, crabs, shrimps, clams, and four fish species are indicated in Table 2. Sediment samples from all three sampling sites were collected at depths varying from 0 to 20 cm. These samples were then pooled and homogenized within polyethylene bags and transported to the laboratory for further processing. In the laboratory, all samples (primary producers - *Ulva* sp., *Padina* sp. and *T. hemprichii*, and a range of animals, including polychaetes, gastropods - *Terebralia* sp., clams - *C. cucullata*, crabs - *Uca* sp. and Penaeid shrimps - *P. monodon*) were kept frozen at −20 °C. Samples were then freeze-dried at −50 °C, milled in an agate mortar, and stored in clean dry plastic containers for further testing and analyses.

2.3. Trace metal analysis

The dry homogenized powdered sediments and biota samples were sieved through a nylon mesh to obtain particles smaller than 0.2 mm in diameter for determination of trace metals. Samples were digested following the US EPA method (EPA, 1996) using an Automatic Microwave Digestion System (Ethos Milestone Pro-24). 0.5 g of dry powdered

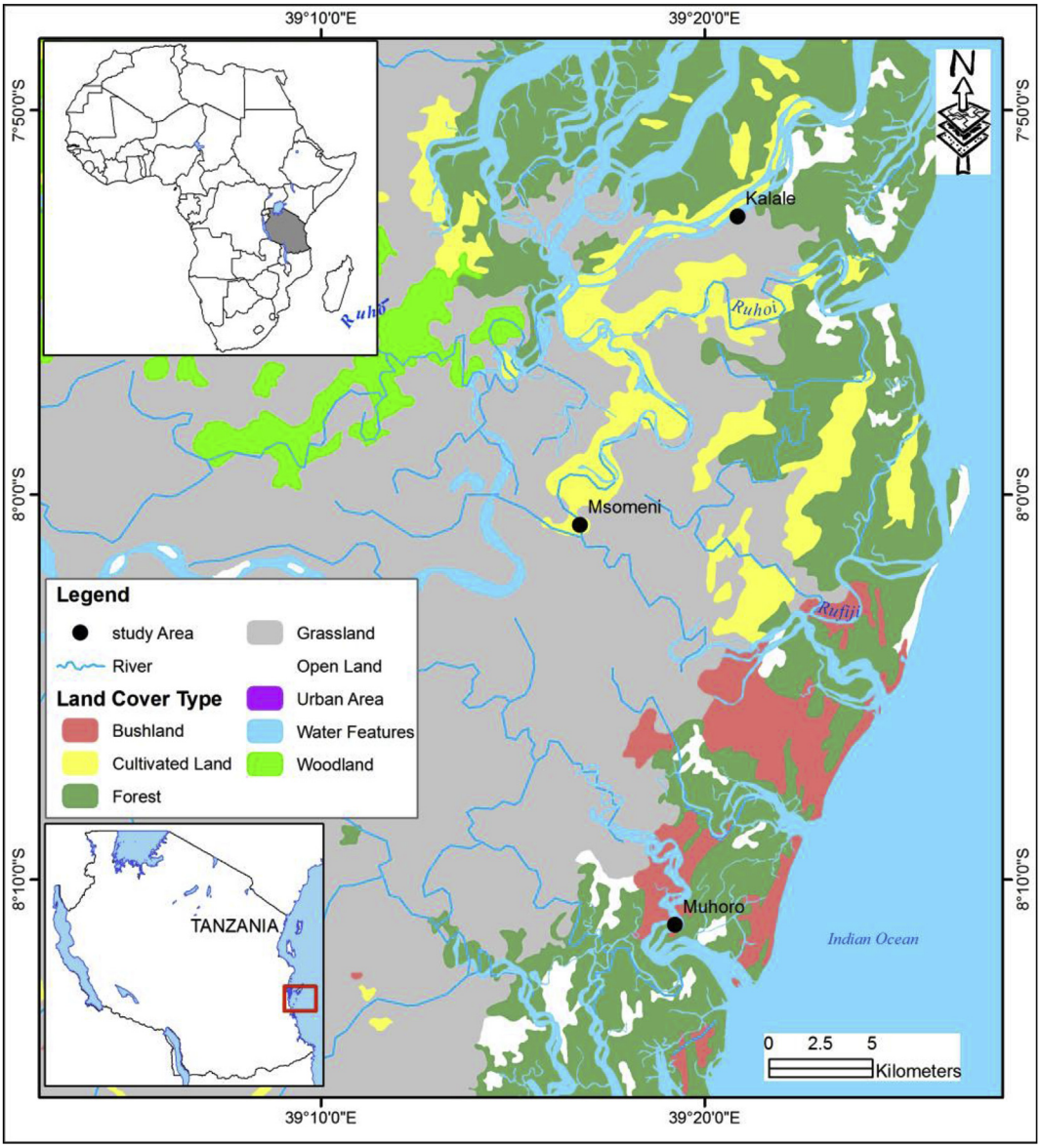

Fig. 1. Map of the Rufiji estuary, Tanzania (adapted from Shilla and Routh, 2018) showing the different sampling sites (Muhoro is station 1; Msomeni is station 2; Kalale is station 3).

Table 1
Two-way PERMANOVA for differences in $\delta^{13}C$ and $\delta^{15}N$ values amongst different aquatic species in the Rufiji estuary.

Source	df	SS	MS	Pseudo-F	P(perm)
Taxa	10	211	21.1	29.6	0.001
Residue	22	15.7	0.72		
Total	32	227			

and sieved sediment samples were weighed in a Teflon vessel and 9 ml of concentrated HNO_3 (65%) and 3 ml of HCl (36.5%) were added and thoroughly mixed. The Teflon vessels were heated to 240 °C over 35 min and then held at 210 °C for at least 15 min in the microwave. The samples were cooled and then filtered to remove the insoluble material before diluted to 50 mL with deionized water. The concentrations of Ag, As, Cd, Cr, Co, Cu, Pb, Mn, Ni, and Zn in the resulting solution were determined on a Perkin Elmer ICP-MS NexION 300D. Quality assurance and quality control were performed by analyzing standard reference materials (PACS-2 marine sediment, Svalvard Rock-1, IAEA organic rich soil, and 350 Tuna fish tissue No. 290). Reagent blanks were digested and analyzed in parallel with each batch of samples. Recovery rates of metals in the standard reference materials ranged between 88 and 110%. Trace metal concentrations in the blanks were < 1% of samples and the relative standard deviation in replicate samples was < 10%.

2.4. Stable isotope analysis

Lipids were extracted from samples with methanol for 12 h in order to reduce the variability caused by isotopically lighter lipids. Samples were further dried at 80 °C for 4 h: 0.5 mg of dried powdered samples and the reference material were weighted into 8 × 5 mm tin capsules and loaded on a Europa Scientific Elemental Analyzer connected to an Isotope Ratio Mass Spectrometer (EA-IRMS). The samples were combusted at 1000–1050 °C and the N_2 and CO_2 gases produced were separated by a packed column gas chromatograph. The gases separated were analyzed using the Europa Scientific 20-20 isotope ratio mass spectrometer (Sercon Ltd. UK).

The stable isotope abundance (δ) was calculated as:

Table 2
Sample details of organisms from the Rufiji estuary.

Names	$\delta^{13}C \pm SD$	$\delta^{15}N \pm SD$	Nd[x]	Na[y]	Body weight (g)		Body length (cm)		TL[z]
					Min-Max	Mean ± SD	Min-Max	Mean ± SD	
Primary producers									
Padina sp.	−13.2 ± 0.50	7.60 ± 0.20	3	3					1.3
Ulva sp.	−13.0 ± 0.40	7.60 ± 0.20	3	3					1.3
T. hemprichii	−14.8 ± 0.60	6.10 ± 0.50	3	3					1.1
Invertebrates									
Polychaetes	−21.8 ± 0.80	9.5 ± 0.80	10	10					1.7
Terebralia sp.	−21.5 ± 0.70	10.8 ± 0.60	12	10					2.0
C. cucullata	−22.3 ± 1.60	10.1 ± 0.50	10	9					2.0
Uca sp.	−21.6 ± 0.80	10.7 ± 0.60	20	20					2.2
P. monodon	−17.3 ± 1.60	13.2 ± 1.00	15	15					2.8
Insects									
Beetles	−23.8 ± 0.60	8.60 ± 2.50	20	20					1.5
Geridae	−25.6 ± 0.50	7.10 ± 1.86	10	10					2.6
Fish									
A. pectoralis	−22.6 ± 1.40	10.7 ± 1.20	6	6					3.2
Hilsa kelee	−27.6 ± 0.60	13.8 ± 0.80	6	6	36.0–108	68.0 ± 9.50	18.8–30.6	22.8 ± 10.2	3.0
Trichiurus lepturus	−15.5 ± 0.70	16.1 ± 1.00	6	6	260–840	460 ± 111	20.6–66.5	48.4 ± 15.4	3.6
Arius thalassinus	−13.9 ± 1.40	16.4 ± 1.20	6	6	115–365	245 ± 98.6	183–295	135 ± 62.8	3.7
Valamugil buchanani	−23.2 ± 0.70	13.0 ± 0.50	6	6	20.0–90.5	45.4 ± 8.60	40.0–87.0	42.6 ± 7.80	2.8

[x] Nd: Number of organisms used to determine trace metal concentrations.
[y] Na: Number of organisms used in stable isotope analysis.
[z] TL: Trophic level.

$$\delta_x = \left(\left(\frac{R_{sample}}{R_{standard}}\right) - 1\right) \times 1000 \quad (1)$$

where x is ^{13}C or ^{15}N and R is the ratio of $^{13}C/^{12}C$ or $^{15}N/^{14}N$. The values in different standards were determined based on the Vienna PeeDee Belemnite (V-PDB) for ^{13}C and atmospheric N_2 for ^{15}N. Replicate measurements of the internal laboratory standard (albumen) yielded measurement errors which were within ± 0.3‰ and ± 0.2‰ for stable C and N isotope measurements, respectively.

Trophic level (TL) for each aquatic organism was calculated as:

$$TL_{consumer} = 2 + (\delta^{15}N_{consumer} - \delta^{15}N_{C.\ cucullata})/3.8 \quad (2)$$

where $TL_{consumer}$ is the trophic level and TL of *C. cucullata* was assumed to be 2.

The food web was then determined according to TL obtained from $\delta^{15}N$. Food sources were determined by $\delta^{13}C$ and the prey-predator relationship described in the literature (Gounter and Furness, 1997; Montesinos et al., 2008).

2.5. Trophic magnification and biomagnification factors

The trophic magnification factor (TMF) was used to measure biomagnification of trace metals in food webs representing the average increase rate per TL rather than specific predator-prey relationship. Trophic level positions for various functional groups in organisms were obtained according to stable N isotope ratios described by Fisk et al. (2001) and Johnson and Schindler (2009). Determination of TMF was based on a linear regression between TL and trace metal concentrations following Hu et al. (2005):

$$Log(HMC) = a + b \times TL \quad (3)$$

where a and b are constants. The slope b was used to calculate food web magnification factor (FWMF) according to (Larissa et al., 2006) and TMF according to Hu et al. (2005) as below:

$$FWMF = b \quad (4)$$

and

$$TMF = 10^b \quad (5)$$

Biomagnification factor (BMF) was calculated according to Larissa et al. (2006) and Hoekstra et al. (2003)

$$BMF = \left(\frac{Metal_{predator}}{Metal_{pray}}\right) / \left(\frac{TL_{predator}}{TL_{pray}}\right) \quad (6)$$

where $Metal_{Predator}$ and $Metal_{Prey}$ are metal concentrations in the predator and prey species, respectively.

2.6. Statistical analysis

Descriptive statistics, linear regression analysis as well as testing the data for homogeneity of variances were performed using the PAST statistical software (version 3.14). The results for this test did not show any homogeneity of variance, and therefore non-parametric tests for multivariate analyses of data were applied using PRIMER v6 multivariate statistics package (Clarke and Gorley, 2006) with the PERMANOVA + add-on module (Anderson et al., 2008). All multivariate tests were performed at a significance level of $p < 0.05$. In order to test the null hypothesis on differences in trace metal concentrations across sampling sites and biota, a permutational multivariate analysis of variance (PERMANOVA) test was used. To perform this test, the Bray-Curtis similarity matrices were constructed by considering two factors (i.e. biota and sites). All data were square root transformed in order to scale down the contributions of quantitatively dominant metal species to similarities between the samples. The same resemblance matrices were used to produce non-metric multi-dimensional scaling (nMDS) ordination plots for visualizing the level and pattern for trace metal distribution between different sites.

3. Results

3.1. Trophic levels in estuarine food webs

The results indicated that $\delta^{13}C$ values in primary producers ranged from −23.2‰ to −13.0‰, whereas $\delta^{15}N$ values varied from 7.1‰ to 16.4‰ (Fig. 2). The permutational multivariate analysis of variance (PERMANOVA) of the data indicated significant differences in $\delta^{13}C$ and $\delta^{15}N$ values among different aquatic species ($P = 0.001$; Table 1). The estimated differences in $\delta^{15}N$ values between the lowest TL primary producers (7.1‰) and the highest TL consumer *A. thalassinus* (16.4‰)

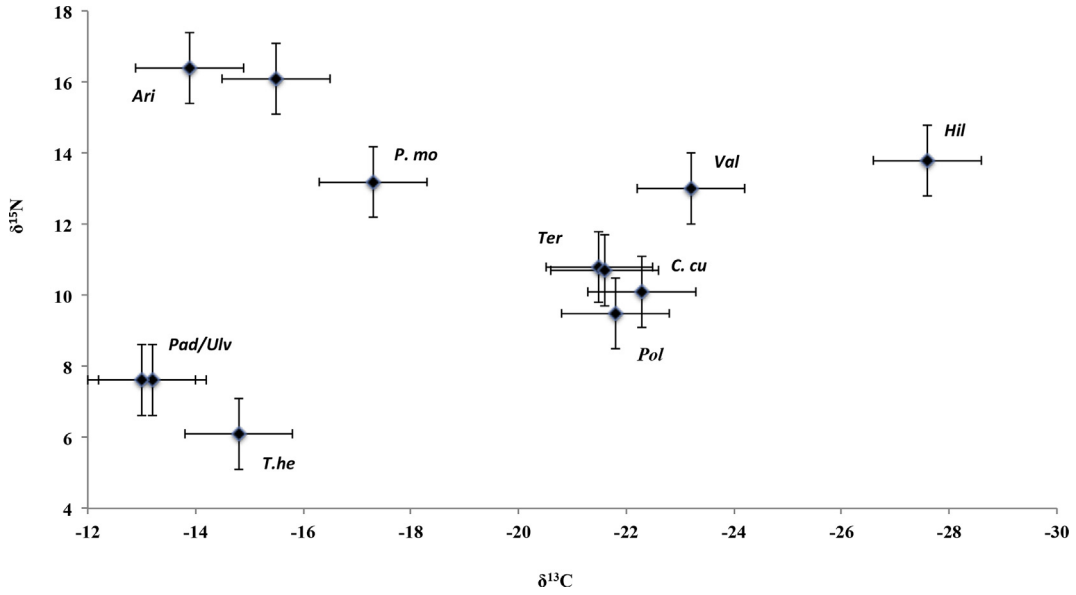

Fig. 2. Stable isotope diagram of members of the estuarine food web in the Rufiji estuary: Cross plot of δ^{15}N ‰ (mean ± SD) versus δ^{13}C ‰ (mean ± SD) in samples.

was ~9.3‰.

The samples were categorized into four groups consisting of primary producers, invertebrates, insects, and fish with trophic levels varying from 1.2 to 3.7 (Table 2).

3.2. Trace metal concentrations

The permutational multivariate analysis of variance (PERMANOVA) of the data revealed no significant differences in trace metals across sampling sites, but significant differences were noted between the species (Table 3). Metal concentrations are the highest in seaweeds and high TL fish species. In general, Cu, Mn, and Zn in biota depicted higher concentrations than other trace metals. The highest concentration of Zn occurred in *T. lepturus* (90.2 mg g^{-1} dry weight), whereas the highest concentration of Mn (95.4 mg g^{-1} dry weight) and Cu (74.1 mg g^{-1} dry weight) occurred in *P. monodon* and Gerridae, respectively (Table 1S; see Supplementary data).

3.3. Trace metal patterns

Ordination of trace metal data shows a clear separation of functional groups between different organisms (Fig. 3). Based on Table 4, the first axis (Axis 1) was associated with higher loads of As, Cr, Cu, Pb, Mn, and Zn towards the right-hand side of the axis, which was associated with *Uca* sp., *C. cucullata,* and polychaetes. Axis 2 was represented by higher loads of Ag towards the top of the axis which was associated with *T. lepturus*, *V. buchanani*, Terebralia sp., *H. kelee,* and *A. thalassinus.* Higher loads of Cd occurred towards the lower side of the axis and was associated with sediments and seaweeds. Axis 3 was characterized by higher Co and Ni loads and was associated with insects *P. monodon*, *A. pectoralis,* and Gerridae. All species were classified into four groups namely primary producers (algae and seaweed *T. hemprichii*), invertebrates, insects, and fish. These samples can also be classified into three functional groups based on their TLs: the first group included algae and seaweed (TL: 1.7–2.2); the second group included *Uca* sp., *C. cucullate,* and polychaetes (TL: 1.7–2.2); and the third group included *T. lepturus*, *V. buchanani*, Terebralia sp, *H. kelee*, *A. thalassinus*, insects, *P. monodon*, *A. pectoralis,* and Gerridae (TL: 1.5–3.7).

Table 3
Two-ways PERMANOVA on trace metal distribution in biota across sampling sites in the Rufiji estuary.

Source of variation	df	SS	MS	Pseudo-F	P(perm)
Species	13	200	15.4	20.8	0.001
Residue	28	20.7	0.74		
Total	41	221			
Sites	2	2.31	1.16	0.21	0.99
Residue	39	219	5.61		
Total	41	221			

3.4. Biomagnification of trace metals

Regression of the TLs plotted vs. trace metal concentrations revealed that As and Zn displayed TL-dependent accumulation processes in the Rufiji estuarine food webs (Fig. 4 and Table 5). Arsenic and Zn increased with the TL, whereas Ag, Cr, Cu, Co, Mn, Ni, and Pb indicated an opposite trend (Fig. 4). Table 5 indicated that FWMF varied from −0.57 for Ni to 0.39 for Zn, whereas TMF varied from 0.27 for Ni to 2.47 for Zn. Ni and Zn bioaccumulated in the Rufiji estuarine food webs and was indicated by a slope > 0. The remaining metals were either eliminated from the food web or their trophic transfer was interrupted.

Low R values of the regression for some metals indicated that this relationship lacked linearity. BMF for trace metals differed between the organisms. The variation ranged from 0.03 for As from *P. monodon* to *A. pectoralis* to 65.0 for Zn from *C. cucullata* to *T. lepturus* (Table 6). Concentrations of Ag, Cd, Co, Cr, Ni, and Pb indicated substantial increase without significant biomagnification effects on species.

4. Discussion

The telescoping food web structure investigated in this study indicate a complex pattern, which is not unexpected in estuarine environments. The δ^{13}C and δ^{15}N values indicate presence of several primary food sources for consumers at higher TLs in the Rufiji estuary. The major primary producers in the estuary (mangroves, microalgae, phytoplankton, and seagrass) are potential food sources for the dominant fish species living at different water depths (Shilla and Routh, 2018). Since terrestrial organic matter is exported further into the open marine environment, it plays an important role as a carbon source in the Rufiji ecosystem. The δ^{15}N values help in distinguishing different TLs in the ecosystem. Considering a trophic fraction of 3.4‰ per TL

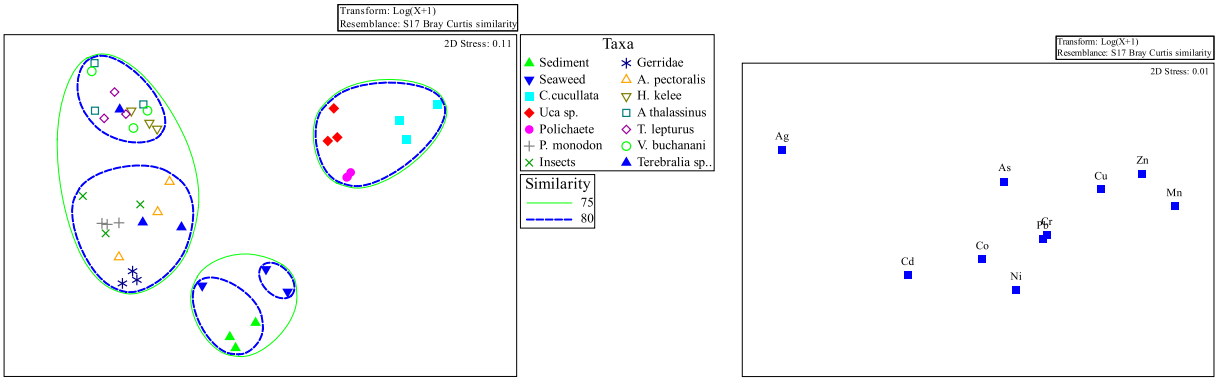

Fig. 3. Ordination (non-metric multi-dimension scaling) of trace metals in the Rufiji estuarine sites. The variables used were Ag, As, Cd, Co, Cr, Cu, Mn, Ni, Pb, and Zn. The graph in the upper right shows the contribution from variables associated with non-metric multi-dimension scaling in a loading plot; stress value is indicated in the plot.

Table 4
Non-metric multi-dimension scaling (nMDS) ordination axes show the correlation statistics for trace metals in the Rufiji estuarine sites.

Metals	Axis 1	Axis 2	Axis 3
Ag	−0.18	−0.82	0.12
As	−0.65	0.27	−0.16
Cd	−0.46	−0.76	0.02
Cr	0.74	−0.14	0.10
Co	0.22	−0.25	−0.82
Cu	0.72	−0.44	−0.25
Mn	−0.76	−0.32	0.20
Ni	0.32	−0.11	−0.74
Pb	−0.79	−0.17	0.28
Zn	−0.79	−0.10	−0.14

based on $\delta^{15}N$ values, the food web structure in the Rufiji estuary spanned over ca. 3 TLs between primary producers and predatory fish species on top of the food chain. The invertebrates were placed between TL 1 and 3.

A comparison of present data with other studies show that metal loads in sediments, primary producers, and consumer organisms in the Rufiji estuary are high (Larissa et al., 2006; Cui et al., 2011, 2012). Although metal concentrations in the Rufiji sediments seem lower compared to levels recorded elsewhere in Tanzania (Shilla, 2016), it far exceeds the average shale values which are commonly used as background values in similar studies focusing on metal contamination (Lopez-Sanchez et al., 1996; Jones and Turki, 1997; Date and Subramanian, 1998; Morillo et al., 2002). Comparison of metal concentrations in sediments with average shale values reveals that sediments from the Rufiji estuary are polluted with high trace metal loads. In a recent study, Minu et al. (2018) indicated enrichment of several trace metals in the Rufiji sediments. The authors indicate that these metals are present in the residual phase (extracted following the BCR scheme; Rauret et al., 1999), and therefore less exposed to chemical weathering. Nevertheless, the Rufiji River is one of the largest rivers in East Africa and drains ~20% of mainland Tanzania (Erftemeijert and Hamerlynck, 2005). Hence, the river has a high potential to transport assorted contaminants from the catchment before draining some of these toxic pollutants into the Indian Ocean.

Trace metal concentrations in biota from the Rufiji estuary are lower compared to levels reported by Shilla (2016) in the Fumba and Malindi coasts of the Tanzanian archipelago except for seaweed, which has higher values. Notably, concentrations of Cr, Cu, Pb, and Zn, in sediments and seaweed show enrichment in the current study. Manganese recorded higher concentrations in all samples from different sites (Table 1S, supplementary data). In general, concentrations of metals are higher in fish than in invertebrates, contradicting the results presented by Marin-Guirao et al. (2008) and Farag et al. (2007). This observation could be due to variation in trace metal concentrations in fish with respect to different species and aquatic environments. Notably, results from this study show differential incorporation of metals in food webs, particularly in *T. lepturus,* which has high ability to accumulate As, Cu, Mn, and Zn from *C. cucullata* and *Uca* sp., whereas polychaetes and *A. pectoralis* accumulate As and Zn from *Uca* sp. Higher concentrations of metals in fish compared to invertebrates in this study can also be explained based on the feeding habits of fish investigated in this study. For example, fish range from pelagic to benthopelagic feeders and they consume phytoplankton, crustaceans, squids, crabs, prawns, and shrimps, but also small fish and mollusks in the Rufiji estuary. These organisms that are consumed are known to live in benthic environments ingesting mud and very fine sediments, which increases the potential for ingesting trace metals (Villares et al., 2005). Hence, differences in trace metal concentrations in different fish are not only related to their TL position, but it also reflects differences in their feeding habits (Lavoie et al., 2010). The process of metal bioaccumulation in biota is however very complex, and it is depends on exposure routes, age, and geochemical effects on the availability of trace metals (Luoma and Rainbow, 2005). Affinity for metal absorption from contaminated water and food may also differ in relation to ecological needs, metabolism, and contamination gradients in water, food, and sediments, as well as other factors such as seasonality, salinity, and temperature (Roméo et al., 1999). Therefore, in order to establish the impact of these factors, it is recommended to pursue long-term studies of these biophysical parameters and monitoring trace metal levels in the food chain.

Biomagnification of selected trace metals in the Rufiji estuarine food webs is not conspicuous and the underlying relationships between TLs and logarithmic concentration of trace metals show non-linearity (Table 5). Researchers have noted that despite metal uptake and absorption through gills and body surfaces are considered important exposure pathways for invertebrates and fish (Costa et al., 2009), tissue obtained from preys exposed to contaminants is assumed to be a good proxy for the total body burden (Jaeger et al., 2009). Accumulation of trace metals is therefore affected by many physical and biological factors due to their dynamic nature (McGeer et al., 2003; Ilbäck et al., 2004; Laura, 2009).

Determination of BMF is based on the comparison of trace metals in predators and preys with the assumption that both predators and preys involved in the comparison have a simple predator-prey relationship, and predators completely consume their preys. However, these assumptions may not be true in complex food webs like that of the Rufiji estuary because consumers, particularly fish have diverse diet options, and carnivorous fish are known to practice opportunism in their feeding habits. Moreover, shifting their diet with age and season and selecting different preys based on the availability of food and sex also influence the BMF trends (Syvaranta et al., 2009). BMF results show that all

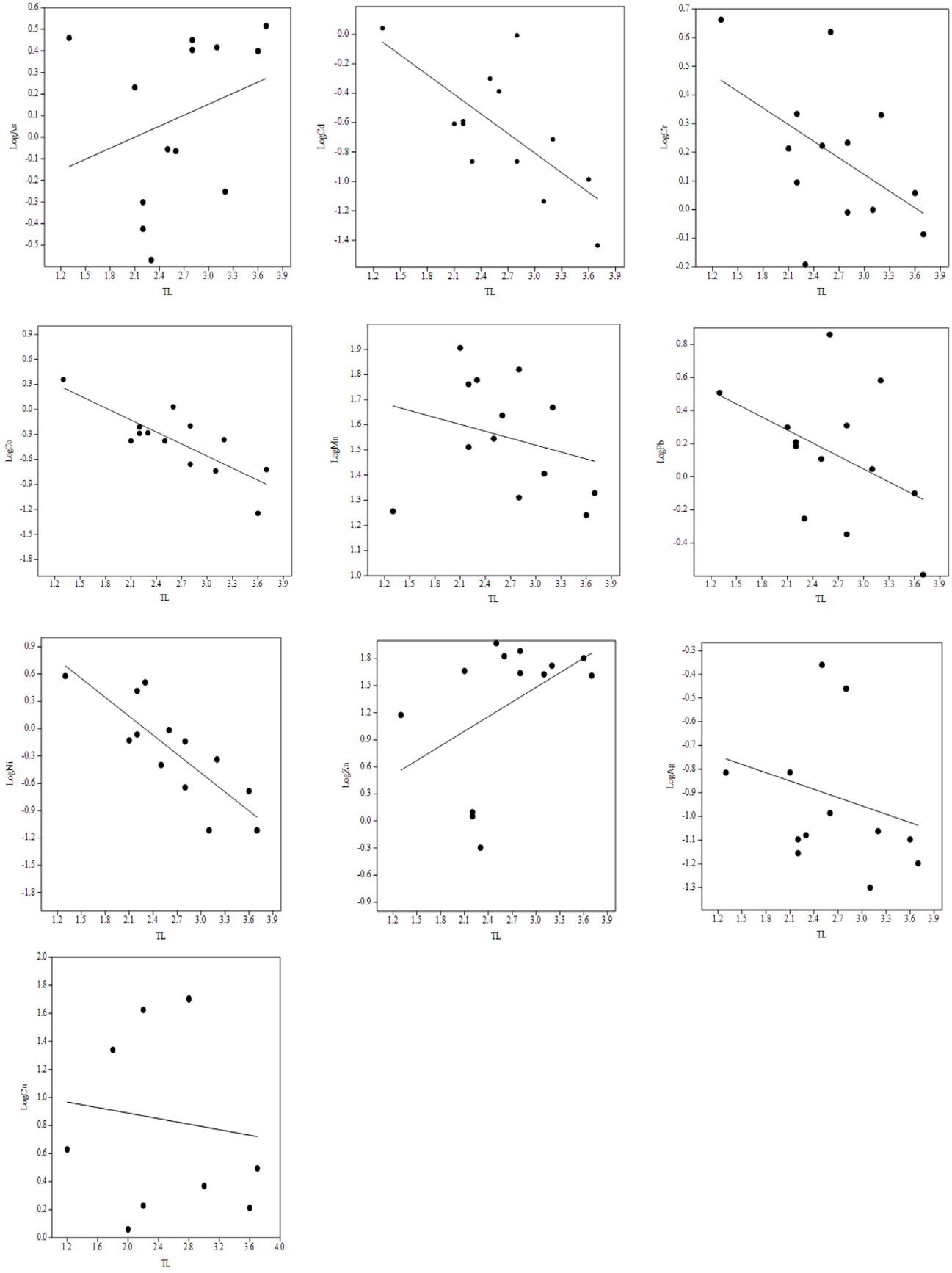

Fig. 4. The relationship between trophic levels and trace metal concentrations in biota in the Rufiji estuarine sites.

Table 5
Regression analysis between logarithm concentrations and trophic levels (slope, p-value of slope) and trophic magnification factors for trace metals in the Rufiji estuarine sites.

	As	*Cd*	*Cr*	*Co*	*Cu*	*Pb*	*Mn*	*Ni*	*Zn*	*Ag*
Slope (FWMF)	0.23	−0.38	−0.16	−0.43	−0.10	−0.27	−0.31	−0.57	0.39	−0.30
Intercept	−0.49	0.32	0.61	0.71	1.09	0.84	2.54	1.17	0.35	−1.92
R	0.87	−0.62	−0.46	−0.77	−0.12	−0.50	−0.52	−0.78	0.89	0.57
TMF	1.70	0.42	0.69	0.37	0.80	0.54	0.49	0.27	2.47	2.00
p-value	0.17	0.05	0.18	0.01	0.75	0.14	0.12	0.01	0.26	0.08

Table 6
Predator-prey biomagnification factors (BMF) in the Rufiji estuary.

Predator	Prey	As	Cd	Cr	Co	Cu	Mn	Ni	Pb	Zn	Ag
Hilsa kelee	*P. monodon*	0.93	0.07	0.53	0.26	0.04	0.49	0.35	0.10	0.50	2.05
	Geridae	2.53	0.15	0.20	0.14	0.04	0.13	0.49	0.07	0.53	2.10
	Beetles	2.39	0.12	0.48	0.35	0.04	0.70	0.59	0.15	0.36	3.36
Trichiurus lepturus	*V. buchanani*	0.69	0.59	0.91	0.20	0.57	1.38	0.66	0.71	1.14	0.50
	Beetles	1.98	0.14	0.48	0.09	0.05	0.43	0.35	0.36	0.47	9.49
	Terebralia sp.	1.09	0.08	0.28	0.26	0.11	0.07	0.15	0.06	0.50	1.27
	P. monodon	15.6	0.06	0.36	0.23	0.05	0.10	0.24	0.08	0.40	7.09
	Uca sp.	3.89	0.09	0.39	0.18	1.09	0.10	0.04	0.05	19.5	2.66
	Polychaetes	5.16	0.08	0.23	0.22	0.93	0.09	0.39	0.02	21.6	3.06
A. pectoralis	*C. cucullata*	1.30	0.88	2.08	0.52	11.9	4.27	0.06	0.09	65.0	0.15
	P. monodon	0.03	0.17	1.10	0.60	0.38	1.64	0.62	0.55	0.60	0.56
	Uca sp.	0.77	0.54	1.18	0.48	8.83	1.72	0.10	0.37	29.0	0.21

metals except As and Zn are generally not biomagnified (biodiluted) in food webs of the Rufiji estuary. Arsenic and Zn concentrations in the biota depend on TL as indicated by the positive linear relationship between TL and these metals (Dietz et al., 2000; Campbell et al., 2005, Fig. 4). Other studies have reported biomagnification of metals in food webs based on the significant positive relationships between log metal concentrations and $\delta^{15}N$ in muscle and liver tissue belonging to the organisms.

We infer from this study that trophic transfer is a prime way for As and Zn accumulation at higher TLs. The life history may affect biomagnification of trace metals even for organisms that are at the same TL. For example, a longer life history of *T. lepturus* reveals higher BMFs of As and Zn than those in *A. pectoralis* even though they share similar preys (Table 6). Moreover, different metal species in food webs can have a different degree of assimilation in stomachs of various organisms at each TL. For example, Wang and Wong (2003) reported variation in the assimilation efficiency of elemental and organic mercury in copepods, silversides, and brine shrimps.

5. Conclusions

Trace metal data in sediments and aquatic organisms collected from the Rufiji estuary were assessed in this study. Stable nitrogen and carbon isotope values revealed a food web length of 3.7 with *A. thalassinus* at the highest TL. Trace metal levels in the Rufiji estuary are lower than levels reported at other Tanzanian sites, but higher than levels reported in similar regions outside Tanzania. Arsenic and Zn in the Rufiji estuary are the only metals found to have biomagnification effects based on FWMF. Based on BMF values, there is significant biomagnification effect from *C. cucullata*, *P. monodon*, *Uca* sp., and polychaetes to *T. lepturus* and *A. pectoralis*. There is no conspicuous difference in trace metal concentrations between the high TL fish species. This is probably due to the opportunistic behavior portrayed by these species in their feeding behavior. We recommend that further investigations be done in order to elucidate a sound basis for protecting the biota living in the Rufiji River estuary.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.apgeochem.2018.11.016.

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