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Bioaccumulation and public health implications of trace metals in edible tissues of the crustaceans *Scylla serrata* and *Penaeus monodon* from the Tanzanian coast

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Abstract The coastal population in East Africa is growing rapidly but sewage treatment and recycling facilities in major cities and towns are poorly developed. Since estuarine mangroves are the main hotspots for pollutants, there is a potential for contaminants to accumulate in edible fauna and threaten public health. This study analysed trace metals in muscle tissues of the giant mud crabs (*Scylla serrata*) and the giant tiger prawns (*Penaeus monodon*) from the Tanzanian coast, in order to determine the extent of bioaccumulation and public health risks. A total of 180 samples of muscle tissues of *S. serrata* and 80 of *P. monodon* were collected from

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Cd, Cr and Pb did not exceed maximum limits for human consumption. Based on the current trend of fish consumption in Tanzania (7.7 kg/person/year), the measured elements (As, Cd, Co, Cu, Mn, Pb and Zn) are not likely to present health risks to shellfish consumers. Nevertheless, potential risks of As and Cu cannot be ruled out if the average per capita consumption is exceeded. This calls for strengthened waste management systems and pollution control measures.

nine sites along the coast. Both species showed high

levels of trace metals in the wet season and significant bioaccumulation of As, Cu and Zn. Due to their

burrowing and feeding habits, mud crabs were more

contaminated compared to tiger prawns sampled from

the same sites. Apart from that, the measured levels of

Keywords Heavy metal pollution · Shellfish · Shrimps · Western Indian Ocean · Target hazard quotient

Introduction

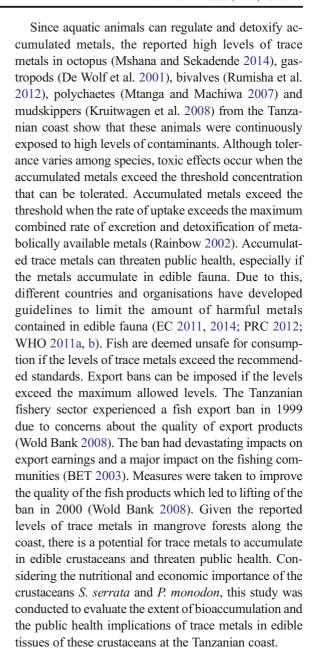
The crustaceans *Scylla serrata* (giant mud crab) and *Penaeus monodon* (giant tiger prawn) provide an important source of income, food and essential nutrients to coastal communities in the Indo-Pacific. These crustaceans spawn offshore producing planktonic larvae that moult into benthic juveniles, which inhabit muddy and sheltered areas among seagrass beds and mangrove ecosystems (Alberts-Hubatsch et al. 2016; Garcia 1988). These crustaceans are usually fished together; however, when the target group is only crabs for special



purchase orders, hook sticks, baited traps and other traditional methods are employed to maximise collection (Kyomo 1999; Mirera 2009). They are typically harvested in estuarine mangroves and sold either to domestic markets for local consumption or to export companies. A total of 41,160 kg of *P. monodon* was exported from Tanzania between January and December 2010 (Mosha and Gallardo 2013). Often, the crustaceans are exported live or frozen to Asia, Europe and other African countries. The main export destination countries include countries in the European Union, Japan, China and the Middle East (URT 2016).

Even though the crustacean fishery generates significant foreign exchange and income, the rapidly growing coastal population in East Africa is threatening its sustainability. Because sewage treatment and recycling facilities in major cities and towns are poorly developed, industrial, institutional and domestic wastes are released to streams or directly to coastal areas with minimum treatment (Kihampa 2013; Mmochi and Francis 2003). In addition, since agricultural activities in the region are normally done along the river basins, residues of fertilisers and pesticides also enter coastal areas (Taylor et al. 2003). As a result, significant enrichment of trace metals is reported in many areas along the Tanzanian coast (Hellar-Kihampa et al. 2012; Kamau et al. 2015; Mtanga and Machiwa 2007; Rumisha et al. 2012, 2016).

Although significant enrichment of trace metals is reported, the bioavailability and toxicity of these elements is affected by pH, temperature, alkalinity and hardness, inorganic ligands, sediments particle size and organic substances. In addition, metal uptake and toxicity is also influenced by size, life stages, species and feeding habits (Wang 1987). Generally, uptake of the trace metals across the plasma membrane is often either via carrier-mediated transport or through protein channels (Rainbow 1997). Apart from that, the crustaceans have the ability to detoxify and prevent build-up of trace metals in their bodies through (i) physiological regulatory mechanisms that balance metal excretion rates with uptake, (ii) intracellular sequestration mechanisms that involve binding to metallothioneins and subsequent elimination through the lysosomal endomembrane system and (iii) intracellular sequestration processes involving specific vacuoles producing solid metallic phosphorous or sulphur granules which subsequently undergo exocytosis for elimination (Ahearn et al. 2004; Deb and Fukushima 1999).



Materials and methods

Study area

The study was conducted along the coastline of the Western Indian Ocean, Tanzania, which extends to over 800 km. Mangrove forests occur along the entire coastline and around the islands (Oliveira et al. 2005). The mangroves are endowed with a diverse range of fauna



including crabs, molluscs, echinoderms, prawns, shrimps and a variety of fish species (Rumisha et al. 2015; Taylor et al. 2003). The giant mud crab S. serrata and Penaeid prawns are among the commercially important crustacean species harvested from the mangroves. Although mud crab and prawn availability is almost all year round, in some areas, prawns are more abundant during the rainy season (Kyomo 1999). Nine sites were selected along the coast based on the availability of giant mud crabs S. serrata and giant tiger prawns P. monodon (Fig. 1). Selection of sites was also based on the reported levels of trace metal accumulation in mangrove sediments and fauna (De Wolf et al. 2001, Mtanga and Machiwa 2007, Kruitwagen et al. 2008, Hellar-Kihampa et al. 2012). Sites 1, 4 and 9 are densely populated (URT 2013) and most industrialised compared to other sample sites. As a result, significant enrichment of trace metals is reported in mangrove ecosystems in these areas (Kruitwagen et al. 2008; Mtanga and Machiwa 2007; Rumisha et al. 2016). The mangrove forests at sites 2, 3 and 5 are located in estuaries of big rivers (river Pangani, Wami, and Rufiji). The rivers contain the most extensive mangrove forests and they are the main prawn fishing grounds. Although the level of industrialisation and population density at sites 7 and 8 is low, a recent study reported high levels of trace metals in mangrove forests in the area (Rumisha et al. 2016). The metals were attributed to agricultural effluents and domestic wastes. The climate of the study area is tropical and it is influenced by seasonal monsoons.

Sampling

Sampling of giant mud crabs *S. serrata* and giant tiger prawns *P. monodon* was conducted between 2014 and 2015. Samples were collected during the wet (March–May) and dry (August–October) season. At each site, 20 individual giant mud crabs were collected during the wet and dry season. In addition, 80 tiger prawns were collected from sites 1, 2, 3 and 4 during the dry season (20 individuals from each site). A section of the muscle tissue was collected from each animal and stored frozen in plastic bags for further analysis. The crabs were collected from their burrows with the help of local fishermen. A thin wire attached to a wooden rod was lowered into the burrow to dislodge crabs. The crabs were caught as they emerged from the burrows. Giant

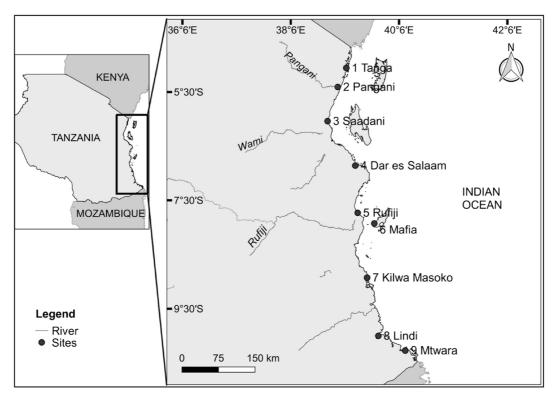


Fig. 1 Map of the Tanzanian coast showing the sample sites



tiger prawns were obtained from local fishermen at landing sites. The geographical coordinates of each site was recorded with a GPS receiver, and it is reported in Table S1.

Laboratory analyses

Tissues samples of each animal from the different sites were pooled, weighed and frozen overnight in a deep freezer. The frozen tissues were freeze-dried in a lyophiliser for 48 h, weighed and homogenised using a mortar and pestle. The moisture content of the samples was estimated as the percent change in mass before and after freeze-drying. Approximately 0.2 g of the homogenised sample was weighed and digested in a CEM Mars 5 Microwave with 5 mL of doubledistilled HNO₃ (65%) and 1 mL of H₂O₂ (30%). Each digestion set up contained 10 digestion vessels with samples, two vessels with reagents only (blanks) and one vessel with a certified reference material (CRM). Dogfish (Squalus acanthias) liver DOLT-3 (National Research Council, Canada) was used as CRM. Digestion was conducted at 180 °C for 15 min followed by a holding time of 15 min. After being allowed to cool for 10 min, the samples were removed from the microwave and 40 mL of deionised water was added to each vessel before the mixture was transferred to polyethylene bottles. The samples were then diluted ten times and analysed for trace metals (As, Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, V and Zn) with a Sector Field Inductively Coupled Plasma Mass Spectrometer (ICP-MS) Element II (Thermo Fisher Scientific, Bremen, Germany). To determine the repeatability of the analytical methods used, three replicate samples of the CRM were analysed using the same equipment. The obtained standard error for each measurement is shown in Table 1. The accuracy of the analytical methods used was determined by comparing the mean value of three replicate samples of the CRM with certified values, and it was expressed as percentage recovery. The obtained recoveries ranged between 73 and 96% (Table 1). Precision was determined as the relative standard deviation (RSD) of the replicated CRM, and it ranged between 2.5 and 5.7% (Table 1).

Data analyses

Bioaccumulation of trace metals in tissues

The Student's *t* test was used to determine whether there are significant differences between the levels of trace metals measured in muscle tissues of giant mud crabs and giant tiger prawns. The same test was used to determine whether there are significant differences in trace metal concentration between samples collected during the wet and dry season. The biota-sediment accumulation factor (BSAF) was used to describe the extent by which trace metals occur in tissues and the associated sediments. The index was estimated using the following equation

$$BSAF = \frac{C_x}{C_s} \tag{1}$$

where C_x and C_s are the concentrations in biota and sediments, respectively. Sediment concentrations (C_s) were obtained from Rumisha et al. (2016).

The metal pollution index (MPI) was used to describe the extent of trace metal contamination at each site. The index was calculated using the following equation:

$$MPI = \sqrt[n]{Cf_1 \times Cf_2 \times ...Cf_n}$$
 (2)

(Usero et al. 2005).where Cf_n = concentration of metal n in the sample.

Table 1 Repeatability, precision and accuracy of the analytical methods performed with the certified reference material (CRM)

	Certified value (µg/g)	Analytical value (µg/g, \pm SE)	Precision (% RSD)	Accuracy (%)
As	10.2	9.1 ± 0.1	3.6	89
Cd	19.4	16.8 ± 0.4	5.5	86
Cu	31.2	29.3 ± 0.6	4.4	94
Fe	1484	1307.3 ± 33.2	5.7	88
Pb	0.32	0.23 ± 0.01	5.7	73
Zn	86.6	83.3 ± 0.9	2.5	96

SE standard error of the mean of three repeated analysis, RSD relative standard deviation



Public health risks

The measured levels of Cd, Cr and Pb were compared with maximum allowed concentrations for human consumption in order to evaluate if they present potential risks to consumers (EC 2011, 2014; PRC 2012). Due to the long half-life of cadmium, daily ingestion in food has a small or even a negligible effect on overall exposure; therefore, to assess long- or short-term health risks due to cadmium exposure, dietary intake should be assessed over months and tolerable intake should be assessed over a period of at least 1 month (WHO 2011b). In view of this, monthly ingestion of Cd through the consumption of mud crabs and tiger prawns was estimated and compared with the provisional tolerable monthly intake established as safe by the Joint FAO/ WHO Expert Committee on Food Additives (WHO 2011a, b). Monthly intake of Cd (MMI) was estimated using the equation:

$$MMI = AfP \times MC \tag{3}$$

(adapted from Ordiano-Flores et al. (2011)).where MC = metal concentration in fish expressed as wet weight, and AfP = average fish portion consumed per month, which was estimated according to Rumisha et al. (2016).

Since the provisional tolerable weekly intake (PTWI) for As and Pb were withdrawn by the joint FAO/WHO expert committee on food additives (WHO 2011a, b), the target hazard quotient (THQ) was used to determine if the measured elements present adverse health risks to consumers. The index compares measured concentration with the oral reference dose. An element is considered to present potential adverse health effects if it has a THQ larger than one (Antoine et al. 2017). The index was estimated based on the US Environmental Protection Agency (US EPA) regional screening level generic tables (US EPA 2016) using the following equation:

$$THQ = \frac{E_{FR} \times Ed \times F_{IR} \times C}{RfD \times BWa \times ATn} \times 10^{-3}$$
 (4)

(Antoine et al. 2017).where E_{FR} = exposure frequency to the trace metal, Ed = exposure duration (equivalent to an average life expectancy, which is 61.8 years for Tanzania), F_{IR} = fraction of shell fish consumed in grams per day, C = concentration of trace metals in shellfish (in wet weight), RfD = oral reference dose of the trace metal in mg/kg/day (Table 2), BWa = average body weight of

an adult (70 kg), ATn = average exposure time (365 days \times 61.8 years), and 10^{-3} is the unit conversion factor.

Studies show that inorganic As, which is toxic, accounts for less than 4% of the levels of total As in tissues of prawns and crabs (Muñoz et al. 2000; Peshut et al. 2008). Since this study measured total As in muscle tissues of *S. serrata* and *P. monodon*, a conversion factor of 4% was used to estimate the THQ for As. To determine risks resulting from cumulative effects of consumption, the hazard index (HI) was estimated using the equation;

$$HI = \sum_{i=1}^{n} THQ_i \tag{5}$$

where $THQ_i = THQ$ of an individual element, and n = number of trace metals used in the analysis (eight in this case).

Results

Concentration of trace metals in muscle tissues

The distribution of trace metals in muscle tissues of giant mud crabs and giant tiger prawns at the Tanzanian coast is shown in Tables 2 and 3, respectively. The Student's t test showed that the measured levels of As, Cd, V and Zn were significantly higher in muscle tissues of mud crabs compared to tiger prawns (p < 0.05). The concentration of Zn was at least three times higher in tissues of giant mud crabs compared to giant tiger prawns from the same site. In addition, the mud crabs showed high levels of As at sites 5, 7 and 9 and high levels of Cd at sites 6, 7 and 9. Apart from that, the measured levels of Co, Cr, Cu, Fe, Mn, Pb and Ni in tissues of mud crabs were not significantly different from the levels measured in tiger prawns (p > 0.05).

The biota-sediment bioaccumulation factor showed significant bioaccumulation of As, Cu and Zn by giant mud crabs and giant tiger prawns in the study area (Fig. 2). Both species showed significant bioaccumulation of Cu and Zn at all sites. Although the concentration of As in tissues of giant mud crabs exceeded environmental concentrations at all sites, tissues of giant tiger prawns showed significant bioaccumulation of As at sites 3 and 7.



Table 2 Average concentration of trace metals ($\mu g/g$ wet weight) in muscle tissues of the giant mud crab ($Scylla\ serrata$) from mangrove forests at the Tanzanian coast

Site	As	Cd	Co	Cr	Cu	Fe	Mn	Ni	Pb	V	Zn	H_2O
1	3.0	0.004	0.01	0.03	12.5	14.6	6.9	0.05	0.02	0.03	68.6	71.9
2	1.0	0.004	0.02	0.04	11.6	27.2	15.9	0.12	0.02	0.06	79.3	72.1
3	3.1	0.003	0.03	0.10	17.4	43.2	7.4	0.06	0.03	0.09	67.4	74.5
4	1.6	0.003	0.03	0.06	11.3	26.3	7.5	0.07	0.07	0.07	58.5	71.0
5	6.0	0.002	0.01	0.02	17.2	15.6	15.3	0.02	0.01	0.04	77.5	72.4
6	2.3	0.010	0.02	0.06	13.6	29.5	7.5	0.14	0.03	0.07	66.5	72.8
7	5.9	0.010	0.01	0.02	14.7	12.0	1.0	0.01	0.01	0.04	69.4	70.6
8	3.1	0.002	0.02	0.04	8.6	21.2	15.6	0.06	0.01	0.05	61.7	73.8
9	4.8	0.013	0.02	0.03	19.5	13.5	5.9	0.12	0.04	0.04	88.4	68.7
RfD		0.001	0.0003		0.04		0.1	0.02	0.02		0.3	
MAL		0.5 ^{ab}		2 ^b					0.5 ^{ab}			

 H_2O percentage of water content in the muscle tissue, RfD oral reference dose of the trace metal in mg/kg/day, MAL maximum allowed levels for human consumption

The concentration of As and Zn measured in muscle tissues of mud crabs showed significant variations between the wet and dry season (p < 0.05). Generally, muscle tissues of mud crabs collected in the wet season contained high levels of Zn compared to samples collected in the dry season (Tables S2 and S3). The concentration of Zn measured in muscle tissues of tiger prawns did not show significant variations between the

wet and dry season (p > 0.05). Apart from that, the measured levels of Co, Cu and Mn in muscle tissues of giant tiger prawns were significantly higher in samples collected during the wet season compared to samples collected during the dry season (p < 0.05). This was also revealed by the metal pollution index, which showed that samples collected during the wet season were more contaminated compared to samples collected

Table 3 Average concentration of trace metals (μ g/g wet weight) in muscle tissues of the giant tiger prawns (*Penaeus monodon*) from mangrove forests at the Tanzanian coast

Site	As	Cd	Co	Cr	Cu	Fe	Mn	Ni	Pb	V	Zn	H_2O	Source
1	0.5	0.012	0.02	0.02	11.5	16.0	0.4	0.03	0.00	0.03	22.5	66.6	c*
2	0.5	0.005	0.01	0.03	9.9	12.2	0.6	0.02	0.09	0.03	21.5	66.0	c*
3	3.6	0.018	0.01	0.05	9.4	15.2	1.2	0.03	0.19	0.04	17.6	59.2	c*
4	1.1	0.016	0.01	0.07	12.4	15.8	1.8	0.08	0.04	0.03	22.8	63.2	c*
5	0.8	0.005	0.01	0.08	10.9	13.3	2.2	0.03	0.01	0.03	19.5	64.6	d^*
7	2.0	0.006	0.01	0.01	9.5	7.8	0.8	0.04	0.01	0.02	22.9	64.4	d^*
8	0.6	0.015	0.03	0.04	14.5	19.0	2.8	0.04	0.02	0.06	22.9	63.7	d^*
9	0.3	0.004	0.03	0.05	11.8	22.5	2.7	0.03	0.01	0.05	22.6	62.5	d^*
RfD		0.001	0.0003		0.04		0.1	0.02	0.02		0.3		
MAL		0.5 ^{ab}		2 ^b					0.5 ^{ab}				

 H_2O percentage of water content in the muscle tissue, RfD oral reference dose of the trace metal in mg/kg/day, MAL maximum allowed levels for human consumption, c^* this study, d^* Rumisha et al. (2016). For sample sites, see Fig. 1

^b PRC (2012)

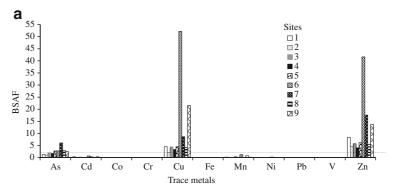


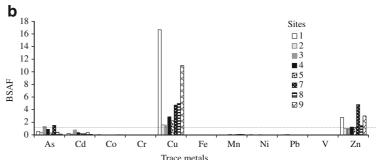
^a EC (2011)

^b PRC (2012). For sample sites, see Fig. 1

^aEC (2011)

Fig. 2 Average biota-sediment bioaccumulation factor showing the relationship between the concentration of trace metals in sediments and muscle tissues of giant mud crabs (*Scylla serrata*) (a) and giant tiger prawns (*Penaeus monodon*) (b) at the Tanzanian coast. The BSAF values above the dotted line represent elements showing significant bioaccumulation





during the dry season (Fig. 3). In addition, the index showed that giant mud crabs from sites 2, 3, 4, 6 and 9 were more contaminated. It also showed that prawns collected from sites 3, 4 and 8 were more contaminated.

(Table S4). The index also exceeded one in giant tiger prawns from all sites except sites 2 and 5.

Public health risks

The measured concentration of trace metals was compared with the maximum allowed levels for human consumption. It was revealed that the measured concentration of Cd, Cr and Pb do not exceed the maximum allowed levels for human consumption (Tables 2 and 3). The measured elements were also compared with the provisional tolerable monthly intake (PTMI) considered as safe by the Joint FAO/WHO Expert Committee on Food Additives (WHO 2011a, b). Based on the current trend of fish consumption in the country, it was revealed that the estimated monthly intake of Cd through consumption of giant mud crabs and giant tiger prawns do not exceed the PTMI (Fig. 4). In addition, the THQ showed that the measured concentration of As, Cd, Co, Cu, Mn, Ni, Pb and Zn do not present potential adverse health risks to giant mud crab and giant tiger prawn consumers (Fig. 5). Apart from that, the hazard index exceeded one in giant mud crabs at all sites

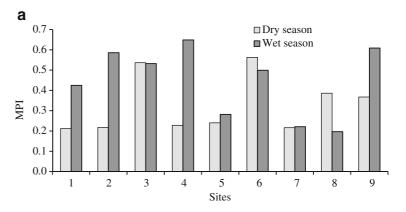
Discussion

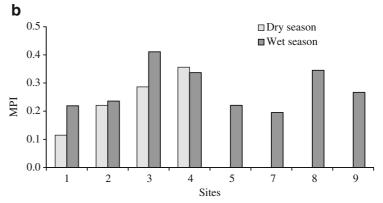
Bioaccumulation of trace metals in muscle tissues

The measured levels of trace metals in muscle tissues of giant mud crabs are low compared to the levels reported in giant mud crabs from Australia (Mortimer 2000), Malaysia (Kamaruzzaman et al. 2012) and India (Krishnamurti and Nair 1999; Mohapatra et al. 2009) (Table 4). The measured trace metals in muscle tissues of giant tiger prawns are also lower than the levels previously reported in giant tiger prawns from Bangladesh (Sarkar et al. 2016), India (Meshram et al. 2014; Mitra et al. 2010) and the Bay of Bengal (Hossain and Khan 2001) (Table 4). Generally, the levels of trace metals in mangroves at the Tanzanian coast are low compared to the levels reported in mangroves in industrialised countries (Rumisha et al. 2016). Although bioavailability might be affected by other biotic and abiotic factors, the fact that Tanzanian mangroves are less contaminated compared to mangroves in the



Fig. 3 Metal pollution index (MPI) showing the total metal content in muscle tissues of a giant mud crabs (*Scylla serrata*) and b tiger prawns (*Penaeus monodon*) at the Tanzanian coast





industrialised world can account for the observed low concentrations of trace metals in Tanzanian crustaceans compared to crustaceans from Australia, Malaysia and India. The findings of this study also show high levels of Cu, Fe, Mn and Zn in muscle tissues of giant mud crabs (*S. serrata*) and the giant tiger prawns (*P. monodon*) (Tables 2 and 3). This observation is in line with the findings of previous studies (Kamaruzzaman et al. 2012; Krishnamurti and Nair 1999; Tu et al. 2008). These elements are essential for many biological processes

including oxidative phosphorylation, gene regulation and free-radical homeostasis (Deb and Fukushima 1999). As a result, all body tissues have significantly higher tendency to accumulate them (Kamaruzzaman et al. 2012), since a certain quantity is required to maintain metabolic activities. Due to this, these elements will still be extracted from the environment, even when environmental concentrations are low (Ahearn et al. 2004). This is supported by the results of the BSAF, which showed that the measured levels of Cu

Fig. 4 Comparison of the cadmium monthly intake (MMI) due to consumption of giant mud crabs (*Scylla serrata*) and giant tiger prawns (*Penaeus monodon*) in Tanzania with the provisional tolerable monthly intake (PTMI) considered as safe by the Joint FAO/WHO Expert Committee on Food Additives (WHO 2011a, b)

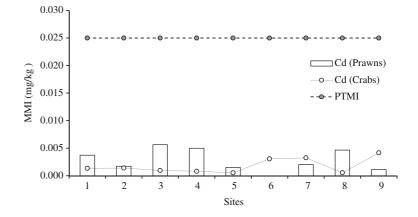
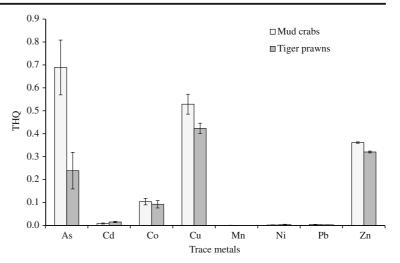




Fig. 5 Average target hazard quotient (THQ) showing risks associated with the measured concentration of trace metals in muscle tissues of giant mud crabs (*Scylla serrata*) and giant tiger prawns (*Penaeus monodon*) from the Tanzanian coast



and Zn in tissues of giant mud crabs and giant tiger prawns exceed environmental concentrations (Fig. 2). The observed significant bioaccumulation also suggests that probably the elements were also accumulated through feeding. Because non-essential metals do not play important biological functions, there are no required minimum tissue concentrations and therefore they need to be detoxified or excreted (Rainbow 2002). Therefore the observed significant bioaccumulation of As suggests that the sampled crustaceans were continuously exposed to this element through either the environment or food. The patterns of accumulation of As, Cd and Pb in muscle tissues of giant mud crabs correspond with the reported patterns of trace metal enrichment in the study area (Rumisha et al. 2016). The pattern of accumulation of As, Cd and Pb in muscle tissues of giant tiger prawns is not consistent with the reported levels of trace metal enrichment (Rumisha et al. 2016). This shows that the giant mud crab S. serrata is a good indicator species, and it is suitable for biomonitoring programs as previously suggested by other researchers (van Oosterom et al. 2010).

Generally, giant mud crabs showed high levels of As and Zn compared to the levels measured in giant tiger prawns and the levels previously reported in Indian white shrimp (Saria 2017) from the Tanzanian coast (Tables 2, 3 and 4). The measured concentrations of Cd and V were also significantly higher in muscle tissues of giant mud crabs compared to giant tiger prawns (p < 0.05). The differences can be attributed to the interspecific differences in uptake and regulation of trace metals since different species have different physiological requirements for essential metals. The

observed differences among species could also be attributed to habitat use and feeding habits. Although both adult giant mud crabs and giant tiger prawns feed on molluscs and crustaceans, the crabs are generally top benthic predators and sometimes they are opportunistic scavengers (Alberts-Hubatsch et al. 2016). As the result, they are likely to pick up particulate matter and sediments during feeding (Kamaruzzaman et al. 2012). Since significant enrichment of trace metals is reported in the study area, this can also account for the observed high levels of trace metals in giant mud crabs. The observed patterns are in line with the findings of a previous study (Krishnamurti and Nair 1999).

The concentrations of As and Zn measured in muscle tissues of mud crabs showed significant variations between samples collected during the wet and dry season (p < 0.05). In addition, the levels of Co, Cu and Mn measured in muscles tissues of giant tiger prawns were significantly higher in samples collected during the wet season compared to samples collected in the dry season (p < 0.05). Theoretically, factors such as salinity, dissolved organics, pH, hardness and sedimentary load affect metal bioavailability and toxicity by influencing the prevailing chemical forms of metals in the aquatic systems (Deb and Fukushima 1999). Generally, salinity drops in estuarine mangroves during the wet season due to dilution resulting from increased river flow. Decrease in salinity during the wet season results to increased abundance of free metal ions due to reduced formation of chloro-complexes (Verslycke et al. 2003). This result in increased bioavailability of trace metals, and it can explain why tissue samples collected during the wet season were more contaminated. In addition, decreased



Table 4 Comparison of the measured levels of trace metals in muscle tissues of giant mud crabs and tiger prawns ($\mu g/g$ wet weight) with the levels previously reported in muscle tissues of crustaceans from the Tanzanian coast and other areas in the tropics and subtropics

Spp.		As	Cd	Cr	Cu	Pb	Zn	Sources
A	Tanzania	13 ± 6.2	0.02 ± 0.01	0.16 ± 0.1	50 ± 11.5	0.09 ± 0.06	252 ± 27	This study
A	Australia	20 ± 7.5	0.08 ± 0.05	0.44 ± 0.1	194 ± 383	0.09 ± 0.05	170 ± 39	Mortimer (2000)
A	Malaysia		0.35 ± 0.04		28.2 ± 12.8	6.52 ± 0.37	362 ± 27	Kamaruzzaman et al. (2012)
A	Thane, India		0.7		73	0.01	376	Krishnamurti and Nair (1999)
A	Mahanadi, India				114 ± 10.4	0.2 ± 0.006	279 ± 20	Mohapatra et al. (2009)
A	Maluan, China		0.22 ± 0.03		3.98 ± 1.2	0.05 ± 0.02	18 ± 3.4	Wang et al. (2011)
В	Tanzania	3.1 ± 2.6	0.03 ± 0.02	0.12 ± 0.05	31.2 ± 4.9	0.13 ± 0.18	60 ± 6.9	This study
В	Vietnam	5.5 ± 1.9	0.01 ± 0.005	0.16 ± 0.05	16.2 ± 2.2	0.01 ± 0.002	51 ± 0.4	Tu et al. (2008)
В	Bay of Bengal		0.3	2.9	21.3	1.3	35.7	Hossain and Khan (2001)
В	Bangladesh		0.05 ± 0.01	0.15 ± 0.02		0.46 ± 0.18		Sarkar et al. (2016)
В	Thailand				6.3 ± 0.02		17 ± 0.1	Sriket et al. (2007)
В	Nayachar, India		1.14 ± 0.1		111 ± 1.2	4.29 ± 0.2	334 ± 3	Mitra et al. (2010)
В	Panvel, India		0.12 ± 0.18			1.54 ± 1.1	22 ± 1.8	Meshram et al. (2014)
С	Tanzania	1.09	0.01		5.97	0.06		Saria (2017)

Spp. species, A Scylla serrata, B Penaeus monodon, C Fenneropenaeus indicus

salinity during the wet season promotes increased active uptake of major ions in the gills (Rainbow 1997). Since some trace metals can follow routes of uptake of major ions, decrease in salinity promotes uptake of such metal ions, irrespective of any physicochemical change affecting the release of the trace metal ions from inorganic complexation (Rainbow and Black 2002). This can also account for the high levels of trace metals in tissues sampled during the wet season (Tables S2 and S3), since the variations cannot be attributed to seasonal differences in trace metal enrichment. This is because a recent study showed that there are no significant seasonal variations in trace metal enrichment in the study area (Rumisha et al. 2016). The decrease in salinity is also associated with increased metal uptake in fish (Shazili 1995), molluscs (Kumar et al. 2015) and other crustaceans (Piazza et al. 2016; Rainbow and Black 2002).

Public health implications

The findings of this study clearly show that the measured concentrations of Cd, Cr and Pb in edible tissues of the giant mud crabs and tiger prawns do not exceed the recommended standards for human consumption (Tables 2 and 3). This is in line with the findings of previous studies on other marine and fresh water fish

from Tanzania (Machiwa 2003, 2005; Mdegela et al. 2009; Saria 2017). Generally, Tanzania export fishery products to Asia, Europe and other African countries, such as the DRC (Democratic Republic of Congo), Malawi and Rwanda. Export to the European Union, DRC and Asia accounts for about 45, 18 and 13% of the total export, respectively (URT 2016). The measured levels of Cd, Cr and Pb do not exceed the legal limits for import into these countries (EC 2011, 2014; PRC 2012). Based on the current trend of fish consumption in Tanzania, it was also revealed that the estimated monthly intake of Cd through consumption of giant mud crabs and giant tiger prawns do not exceed the provisional tolerable monthly intake level (PTMI) considered as safe by the Joint FAO/WHO expert committee on food additives (Fig. 4). Since the PTWI for As and Pb were withdrawn by the joint FAO/WHO expert committee on food additives (WHO 2011a, b), the target hazard quotient was used to evaluate risks associated with consumption of giant mud crabs and giant tiger prawns from the study area. It was revealed that the measured levels of As, Cd, Co, Cu, Mn, Pb and Zn do not present adverse health risks to shellfish consumers in the country (Fig. 5). This indicates that health risks for the measured trace metals are unlikely in areas where fish consumption does not exceed the average per capita



consumption (7.7 kg/person/year). Since arsenobetaine (nontoxic) accounts for 50-95% of total As content in crustaceans and teleosts (Calatayud and Llopis 2015), the THO values reported by Saria (2017) do not necessarily imply that As is likely to cause health risks to Blacktail snapper (Lutjanus fulvus), Indian Mackerel (Rastrelliger kanagurta) and Indian white shrimp (Fenneropenaeus indicus) consumers in the country. Apart from that, the high THQ values displayed by As and Cu (Fig. 5) gives an alarming sign and it indicates that potential health risks to people consuming shellfish above the per capita consumption cannot be ruled out. Considering that shellfish consumption in fishing communities is likely to exceed the average per capita consumption, regular monitoring is required. The fact that the hazard index (HI) exceeded one (Table S4) shows that although the cumulative exposure to multiple elements may not necessarily be additive (Antoine et al. 2017), the measured levels are alarming. Considering the nutritional and economic importance of these crustaceans, waste management systems and pollution control measures should be strengthened.

Conclusion

The present study showed significant bioaccumulation of As, Cu and Zn in edible tissues of the crustaceans S. serrata and P. monodon from the Tanzanian coast. This suggests that the crustaceans were either exposed to trace metals through the environment or food. It was also revealed that giant mud crabs were more contaminated compared to giant tiger prawns sampled from the same sites. Generally, giant tiger prawns migrate to the subtidal during low tide, while some mud crabs (mostly males) take refuge in mangrove burrows. Therefore, the crabs are more likely to mobilise sediment-bound metals and become exposed to trace metals from the sediments and pore waters compared to mobile prawns. The pattern of trace metal accumulation in giant mud crabs was consistent with the reported patterns of trace metal enrichment in the study area. This shows that the giant mud crab S. serrata is a good indicator of environmental quality and that it is suitable for biomonitoring programs. It was also revealed that the measured levels of Cd, Cr and Pb in muscle tissues of S. serrata and P. monodon do not exceed the maximum limits for consumption. Based on the current trends of fish consumption in Tanzania, As, Cd, Co, Cu, Mn, Pb and Zn

are not likely to present adverse health risks to an average mud crab and tiger prawn consumer. Apart from that, the high THQ displayed by As and Cu suggest that potential health risks to people consuming shellfish above the per capita consumption cannot be ruled out. This calls for strengthened pollution control measures and regular monitoring. Since arsenobetaine is known to account for 50–95% of total As content in crustaceans, analysis of As species would have given a more meaningful characterisation of the risks of As to shellfish consumers. Apart from that, the findings of this study provide baseline information for future studies.

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