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Heavy metal levels in the sediments of four Dar es Salaam mangroves Accumulation in, and effect on the morphology of the periwinkle, *Littoraria scabra* (Mollusca: Gastropoda)

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Abstract

Heavy metals were determined in the soft tissue and shells of the littorinid, *Littoraria scabra*, and in the sediments of four mangrove areas along the Dar es Salaam coastline where *L. scabra* was collected. Several metals accumulate, preferentially in the animals' soft body parts, but do not seem to affect the shell morphology of this species. Sediment-metal levels, measured in the direct vicinity of Dar es Salaam have increased dramatically over the last decade. Nonetheless, these levels are still lower compared to metal-sediment levels reported in polluted European and American estuaries. Soft-tissue metal levels detected in *L. scabra* are, nevertheless, with the exception of Cr and Zn, comparable to metal levels reported in other gastropod species. © 2001 Elsevier Science Ltd. All rights reserved.

Keywords: Heavy metals; Mangrove; *Littoraria Scabra*; Dar es Salaam

1. Introduction

Like many other developing countries, Tanzania, one of the less industrialised nations on the Western Coast of the Indian Ocean, is experiencing increasing impacts of environmental degradation. Tanzania has a few industrial centres, of which Dar es Salaam is the most important one. It is not only the major industrial, but it is also the largest, city of the country (Machiwa, 1992). Its population and level of industrial activities have steadily increased during the past three decades, with vast open spaces being converted into residential or industrial areas (Machiwa, 1992). This increased development has resulted in an uncontrolled disposal of domestic and industrial wastes (Ak'habuhaya and Lodenius, 1988; Machiwa, 1992). Waste is seldom treated and, as such, transported towards the coastal area by a series of rivers (Mgana and Mahongo, 1997). Msimbazi river, for instance, which flows through the Dar es Salaam industrial area, has an average sewage and industrial effluents rate of 256 m^3/h with peak values of 606 m^3/h, just before it enters the Indian Ocean (Ak'habuhaya and Lodenius, 1988). Pollutants such as PAHs, PCBs, and heavy metals, which are drained from the city towards the Ocean, tend to accumulate in the coastal sediments (Machiwa, 1992), where they pose a threat to the intertidal communities. Of special interest are the mangrove forests, located along the Tanzanian coast, as mangroves are one of the most productive and biodiverse wetlands on earth. The Tanzanian mangroves are not only affected by the pollution drain, but they are also cleared by man in order to obtain land for salt extraction (Ak'habuhaya and Lodenius, 1988). Thus, despite its ecological importance, the Tanzanian mangrove ecosystem is continuously being degraded and depleted (Ak'habuhaya and Lodenius, 1988).

Several mangrove areas are located along the Dar es Salaam coastline (Fig. 1). Msimbazi mangrove receives water from the Msimbazi river and is therefore likely to be a polluted area (Fig. 1). Mtoni mangrove is located along the Dar es Salaam harbour channel (Fig 1). Also, this mangrove is likely to be polluted due to the heavy traffic of fishing boats and ferries, while some municipal outflows are directly discharged in this area (Machiwa, 1992). Finally, Kunduchi and Mbweni mangroves are situated, respectively, 20 and

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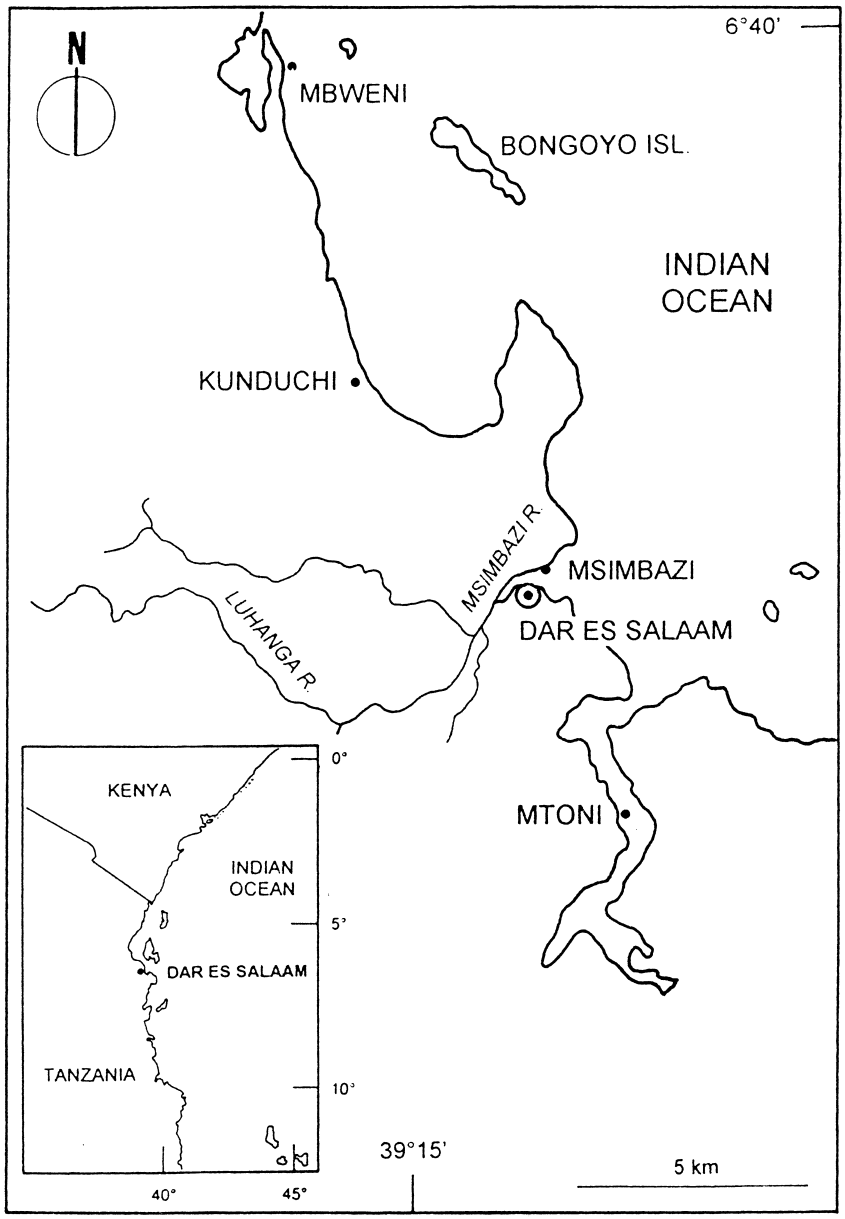


Fig. 1. Sampling area.

40 km north from the city centre (Fig. 1). With the exception of a few hotels located at Kunduchi, there is virtually no source of pollution visible at these mangrove areas.

Despite efforts undertaken to evaluate pollution levels in and around the Dar es Salaam area (Ak'habuhaya and Lodenius, 1988; Machiwa, 1992), very few studies have considered the accumulation of pollutants in, and the effect they have on the local marine fauna and flora. The mangrove areas along the coast of Dar es Salaam, each affected by different pollution levels, thus provide an excellent setting to test for such factors.

In this study, we investigate the morphological population structure of the dominant mangrove-dwelling periwinkle, *Littoraria scabra*, collected at all four mangrove areas, while we determine and compare sediment heavy-metal concentrations with each other and with heavy-metal concentrations measured in the soft body parts, as well as in the shells of *L. scabra*. In addition, a comparison of heavy-metal levels measured in the 1988 Msimbazi sediments (Ak'habuhaya and Lodenius, 1988) enables us to evaluate any changes in heavy-metal pollution around Dar es Salaam harbour during the past decade.

2. Material and methods

On 20–22 December 1998, *L. scabra* was collected at four similar mangrove sites along the Dar es Salaam coastline. These sites included, in order of expected pollution level: Msimbazi, Mtonoi, Kunduchi, and Mbweni (Fig. 1).

Table 1
Detection limits (DL) of ICP-AES in the determination of metals in the sediment, soft tissue, and recovery percentage of some of the elements, obtained by comparison with available standard reference material concentrations

Element	DL of ICP-AES using a V-groove nebulizer (μg/l)	% Recovery of the reference sample
Ag	0.5	
Al	0.4	
As	8	
Cd	0.2	106.74
Co	0.2	
Cr	0.2	
Cu	0.5	88.31
Fe	3	
Mn	0.05	91.98
Ni	0.6	
Pb	1	114.28
Sr	0.6	
Zn	0.1	97.19

A total of 80 specimens were collected, 20 at each site, and were morphometrically characterised. Shell height (HS) and width (WS), aperture height (HA) and width (WA), and shell-top height (HT) were measured to the nearest 0.1 mm using calipers, while the animals were sexed, based on the presence/absence of a penis, and weighed with [total weight (TW)] and without their shell [body weight (BW)].

An additional 120 specimens collected at the same sites were used for heavy-metal analysis. Shells and corresponding soft body parts were separated and were dried for 24 h at 60°C. Individual shell and soft-tissue samples were subsequently digested in a microwave oven, adding a mixture (5:1) of nitric acid (70%) and peroxic acid (30%). The digested samples were stored at 4°C until further analysis.

At each site, two sediment samples were taken, using a 5-cm ∅ hand core, to a depth of 10 cm. Wet sediment samples were centrifuged at 1000 rpm for 30 min. The supernatants were removed and the sediment particles were dried for 24 h at 60°C. Sediment samples were put in a Teflon bomb and digested in a microwave, adding a mixture (1:3) of nitric acid (70%) and HCl (37%), following the protocol described by Blust et al. (1988). After digestion, the solution was filtered through a 0.45-μm polyethersulfore-membrane filter, which was soaked for a few minutes in nitric acid (5%).

Table 2
Results of a two-way MANOVA, contrasting the random factor site with the fixed factor site

Effect	Wilks' λ	df_1	df_2	P
Site	0.443	21	190	.0001
Sex	0.891	7	66	.3411
Site × Sex	0.671	21	190	.1488

Table 3
Results of the post hoc Sheffé tests for interpopulational comparisons of morphological shell variability

Dependent variable	Site	Mtoni	Kunduchi	Mbweni
TW	Msimbazi	0.0012	0.5323	0.9475
BW		0.0215	0.3565	0.8696
HS		0.0165	0.1880	0.9487
WS		0.0100	0.1216	0.8467
HA		0.0139	0.6348	0.9337
WA		0.0122	0.1287	0.9629
HT		0.0135	0.0613	0.9939
TW	Mtoni		0.0705	0.0002
BW			0.5874	0.0019
HS			0.7555	0.0030
WS			0.7786	0.0006
HA			0.3763	0.0002
WA			0.4461	0.0001
HT			0.9456	0.0061
TW	Kunduchi			0.2333
BW				0.0795
HS				0.1104
WS				0.0558
HA				0.0148
WA				0.0405
HT				0.0053

For abbreviations, see Material and Methods section.

Silver, aluminium, arsenic, cadmium, cobalt, chromium, copper, iron, manganese, nickel, lead, strontium, and zinc were measured in the sediment, soft body parts, and shell samples by means of inductively coupled plasma atomic emission spectroscopy (ICP-AES), using a Varian Liberty Series II spectrophotometer. Analytical efficiency was checked using standard reference material (*Mytilus edulis*, CRM 278R) from the Community Bureau of Reference (BCR), digested and analysed in the same way as the samples (De Wit and Blust, 1998). The detection limits of

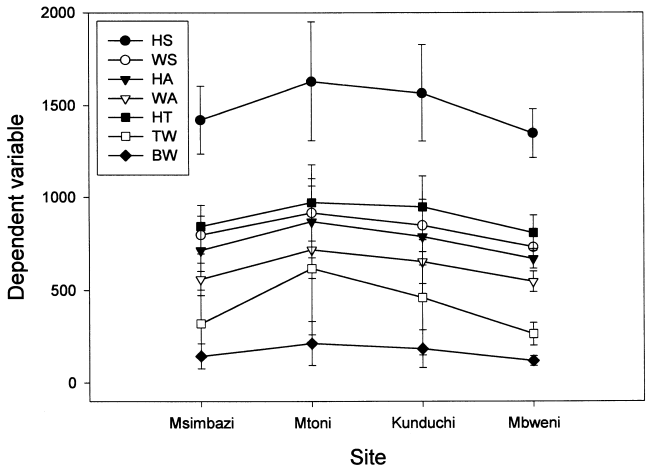


Fig. 2. Graphical representation of the dependent shell variables (HS, WS, HA, WA, HT, TW, BW). All variables are given as 10^{-2} mm except for TW and BW, which are given as 10^{-3} g.

the ICP-AES heavy-metal determination and recovery percentages are given in Table 1.

Morphometric patterns were investigated by means of a two-way multivariate analysis of variance (MANOVA), contrasting the fixed factor sex and the random factor site. Interpopulational differences were further investigated using a post hoc Sheffé test.

Possible differences between heavy-metal concentrations in the sediments, shells, and soft tissues, expressed as microgram per gram dry weight, were investigated using a Kruskal–Wallis single factor ANOVA.

The heavy-metal differentiation among the different mangroves was further analysed by means of a correspondence analysis executed with the NTSYS v. 1.80 software (Rohlf, 1993). With this method, the ordinates of the mangroves are taken on the basis of their sediment-metal distribution and of the metal distribution in the shells and soft tissues of *L. scabra*.

All statistical analyses, except for the correspondence analysis, were performed using the software package, Statistica v. 5.0 (Statsoft 1995).

3. Results

The results of the two-way MANOVA are summarised in Table 2. No sexual dimorphism is found, as only the factor site explains a significant part of the variation (Table 2). Morphological differences between sampling sites are almost entirely attributable to morphological differences between two sampling sites, Mtoni and Mbweni (Table 3). *L. scabra* collected from Mbweni weighs less and has a smaller and narrower aperture size with a more elongated shell compared to *L. scabra* collected at Mtoni (Table 3, Fig. 2).

For the heavy-metal sediment analysis, all elements, except for As and Cd, fell within the detection limits and could be measured in the sediments of at least one sampling site (Table 1, Fig. 3). Of these elements, only Ag, Fe, Mn, and Zn differed significantly between the sampling sites. Fe, Mn, and Zn concentrations decreased from Msimbazi to Mbweni (i.e., from a polluted to a clean area) (Figs. 3 and 4), whereas Ag did not reveal a clear geographical pattern (Figs. 3 and 4). All other elements were either only detected at Msimbazi (i.e., Cu and Pb) (Table 4) or revealed a trend of decreasing concentration from Msimbazi to Mbweni (i.e., Al, Co, Cr, Ni, and Sr) (Fig. 3). With the exception of Mn and Ni, the concentrations of all metals increased in the Msimbazi sediments between 1988 and 1999 (Fig. 3). This increase is most obvious for Al (from 530 to 6375 μg/g dry weight), Fe (from 630 to 3539 μg/g dry weight), and Cr (from 2.7 to 10.1 μg/g dry weight).

With respect to the heavy-metal soft-tissue analysis, all elements, except for Al, fell within the detection limits and were present in at least one site (Tables 1 and 4, Fig. 4). Significant sampling-site differences were observed for Cu, Mn, Sr, and Zn (Table 4). Of these elements Cu, Mn, and Zn decreased from Msimbazi to Mbweni (Fig. 4). Other elements revealed no clear geographical pattern (i.e., Ag, Cd, Cr, and Fe) (Fig. 4), were only detected in Msimbazi (i.e., As, Co, Ni, Pb) (Table 4), or revealed a trend of decreasing concentration towards the less polluted sites (i.e., Sr) (Fig. 4).

Except for Sr, all other metals could either not be detected in the shells of *L. scabra* (i.e., Ag, Al, As, Cd, Co, Cr, and Ni) (Table 4) or occurred at much lower

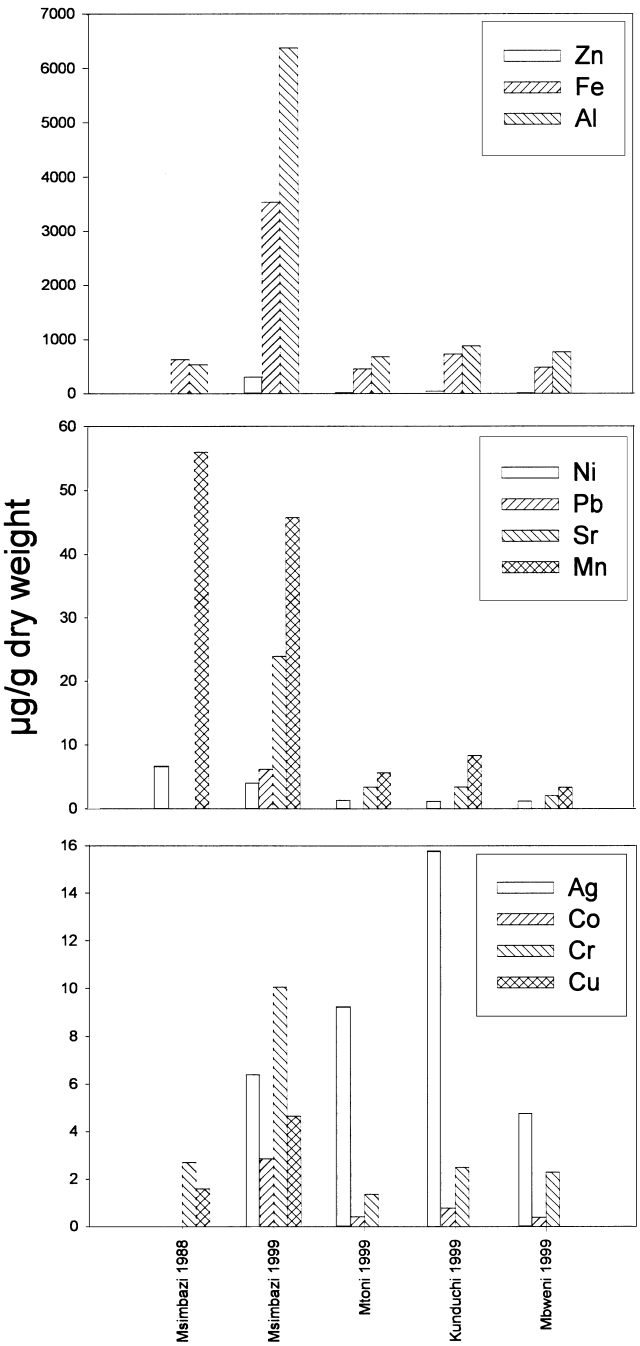


Fig. 3. Mean sediment heavy-metal concentrations (Msimbazi, 1988, 1999; Mtoni, 1999; Kunduchi, 1999; Mbweni, 1999).

concentrations compared to those measured in the soft tissue of *L. scabra* (Fig. 4). Only the shell Mn concentrations differed significantly between the different sites, decreasing from Msimbazi towards Mbweni (Table 4, Fig. 4). The shell concentrations of other elements did not reveal a geographical pattern (i.e., Cu, Fe, Sr, and Zn) (Fig. 4) or were only detected at Msimbazi (i.e., Pb) (Table 4).

The results of a correspondence analysis are graphically summarised in Fig. 5. The first axis is the most important one, explaining 91.1% of the total variation. It is mainly an expression of the Al, Fe, and Zn sediment measurements and is used to discriminate Msimbazi from the remaining sites. It indicates that these metals occur at the highest concentrations in Msimbazi, whereas the angle between the vectors illustrates that the three metals show a similar pattern of occurrence in the sediments of the four mangrove sites. Other important discriminating variables include Fe, Zn, and Sr, which were measured

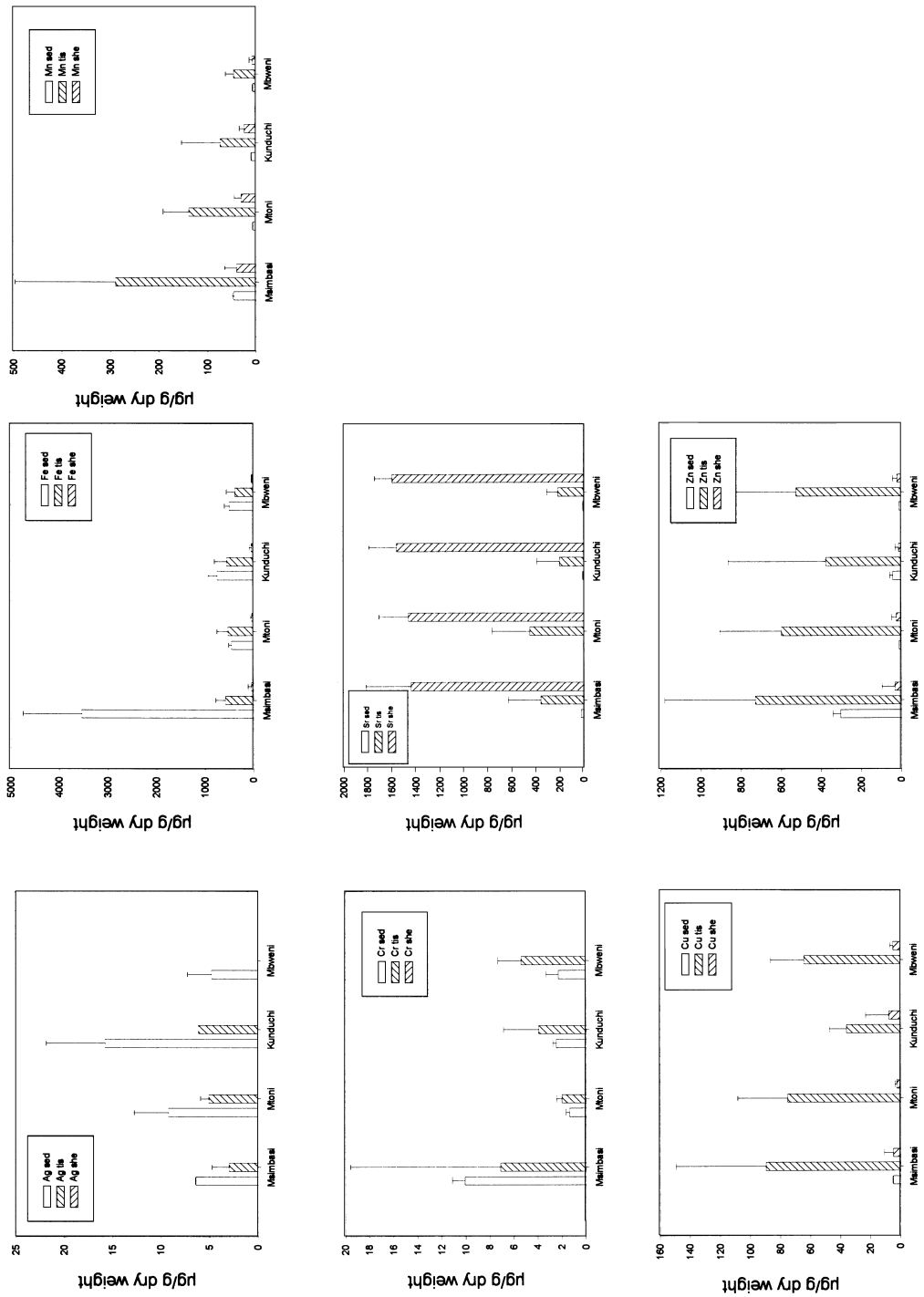


Fig. 4. Mean and standard deviations of sediment, soft-tissue, and shell heavy-metal concentrations.

Table 4
Results of the Kruskal–Wallis ANOVA for possible differences in heavy-metal concentrations in sediment, soft tissue, and shell measurements

	P		
Element	Sediment	Soft tissue	Shell
Ag	.0460	.7212	n.a.
Al	.2615	n.a.	n.a.
As	n.a.	–	n.a.
Cd	n.a.	.2330	n.a.
Co	.0719	–	n.a.
Cr	.2615	.0685	n.a.
Cu	–	.0001	.0599
Fe	.0460	.2615	.2847
Mn	.0460	.0001	.0001
Ni	.1116	–	n.a.
Pb	–	–	–
Sr	.2615	.0004	.2341
Zn	.0460	.0065	.1042

n.a., nonavailable, element that fell below our detection limits; (–) element measured in one population only (i.e., Msimbazi).

in the soft tissues, and Sr, which was measured in the shells of *L. scabra*. It reveals that the sediment and soft-tissue heavy-metal profiles of both Fe and Zn do not correspond well, whereas the tissue Fe and Zn measurements show a similar pattern of occurrence. Sr measured in the shells does not correspond well with Sr measured in the soft tissue of *L. scabra*, as illustrated by the opposing positioning of both vectors. Finally, the second axis is far less informative (6.3%), and its interpretation is therefore less straightforward.

4. Discussion

As was expected, Msimbazi mangrove is, with the exception of Ag, the most heavy-metal-polluted spot. Although the concentrations of several metals, except for Mn and Ni, have increased at this site during the last decade, sediment concentrations of Cd, Cu, and Pb, for instance, measured in industrialised estuaries in Europe and America, are still higher (Nolting et al., 1989; Benoit et al., 1994; Comber et al., 1995; Bayens, 1998). Nevertheless, comparable metal levels might be reached in a few years time, if the heavy-metal pollution, seen at Msimbazi keeps increasing. Zn, for instance, occurs already at comparable concentrations and is only detected at higher concentrations in the heavily polluted Rhine estuary (i.e., 714 μg/g dry weight) (Nolting et al., 1989). Several heavy metals, except for Ag, decrease in sediment concentration towards Mbweni, as they are probably trapped in the Msimbazi sediments and/or are dispersed on their way to Mbweni. Nevertheless, it should be stressed that sediment concentrations of a series of heavy metals, such as Al, Cr, and Fe, measured at the relatively "clean" sites of Kunduchi and Mbweni, are currently already comparable to particulate heavy-metal concentrations that were measured in the polluted Msimbazi sediments 10 years ago (Ak'habuhaya and Lodenius 1988).

Periwinkles may accumulate a considerable amount of these metals, either from the surrounding water or from their diet (Bryan et al., 1983). These metals may accumulate in the soft body parts, as well as in the shells of gastropods (Krolak, 1998). Likewise, *L. scabra* accumulates a substantial amount of metals. Not all metals accumulate uniformly in both the shell and soft body parts of *L. scabra*. While all metals that appeared in the shells also appeared in the tissues, some elements could not be detected in the shells. Compared to other gastropod species, *L. scabra* has relatively comparable soft-body levels of Ag, As, Cd, Co, Fe, Mn, Ni, and Pb (Bryan et al., 1983; Krolak, 1998; Wright and Mazon, 1999). Cu levels are relatively low, but Cr and Zn exceed previously published gastropod soft-body heavy-metal levels. Values reported in literature for Cr range from 0.03 to 0.7 μg/g dry weight measured in *Littorina littorea* collected, respectively, from the Teifi and Thames estuaries (GBR) (Bryan et al., 1983), and for Zn, from 51.7 to 553 μg/g dry weight measured in *Littorina littorea* collected, respectively, from the Stour and Fal estuaries (Great Britain) (Bryan et al., 1983; Wright and Mazon, 1999). Fe appears to be regulated by *L. scabra*, within the exposure window, as it occurs at comparable soft-body concentrations irrespective of the ambient Fe levels. The fact that an animal is able to regulate a metal does not necessarily mean that this metal is not harmful, as the regulation might put a demand on the animal's energy budget, possibly decreasing its growth and/or survival rate (Rainbow, 1997).

In any case, we did not find much intraspecific shell variation, which might indicate differences in growth and/or survival rate between the different sampling sites. The morphologies of the periwinkles differed significantly from each other at only two sites, Mtoni and Mbweni, with animals collected at the latter site being more elongated, having a smaller and narrower aperture, and weighing less. Shell variability in *L. scabra* is rather rare, and if it occurs, it is likely to be associated with habitat differences (Reid, 1986). Since Mbweni and Mtoni are seemingly comparable

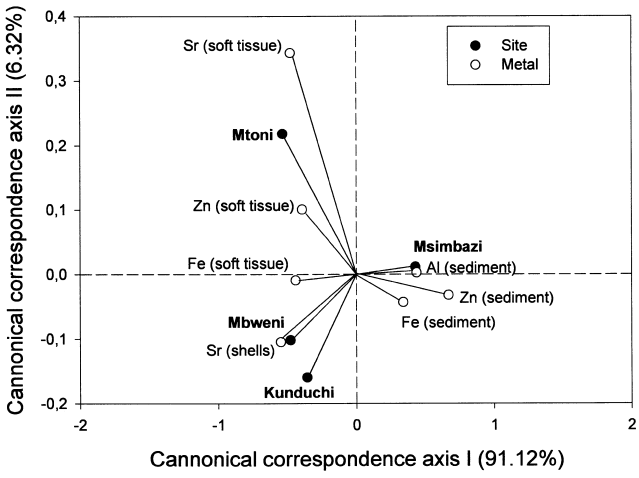


Fig. 5. Graphical representation of the results of a correspondence analysis.

mangrove habitats that differ in metal pollution, it might be tempting to attribute the observed morphological differences to differences in heavy metal or other types of pollution. However, this seems rather unlikely, as (1) Msimbazi is clearly the most polluted spot, but *L. scabra* from Msimbazi does not differ morphologically (i.e., shell measurements) from those at the Mbweni population, and (2) increased pollution by heavy metals would probably result in a growth rate and/or survival rate decrease (Widdows et al., 1995), shifting the more polluted animals towards smaller sizes, which is not the case here, as the smallest animals were collected at the less polluted site. Other factors besides pollution by heavy metals must be invoked to explain these results and will be analysed in the future. Care should, however, be taken with the current morphological results as only 20 individuals were analysed per population.

Based upon the increase in metal contamination levels over the last 10 years, it seems that pollution by heavy metals is rapidly increasing around the Dar es Salaam area. This increase is related to the rapid and uncontrolled industrial development that has been pursued without specific provision for handling the environmental pollution that is related to this industrial development (Mgana and Mahongo, 1997). Despite the currently relative low levels of heavy metals found in the sediments and soft body parts of *L. scabra* collected around the Dar es Salaam area, caution is necessary, and steps should be undertaken in order to control this pollution trend and ensure the protection of the biodiverse mangrove Tanzanian aquatic ecosystem.

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References

Ak’habuhaya J, Lodenius M. Metal pollution of river Msimbazi, Tanzania. Environ Int 1988;14:511–4.

Bayens W. Evolution of trace metal concentrations in the Scheldt estuary (1978–1995): a comparison with estuarine and ocean levels. Hydrobiologia 1998;366:157–67.

Benoit G, Oktay-Marshall SD, Cantu E, Hood EM, Coleman CH, Corapcioglu MO, Santschi PH. Partitioning of Cu, Pb, Ag, Zn, Fe, Al and Mn between filtered–retained particles, colloids, and solution in six Texas estuaries. Mar Chem 1994;45:307–36.

Blust R, Van de Linden A, Verheyen E, Decleir W. Evaluation of microwave heating digestion and graphite furnace atomic absorption spectrometry with continuum source background correction for the determination of iron, copper and cadmium in Brine shrimp. JAAS 1988;3:387–93.

Bryan GW, Langston WJ, Hummesrtone LG, Burt GR, Ho YB. An assessment of the gastropod *Littorina littorea*, as an indicator of heavy-metal contamination in United Kingdom estuaries. J Mar Biol Assoc UK 1983;63:327–45.

Comber SDW, Gunn AM, Whaley C. Comparison of the partitioning of trace metals in the Humber and Mersey estuaries. Mar Pollut Bull 1995; 30:851–60.

De Wit M, Blust R. Determination of metals in saline and biological matrices by axial inductively coupled plasma atomic emission spectrometry using microcentric nebulization. JAAS 1998;13:515–20.

Krolak E. Concentration of heavy metals in the snails *Lymnaea* (Radix) *peregra* (O.F. Mull) and *Lymnaea stagnalis* (L.) occurring in rivers near Siedlce town. Pol Arch Hydrobiol 1998;45:553–63.

Machiwa JF. Heavy metal content in coastal sediments off Dar Es Salaam, Tanzania. Environ Int 1992;18:409–15.

Mgana S, Mahongo S. Land-based sources and activities affecting the quality and uses of marine, coastal and associated freshwater environment. Report presented at the UNEP Regional Workshop, Institute of Marine Sciences, Zanzibar, 1997.

Nolting TF, Sunby B, Duinker JC. Minor and major elements in suspended matter in the Rhine and Meuse rivers and estuary. Neth J Sea Res 1989; 23:255–61.

Rainbow PS. Trace metal accumulation in marine invertebrates: marine biology or marine chemistry? J Mar Biol Assoc UK 1997;77:195–210.

Reid DG. The littorinid molluscs of mangrove forests in the Indo-Pacific region. London: British Museum of Natural History, 1986.

Rohlf FJ. NTSYS-PC. Numerical taxonomy and multivariate analysis system. Version 1.80 Setauket, NY: Applied Biostatistics, 1993.

Widdows J, Donkin P, Brinsley MD, Evans SV, Salkeld PN, Franklin A, Law RJ, Waldock MJ. Scope for growth and contaminant levels in North sea mussels *Mytilus edulis*. Mar Ecol: Prog Ser 1995;127: 131–48.

Wright P, Mazon CF. Spatial and seasonal variation in heavy metals in the sediments and biota of two adjacent estuaries, the Orwell and the Stour, in eastern England. Sci Total Environ 1999;226:139–56.