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Heavy Metals (Cd, Cu, Cr, Pb and Zn) in Meretrix meretrix Roding, Water and Sediments from Estuaries in Sabah, North Borneo

[1*]Mohd. Harun Abdullah, [1]Jovita Sidi and [2]Ahmad Zaharin Aris
[1] School of Science and Technology, Universiti Malaysia Sabah, 88999 Kota Kinabalu, Sabah, MALAYSIA
[2]Faculty of Environmental Studies, Universiti Putra Malaysia, 43400 Serdang, Selangor, MALAYSIA
* Corresponding author: harunabd@ums.edu.my

Abstract: Concentrations of heavy metals (Cd, Cu, Cr, Pb and Zn) in tissues of Meretrix meretrix Roding (M. meretrix R.), water and sediments from two estuaries were determined. One estuary is located in an urban area of Kota Kinabalu (Likas estuary) and the other in a rural district of Kota Belud (Kota Belud estuary), where both are in Sabah, North of Borneo island. M. meretrix R. is a kind of mollusk commonly consumed by the inhabitants of many parts of the island. This study indicated that species of mollusk, water and sediment from the urban estuary (Likas) contained higher concentration of some heavy metals than those in rural estuary (Kota Belud). It was found that the mollusk has a potential to be used as bioindicator for the contamination of Cd and Zn in water and sediment of an estuarine environment, as indicated by its high bioconcentration factors (BCFs) values. Overall, this species seemed to accumulate certain metals in its tissue and resisted the entry of others from its surrounding environment.

Key words: Heavy Metals; Mollusk, Water, Sediment, Bioconcentration Factor (BCF).

INTRODUCTION

Water bodies contaminated by heavy metals may lead to bioaccumulation in the food chain of an estuarine environment. Normally, such contaminants are transported from its sources through river system and deposited downstream. Since most of the pollutants could be mixed and became suspended solid and bottom sediment through sedimentation, therefore estuary is a potential sink for these pollutants for a long period of time (Morrisey et al., 2003). The presence of heavy metals in sediments can lead to greater environmental problem when the contaminated sediments resuspended and such metals are uptaken by filter feeder mollusk. Hence, consumption of such kind of mollusk may form a significant pathway to metals contamination in the human being and eventually poses greater health risk.

Mollusk, especially the species from bivalve class is frequently used as bioindicator in environmental monitoring. Bivalve is one of the organisms that has the criteria of potential bioindicator due to its ability to accumulate pollutants from its ambient. Usually, the level of pollutant accumulated in such an organism's tissue is used for assessing the level of pollution in its habitat (Al-Madfa et al., 1998; AbdAllah and Moustafa, 2002). A lot of studies on mollusks associated with heavy metal pollution have been done by many researchers, but few studies have been published related to Meretrix meretrix species. For example, Hung et al. (2001) compared trace metals contents in different species of mollusk species that included Meretrix lusoria, from Taiwan coastal area. On the other hand, Meretrix meretrix was one of the mollusks studied by Wang et al. (2005) in relation to the heavy metals contamination in coastal sites along the Chinese Bohai Sea. However, none of those studies specifically investigated the biological cumulative factor of such a species. The level of concentration of heavy metals in Meretrix meretrix Roding (M. meretrix R.) living in estuaries has never been investigated and reported. M. meretrix R. is a bivalve mollusk that live on the loose bottom of water bodies, and widely found in the coastal areas of Borneo island. They are filter feeder and thus very potential to accumulate toxic substances from water and sediment.

Aim and Purpose of the Research

This paper reports the results of our investigation of heavy metals in M. meretrix R., water and sediment samples collected from two estuaries of different environmentally background. This study would provide an understanding about the trace metals concentration in the tissues of M. meretrix R. in two different estuaries of Sabah. Interest in this species was motivated by the fact that it is one of the most commonly consumed mollusks in North of Borneo. Further more, there have never been any published reports on the background of heavy metals in such a mollusk species. The study on the potential of M. meretrix R. as bioindicator is an important effort that contributes to the findings of method in monitoring pollution in an estuarine environment of Tropical regions.

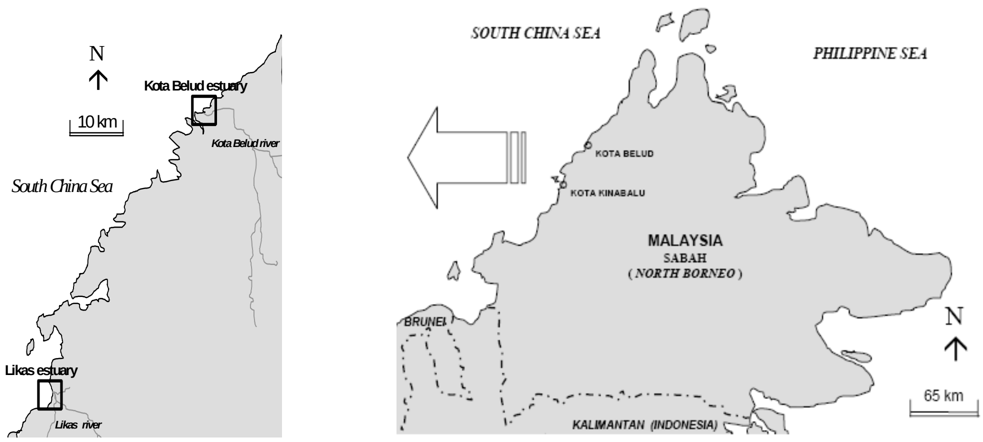

Figure 1. The geographical location of Likas and Kota Belud estuaries.

RESEARCH METHODOLOGIES AND METHODS

Study Area

The study areas are located at Likas estuary (between 13º07'N – 13º02'N and 137º35'E – 137º30'E) and Kota Belud estuary (between 6º 25'N – 6º 20'N and 116º 22'E – 116º 17'E) on the west coast of Sabah, Malaysia as shown in Figure 1. Likas estuary is shared by three rivers namely Likas river, Inanam river and Buat river. Likas river has been reported to be polluted by industrial effluent, whereas Inanam and Buat rivers were polluted by domestic wastewaters (Mokhtar et al., 1994; ECD and UMS, 2002). The Kota Belud estuary is believed to be in its natural condition where it receives Kota Belud river water that originates from the Crocker Range of Borneo.

Sampling and Analysis

Samples of M. meretrix R., water and sediments were collected from two estuaries, one situated in Kota Kinabalu (Likas estuary) and the other in Kota Belud district (Kota Belud estuary), both in the state of Sabah, Malaysia. Likas estuary represented an estuarine environment of an urban river, whereas Kota Belud estuary represented an estuarine of a rural river. Water samples were collected and filtered through a 0.45 µm membrane filter before being acidified with concentrated nitric acid in plastic bottles. Sediments were collected using grab sampler, and mollusks were obtained manually from the sites. During the samplings, both types of samples were kept in polyethylene bags and brought to laboratory for digestion and analysis.

In laboratory, the mollusk tissue samples were dried in oven at 80 ºC for 24 h and followed by digestion of 2 g of a dried sample in 10 ml of concentrated nitric acid at 60 ºC for 30 min. Subsequently, 1 ml of hydrogen peroxide was added to further oxidize any recalcitrant lipid materials in the sample. The digested tissues were then diluted to 100 ml with deionized water, and filtered. All samples were analyzed for cadmium (Cd), copper (Cu), chromium (Cr), lead (Pb) and zinc (Zn) using a Flame Atomic Absorption Spectrophotometer (Perkin Elmer). Heavy metals concentration in the water was expressed as milligram per liter (mgl-1), whereas those in the sediment and the mollusk tissue as microgram per gram (µgg-1).

RESULTS AND DISCUSSION

Heavy metals in mollusk tissue

In this study, a total number of 50 and 40 specimens of a mollusk species were collected from two estuaries, viz Likas estuary and Kota Belud estuary, respectively. Concentrations of heavy metals (Cd, Cu, Cr, Pb and Zn) in the M. meretrix R., collected from those estuaries were compared. The overall concentrations in this studied mollusk are provided in Table 1, and presented in µg g-1 dry weight. It was found that all of the metals concentration in the tissues of the mollusk collected from Likas estuary were significantly higher ($p<0.05$) than those specimens collected from Kota Belud estuary. It indicated that the Likas river has been polluted by heavy metals which eventually lead to bioaccumulation of those pollutants in the food chain of the estuary. Due to the nature of an estuary where river freshwater and seawater meets, it is

Table 1. Heavy metals concentration (µg g^{-1}) in *M. meretrix* R. from two estuaries.

Locations	Heavy metals				
	Cd	Cu	Cr	Pb	Zn
Likas estuary	3.27 ± 1.46 (1.24 – 6.78)	6.62 ± 1.30 (4.26 – 10.30)	2.28 ± 2.60 (0.15 – 9.32)	1.72 ± 0.58 (0.95 – 3.90)	106.7 ± 28.8 (50.3 – 223.5)
Kota Belud estuary	1.68 ± 0.65 (0.82 – 3.93)	5.78 ± 0.83 (3.80 – 8.02)	1.19 ± 0.93 (0.07 – 3.75)	1.09 ± 0.46 (0.26 – 2.27)	83.1 ± 26.0 (42.6 – 149.8)

Table 2. Heavy metals concentration (µg g^{-1}) in *M. meretrix* R. and other mollusks (*Anadara granosa* and *Crassostrea iredalei*) from Likas estuary.

Heavy metals	Mollusks species		
	M. meretrix R.	*Anadara granosa*	*Crassostrea iredalei*
Cd	3.27 ± 1.46	0.63 ± 0.44	0.68 ± 0.62
	(1.24 – 6.78)	(nd – 1.70)	(0.04 – 3.11)
Cu	6.62 ± 1.30	6.89 ± 1.66	13.22 ± 8.06
	(4.26 – 10.30)	(3.51 – 9.80)	(4.08 – 47.17)
Cr	2.28 ± 2.60	nd	nd
	(0.15 – 9.32)	-	-
Pb	1.72 ± 0.58	4.74 ± 2.37	4.61 ± 3.26
	(0.95 – 3.90)	(0.51 – 14.46)	(1.32 – 17.61)
Zn	106.7 ± 28.8	96.0 ± 17.0	397.2 ± 319.4
	(50.3 – 223.5)	(68.8 – 137.7)	(77.2 – 1445.5)

* nd = not detected

likely that the entrapment of pollutants in such an area to be expected.

Among the five metals tested for Likas and Kota Belud estuaries, Zn concentration was the highest (106.7 µg g-1 and 83.1 µg g-1, respectively), and Pb was the lowest (1.72 µg g-1 and 1.09 µg g-1, respectively). All the metals in both estuaries shared similar order of concentration, that is Zn > Cu > Cd > Cr > Pb. As indicated in Table 1, M. meretrix R. was found to have a large capacity for Zn and Cu intake. This could be explained by the role of those metals as essential elements for aquatic organism (Drexler et al., 2003). Comparatively, lower Pb, Cr and Cd contents in the tissue possibly due to its toxicity and are non essential metals to mollusk.

The metals content in M. meretrix R. of Likas estuary were also pictured with other edible mollusk species as a comparative study. For such a purpose, mollusks namely Anadara granosa and Crassostrea iredalei tissues were analyzed. The concentration of Cd and Cr in M. meretrix R. tissues (Table 2) were found to be significantly higher ($p<0.05$) than Anadara granosa and Crassostrea iredalei. The tissues of Anadara granosa contained Cd and Zn in the concentration of 0.626 ± 0.438 µg g-1 and 96.0 ± 24.7 µg g-1, respectively, whereas Cr was not detected. On the other hand, the content of Cu and Pb in this mollusk were 6.89 ± 1.66 µg g-1, and 4.74 ± 2.37 µg g-1, respectively, indicated a higher concentrations than M. meretrix R..

The comparison between M. meretrix R. and Crassostrea iredalei showed that the concentrations of Cd and Cr were found to be higher in the tissues of M. meretrix R. (Table 1 and Table 2) . The Crassostrea iredalei contained Cd in the concentration of 0.68 ± 0.62 µg g-1, and Cr was not detected. However, apparently the concentration of Cu (13.22 ± 8.064 µg g-1), Pb (4.61 ± 3.26 µg g-1) and Zn (397.2 ± 319.4 µg g-1) in Crassostrea iredalei were significantly higher ($p<0.05$) than the M. meretrix R.. Further explaination related to bioaccumulation of those metals in the tissues of this mollusk are provided under the bioconcentration factor (BCF).

Overall, it was noted that the M. meretrix R. contained the highest concentration of Cd and Cr among the studied mollusks (Table 2). The concentration of heavy metals in the tissues of this mollusk decreased in the order of Zn > Cu > Cd > Cr > Pb. Based on those results, it showed that the magnitude of the heavy metals accumulation in mollusks tissues depend on the type of heavy metals and the species of the mollusks. Both M. meretrix R. and Anadara granosa are the species of mollusk that live on the loose bottom of water bodies, where they burrow in the sand or mud, and they are known as infaunal deposit feeder (Han et al., 1996). As a consequence, they are very much exposed to the bottom water and the heavy metals accumulated in sediments. Such a condition has a major impact on the accumulation rate of heavy metals in those mollusks. On the other hand, Crassostrea iredalei is a bivalve mollusk that live by sticking to any solid materials such as rocks and concretes in the water environment. As a result, it is more exposed to water and suspended solids, and accumulates heavy metals by trapping food from water through filter-feeding process. Thus, the accumulation of heavy metals were different between species and possibly exhibited different accumulation strategies for trace elements.

Heavy metals in water and sediments

1. Water

Table 3. Concentration of heavy metals (mgl^{-1}) in water from two estuaries.

Heavy metals	Estuaries	
	Likas	Kota Belud
Cd	0.006 ± 0.003 (0.001 – 0.010)	0.004 ± 0.005 (0.001 – 0.015)
Cu	0.042 ± 0.001 (0.041 – 0.043)	0.013 ± 0.003 (0.009 – 0.020)
Cr	0.009 ± 0.002 (0.006 – 0.012)	0.009 ± 0.004 (0.002 – 0.013)
Pb	0.100 ± 0.005 (0.094 – 0.108)	0.010 ± 0.011 (0.002 – 0.035)
Zn	0.043 ± 0.026 (0.014 – 0.107)	0.031 ± 0.005 (0.23 – 0.036)

Table 4. Concentration of heavy metals (μgg-1) in fine sediments from two estuaries.

Heavy metals	Estuary	
	Likas	Kota Belud
Cd	4.04 ± 1.18 (2.5 – 5.7)	0.40 ± 0.10 (0.3 – 0.6)
Cu	76.9 ± 45.0 (45.5 – 142.5)	77.6 ± 4.5 (68.6 – 82.7)
Cr	19.40 ± 1.51 (16.3 – 21.1)	36.06 ± 3.23 (31.5 – 41.8)
Pb	25.98 ± 1.63 (22.2 – 27.4)	13.54 ± 1.09 (12.0 – 15.1)
Zn	368.7 ± 43.9 (286.0 – 420.0)	53.2 ± 3.2 (47.9 – 57.6)

Overall, the heavy metals concentraion from both estuaries water were low, except Cu and Pb, as shown in Table 3. In this study, the concentrations of Cu (0.042 ± 0.001 mgl-1) were higher and Pb (0.100 ± 0.005 mgl-1) were lower than those reported earlier (Cu<0.01 mgl-1 and Pb=0.39 mgl-1; ECD and UMS, 2002), respectively. Though Zn in Likas water was slightly higher than Kota Belud, the range was still within normal concentration of natural water. The Cr quantity occurred within the normal value for natural water (0.04-0.05; Sarmani, 1985). According to Stickney (2000), the optimum values of certain metals for aquaculture, in terms of parts per million are: <0.15 (Cd), <0.1 (Cu) and <0.25 (Zn). High values of Pb in Likas estuary indicated pollution of the estuarine water bodies. The recommended safe level of dissolved Pb is <0.05 ppm (Krenkel and Novotny, 1980). Nevertheless, low concentration of metals in water might not necessary reflected that the area was pollution free. The biota lives in such an area might have accumulated the metals from water from time to time. Such a situation could be observed from the higher concentration of heavy metals in the mollusks tissue found in the Likas estuary.

2. Sediments

Fine fraction (<0.63μm) of sediment contains heavy metals more than total sediment (Forstner and Solomons, 1980). Thus, in this study the analyses were done on the surface sediments of fine fraction. Table 4 shows the results of analyses on the content of heavy metals in the sediments of Likas and Kota Belud estuaries. It was found that the concentration of Cd, Pb and Zn in the sediments of Likas estuary were significantly higher ($p<0.05$) than those from Kota Belud estuary. The contents of Cu and Cr in Kota Belud sediments were possibly contributed by the natural geological sedimentation of the area where Mountain Kinabalu and copper mining area are located at the upstream of the river. Basically, the concentration of the heavy metals in the Likas estuary decreased in the order of Zn > Cu > Pb > Cr > Cd, whereas in Kota Belud the order was Zn > Cu > Cr > Pb > Cd. The findings of this investigation revealed that the higher concentration of heavy metals (such as Cd, Pb and Zn) in the sediment of Likas concurred with the pattern of those metals found in the tissue of M. meretrix R..

Accumulation of metals in *M. meretrix* R.

1. Metal concentration ratio of mollusks to water and sediments

A mollusk's ability to accumulate metals from a medium into its tissue can be estimated using bioconcentration factor (BCF). By comparing BCF, we can compare the ability of those mollusks in taking up metals from water and sediments. Tolerant mollusk tend to restrict water-sediment-tissue transfers, and thus have less accumulation in its tissue. In this study, BCFmollusk-water (BCFm-w) refers to the concentration of a particular metal in a tissue of a mollusk per concentration of that metal in water. The categories of BCFm-w are presented as: High potential if BCFm-w > 1000; Moderate potential if 1000 > BCFm-w > 250; and Low potential if BCFm-w.< 250. Table 5 shows value of high potential factor (BCFm-w = 2500-2700) for Zn in both estuaries, indicating that the cumulative factor of the studied heavy metals were higher in the M. meretrix R. compared to the water. Such values indicated that the mollusk accumulated the essential metals in considerable quantity in its tissue. On the other hand, Pb exhibited low potential factor (BCFm-w = 25-27) in both estuaries. Though Pb (0.100 ± 0.005 mgl-1) was recorded with the highest concentration in the Likas estuarine water compared to the other heavy metals (Table 3), yet it presented low BCFm-w value. Such a low value indicates that the mollusk was unwilling to absorb the metals from water possibly due to Pb toxicity.

BCFmollusk-sediment (BCFm-s) is defined as the ratio of metal concentration in the mollusk to that in sediment. In this study, it was found that M. meretrix R. from Kota Belud estuary showed higher BCFm-s values for Cd (BCFm-s = 4.2) and Zn (BCFm-s = 1.6), indicating that the mollusk contained more metals than the sediment. Similar pattern was also found for Cd and Zn in Likas estuary (Table 5), although the BCFm-s values were less than 1.0. It should also be noted that the concentration of these metals in the sediment and M. meretrix R. from Kota Belud were lower than Likas estuary, as shown in Table 3 and Table 4. Thus, this investigation indeed indicated that the M. meretrix R. were able to accumulate higher quantity of Cd and Zn than Cu, Cr and Pb. The BCFm-s values for Cu, Cr and Pb were lower than1.0, which means limited ability of these heavy metals to be accumulated by the mollusk.

2. Comparison of BCF in M. meretrix R. with other mollusks

The M. meretrix R. cumulative factor of heavy metals from water were compared with other mollusks (Anadara granosa and Crassostrea iredalei) found in the study area of Likas estuary, as shown in Table 6. All of the mollusks have high potential factor (BCFm-w > 1000) for Zn, exhibiting that this element possibly essential in its tissue. Comparatively, M. meretrix R. recorded higher factor value for Cd, and low factor value for Pb. Such a condition indicated that this species is capable of accumulating that metal (Cd) in relatively larger quantity, which indeed show that it is potential bioindicator for Cd pollution in water. On the other hand, it has low potential factor for the accumulation of Pb from water bodies.

Table 7 compares the M. meretrix R. cumulative factor of heavy metals from sediments with other mollusks (Anadara granosa and Crassostrea iredalei) found in the Likas estuary. The BCFm-s values obtained by all of the mollusks were below 1.0 (except Crassostrea iredalei for Zn), indicating higher content of those metals were found in the sediments compared to the tissues of the mollusks. Such a condition possibly reflected that the mollusk were rather selective in accumulating metals into their tissues. Comparatively, an interesting observation indicated that M. meretrix R. from the study area showed highest BCFs-m for Cd

Table 5. Accumulation of heavy metals in *M. meretrix* R. from two estuaries.

Heavy metals	Bioconcentration factor			
	Water (BCF_{m-w})		Sediment (BCF_{m-s})	
	Likas	Kota Belud	Likas	Kota Belud
Cd	5.5×10^2	4.2×10^2	0.8	4.2
Cu	1.6×10^2	4.4×10^2	0.1	0.1
Cr	2.8×10^2	2.1×10^2	0.1	0.1
Pb	1.7×10^1	1.1×10^2	0.1	0.1
Zn	2.5×10^3	2.7×10^3	0.3	1.6

Table 6. Comparison of BCF of heavy metals from water between mollusks in the study area.

Heavy metals	Cumulative factor		
	M. meretrix R.	*Anadara granosa*	*Crassostrea iredalei*
Cd	5.5×10^2	1.0×10^2	1.1×10^2
Cu	1.6×10^2	1.6×10^2	3.1×10^2
Cr	2.8×10^2	nd	Nd
Pb	1.7 x 10	4.7 x 10	4.6 x 10
Zn	2.5×10^3	2.2×10^3	9.2×10^3

* nd = not detected

Table 7. Comparison of BCF of heavy metals from sediments between mollusks in the study area.

Heavy metals	Cumulative factor		
	M. meretrix R.	*Anadara granosa*	*Crassostrea iredalei*
Cd	0.8	0.2	0.2
Cu	0.1	0.1	0.2
Cr	0.1	t.k	t.k
Pb	0.1	0.2	0.2
Zn	0.3	0.3	1.1

(BCFs-m = 0.8; Table 7), approaching the value of one. This showed that such a species was efficient in taking Cd, despite the toxicity of that element. On the other hand, Zn as an essential nutrient for mollusk, was recorded with BCs-m = 0.3. Based on the results in Table 7, apparently this species was tolerant to the intake Cu, Cr and Pb (all recorded low cumulative factor with BCFs-m = 0.1). Therefore, it indicated that the accumulation of heavy metals were different between species and might have performed different accumulation strategies for trace elements.

CONCLUSIONS

The findings of this study showed that M. meretrix R., water and sediment from the urban estuary (Likas) contained higher concentration of some heavy metals than those in rural estuary (Kota Belud). Based on BCFs values obtained from both estuaries, it was found that this species of mollusk has the potential to be used as bioindicator for the contamination of Cd and Zn in water and sediment of an estuarine environment. Low BCFs values for Cu, Cr and Pb means limited ability of these heavy metals to be accumulated by the mollusk. It showed that the accumulation of heavy metals in a mollusk depends on the species of heavy metals and organism. Overall, this study indicated that M. meretrix R. resists accumulation of certain metals and promotes that of others as shown by the BCFs values.

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