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HEAVY METAL ACCUMULATION IN MOLLUSCS AND SEDIMENT FROM UPPANAR ESTUARY, SOUTHEAST COAST OF INDIA

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ABSTRACT

The concentration of (Cd, Co, Cu, Fe, Mg, Mn, Pb, Zn) were analyzed in sediments, shells and tissues of the molluscs *Meretrix meretrix, Crassostrea madrasensis* and *Cerithidea cingulata* from two stations of Uppanar Estuary, southeast coast of India. The concentrations of the heavy metals analyzed exhibited variations in sediments, tissues and shells of the study animal from all the two stations. Zn and Cu concentration were below the alarming level. In the present study Mg content was found to be higher in shell and tissue. The Fe content was found to be higher in sediment in both the stations, but it was the second highest metal recorded in shell and tissue. Cd and Co was minimum accumulated of all the metals estimated in sediment and animal whereas Mg was recorded maximum (548.3 ± 5) in *C. cingulata* shell. All the metals analysed were within the safety level. The combined correlation of both the stations between the sediment vs. sediment, tissue vs. tissue, shell vs. shell were positively correlated and significant at $P < 0.001$ and $P < 0.05$ level. In addition to continuous contribution of pollutants due to human activities, industries may have a significant contribution of these metals in all the stations. As suggested by many reported studies found in the literature, regular biomonitoring of heavy metal concentrations at these sites is needed since *Meretrix meretrix*, *Crassostrea madrasensis* are a popular commercial bivalve in India.

Key words: Uppanar, Heavy metals, sediment, gastropod, bivalve.

RESUMEN

Se analizaron las concentraciones de Cd, Co, Cu, Fe, Mg, Mn, Pb, Zn en sedimentos y en conchas y tejidos de los moluscos *Meretrix meretrix, Crassostrea madrasensis* y *Cerithidea cingulata* en dos estaciones de muestreo del estuario del río Uppanar, en el sudeste de la costa de la India. Las concentraciones de los metales pesados analizados mostraron diferencias en los dos puntos de muestreo tanto en sedimentos como en conchas y tejidos. Las concentraciones de Zn y Cu estuvieron por debajo de niveles considerados alarmantes. El contenido de Mg en el presente estudio fue alto en conchas y tejidos. El contenido de Fe fue muy alto en los sedimentos en ambas estaciones, siendo el segundo metal en concentración en las conchas y tejidos. Los niveles de Cd y Co fueron mínimos tanto en sedimentos como en muestras animales y la concentración máxima fue la de Mg (548,3 ± 5) en conchas de *C. cingulata*. Todos los metales analizados estuvieron dentro del nivel de seguridad. La correlación combinada de ambas estaciones analizando sedimento contra sedimento, tejido contra tejido, concha frente a concha mostraron correlaciones positivas con significación a nivel P <0,001 y P <0,05. Las actividades industriales pueden contribuir significativamente a los valores encontrado. Según lo sugerido por muchos estudios reportados en la literatura, la biomonitorización regular de las concentraciones de metales pesados en estos sitios es necesaria dado que tanto *Meretrix meretrix* como *Crassostrea madrasensis* son bivalvos comerciales populares en la India.

Palabras clave: Uppanar, metales pesados, sedimentos, gasterópodos, bivalvos.

INTRODUCTION

More than half the world's population live within 60 km of the shoreline, and this could rise to three-quarters by the year 2020. Adverse anthropogenic effects on the coastal environment include eutrophication, heavy metals, organic, microbial pollution and oil spills. Consequently, levels of contaminants in the marine environment are increasing continuously. In order to establish adequate coastal management programs, it is important to characterize the environment of concern chemically. The extent of contamination can be assessed by measuring pollutant concentrations in water, sediments and organic tissue samples.

High metal concentrations in the environment are the result of both natural and anthropogenic sources. The accumulation of metals in water and sediments affects various organisms in the environment, influencing their functions in several different ways (Regoli and Principato 1995). As compared with the open sea, lagoons are more subject to pollution, particularly by heavy metals from industrial, agricultural and urban origin. These sources contribute to the lagoon environment either directly or by means of water courses discharging their contents into the lagoon. Near-shore sediments are found in a wide variety of environments (bays, lagoons, deltas). The sediments at the water-sediment interface are more important to biological fauna as compared with subsurface sediments since meiofauna live above the reduced zone in sediments. Therefore, the composition of surface sediments has a significant influence on the living conditions of some marine organisms. The trace metal results obtained by sediment analysis, unlike sea water analysis, are generally above the detection limit and contamination risks are significantly reduced (Brugmann, 1981). Sediment data are therefore utilized as a total for assessing sources and distribution of some elements in aquatic environments.

It is well known that molluscs accumulate organic and metallic pollutants at concentrations several orders of magnitude above those observed in the field environment (Bryan *et al*., 1983). Owing to the widespread application of bivalve molluscs as biomonitoring organisms in the aquatic environment, they have been the subject of several studies on the interaction of heavy metals. Fewer studies have been done on gastropod molluscs, some of which are also considered as useful biomonitors of certain metals (Bryan *et al*., 1983). Most metals are generally concentrated many times over within an organism's soft tissue, rather than the shell, and so the vast majority of studies concentrate on the soft tissue. However, some studies of the shell material have also been conducted and many authors suggest that shells can provide a more accurate indication of environmental change and pollution; they exhibit less variability than the living organism's tissue and they provide a historical record of metal content throughout the organism's life time. This record still preserved

Figure 1: *Map Showing the Study Area.*

after death (Huanxin, *et al.,* 2000). The fact that a metal would be regulated by living organisms does not, however, signify its harmlessness. Exposures to environmental concentrations lower than the threshold of non-regulation can induce physiological changes: variations of enzymatic activities and alteration of hormonal metabolism (Amiard- Triquet *et al*., 1986). Heavy metals that are present at concentrations common in ambient marine waters can cause adverse effects in shellfish. Such effects can significantly impact the trophic structure of a biological community. Heavy metals uptake is dependent on both geochemical and biological factors.

The present investigation was undertaken to study the status of bioaccumulation of heavy metals in the soft tissue and shell of three different species of molluscs apart from the sediment in Uppanar estuary.

STUDY AREA

The Uppanar estuary is situated at Cuddalore which is about 25 km away from the Parangipettai coast. Uppanar estuary is also an open type estuary and the width of the mouth is around 30 m. The Cuddalore fishing harbour is situated near the mouth of the Uppanar estuary, which is one among the important fishing harbours of Tamil Nadu. SIPCOT industrial park (State Industries Promotion Council of Tamil Nadu) is located on the northern bank of Uppanar estuary covering an area of about 520 acres with 44 industries, which include chemicals, petrochemicals, pharmaceutics, pesticides, fertilizers and metal processing industries (Fig 1).

Station 1- SIPCOT (Lat. 11º 41' N; Long 79º 45' E): It is situated near where the SIPCOT industrial units are discharging. Municipal and domestic sewage from the nearby Cuddalore town and coconut retting effluents are also released here.

Station 2– Landing centre (Lat 11º 44' N; Long 79º 46' E): This station lies near the mouth of the estuary, which is nearly 1km away from station 1, and in close proximity to the fishing harbour.

MATERIALS AND METHODS

Trace metal content (Magnesium, Iron, Zinc, Manganese, Cadmium, Cobalt, Lead, Copper) were analysed in the soft tissue, shell of three different species of molluscs such as *Cerithidea cingulata, Crassostrea madrasensis* and *Meretrix meretrix* and sediment collected from two stations of Uppanar estuary, southeast coast of India.

Sediment

Sediment samples were collected using a pre cleaned and acid washed PVC corer and immediately kept in pre cleaned and acid washed polythene bags, which were sealed and kept in an ice box until further analysis in the laboratory. Sediment samples were washed with metal free double distilled water. The sediment samples were then dried in an oven at 60ºC for 5-6 hours. Dried sediment samples were ground in a glass mortar and reduced into fine particles.

Tissues and shells

The molluscs were collected by hand picking. The soft tissue was removed from the shells with knife and dried at 60ºC. The dried tissue was reduced into fine powder in a pestle and mortar and was stored in dessicator for further analysis. The shell of individual species of molluscs was also finely ground. The resulting powder was selected, using a plastic sieve with 0.2mm opening size and was stored in dessicator for further analysis.

To estimate the trace metal content samples were digested (1g) with conc. HNO_3 and conc. $HClO_4$ (4:1) and analysed in optical emission spectrophotometer (optima 2100DV) (Topping, 1973). The metal content in sediment was determined by following the method of Chester and Hughes (1967). The values were expressed in ppm. The standard deviation and the correlation coefficient analyses were performed.

RESULTS

Station 1: SIPCOT (Table 1)

Sediment

In Station 1, the values of heavy metals in sediment were ranging from 0.005 ppm (Cd) to 65.45 ppm (Fe).

Meretrix meretrix

In *M. meretrix*,tissue recorded the highest magnesium content of 87.79 ppm and the lowest cobalt concentration of 0.037 ppm, while the concentration of the heavy metals in the shell was ranging from 34.85 ppm (Mg) to 0.004 ppm (Cd).

Crassostrea madrasensis

Among the heavy metals estimated, magnesium reported the highest values of 128.1 ppm and Cobalt, the lowest value of 0.023 ppm in the tissue. At the same time,

Table 1:

STATION 1 - SIPCOT

Species	Cd	Co	Cu	Fe	Mg	Mn	Pb	Zn
	ppm							
M. meretrix **Tissue**	0.055 ± 0.01	**0.037 ± 0.001**	0.331 ± 0.05	12.71 ± 0.5	**87.79 ± 10**	0.929 ± 0.05	0.278 ± 0.01	1.799 ± 0.05
M. meretrix **Shell**	**0.004 ± 0.01**	0.006 ± 0.001	0.109 ± 0.05	4.691 ± 0.5	**34.85 ± 10**	0.274 ± 0.05	0.083 ± 0.01	3.639 ± 0.05
C. madrasensis **Tissue**	0.123 ± 0.01	**0.023 ± 0.001**	0.311 ± 0.05	6.424 ± 0.5	**128.1 ± 10**	1.131 ± 0.05	0.285 ± 0.01	3.838 ± 0.05
C. madrasensis **Shell**	**0.002 ± 0.001**	0.014 ± 0.001	0.112 ± 0.05	18.35 ± 0.5	**74.56 ± 10**	1.597 ± 0.05	0.034 ± 0.01	0.293 ± 0.05
C. cingulata **Tissue**	**0.026 ± 0.01**	0.333 ± 0.001	3.006 ± 0.05	25.65 ± 0.5	**144.5 ± 10**	22.24 ± 0.05	0.072 ± 0.01	7.769 ± 0.05
C. cingulata **Shell**	0.099 ± 0.01	**0.003 ± 0.001**	0.102 ± 0.05	1.609 ± 0.5	**13.69 ± 10**	0.348 ± 0.05	0.070 ± 0.01	0.993 ± 0.05
Sediment	**0.005 ± 0.001**	0.029 ± 0.001	0.091 ± 0.05	**65.45 ± 0.5**	12.05 ± 10	0.560 ± 0.05	0.101 ± 0.01	0.185 ± 0.05

the shell recorded the maximum magnesium content of 74.56 ppm and minimum cadmium content of 0.002 ppm.

Cerithidea cingulata

In this species, the tissue recorded the highest concentration of 144.5 ppm (Mg) and lowest of 0.026 ppm (Cd) whereas in shell it was 13.69 ppm (Mg) and the lowest of 0.003 ppm (Co) respectively.

The correlation was made between the sediment and the animals. The result between the sediment vs. *C. cingulata* shell and *M. meretrix* shell was found not significant at any level. But, the sediment vs. *C. madrasensis* tissue ($r = 0.782$, $P < 0.01$) and shell ($r = 0.782$, $P < 0.01$) were positively correlated and significant at $P < 0.01$ level, whereas the sediment vs. *M. meretrix* tissue ($r = 0.703$, $P < 0.05$) and *C. cingulata* tissue ($r = 0.667$, $P < 0.05$) showed positive correlation and significant at $P < 0.05$ level.

Station 2: Landing center (Table 2)

Sediment

In station 2, the values of heavy metals in sediment were ranging from 0.003 ppm (Cd) to 64.07 ppm (Fe).

Meretrix meretrix

In *M. meretrix*, the tissue recorded the highest magnesium content of 52.46 ppm and the lowest cadmium concentration of 0.016 ppm, while the concentration of the heavy metals in the shell was ranging from 6.159 ppm (Mg) to 0.002 ppm (Cd).

Crassostrea madrasensis

Among the heavy metals estimated, magnesium reported the highest values 59.36 ppm and Cadmium, the lowest value of 0.014 ppm in the tissue. At the same time, the shell recorded the maximum magnesium content of 71.84 ppm and minimum cadmium content of 0.002 ppm.

Cerithidea cingulata

In this species, tissue recorded the highest concentration of 29.70 ppm (Mg) and the lowest of 0.001 ppm (Cd) whereas in shell it was 548.3 ppm (Mg) and the lowest 0.006 ppm (Cd) respectively.

The correlation was made between the sediment and the animals. The result between the sediment vs. *C. madrasensis* tissue was found not significant at any level. But, the sediment vs. *C. madrasensis* shell ($r = 0.888$, $P < 0.01$), *M. meretrix* tissue ($r = 0.925$, $P < 0.01$), shell ($r = 0.856$, $P < 0.01$), *C. cingulata* tissue ($r = 0.841$, $P < 0.01$) and shell ($r = 0.835$, $P < 0.01$) were positively correlated and significant at $P < 0.01$ level.

One way ANOVA was performed between the two stations of the study area in Uppanar. In between stations (SIPCOT and Landing center), there was much variation in heavy metal concentrations and showed they were not significant for both the animal and sediment. The values obtained in sediment (F=2.100, p=NS), *M. meretrix* tissue (F=.034, p=NS), *M. meretrix* shell (F=1.154, p=NS), *C. cingulata* tissue (F=.869, p=NS), *C. cingulata* shell (F=.745, p=NS), *C. madrasensis* tissue (F=.000, p=NS) and *C .madrasensis* shell (F=.000, p=NS).

Table 2:
STATION 2 - LANDING CENTER

Species	Cd	Co	Cu	Fe	Mg	Mn	Pb	Zn
	ppm							
***M. meretrix* Tissue**	0.024 ± 0.01	**0.016 ± 0.01**	1.009 ± 0.05	5.476 ± 2	**52.46 ± 5**	0.978 ± 0.5	0.091 ± 0.01	13.09 ± 0.5
***M. meretrix* Shell**	**0.002 ± 0.01**	0.003 ± 0.01	0.099 ± 0.05	2.159 ± 2	**6.159 ± 5**	0.164 ± 0.251	0.033 ± 0.01	0.667 ± 0.5
***C. madrasensis* Tissue**	**0.014 ± 0.001**	0.027 ± 0.001	0.543 ± 0.05	6.346 ± 2	**59.36 ± 5**	2.004 ± 0.5	0.073 ± 0.01	1.661 ± 0.5
***C. madrasensis* Shell**	**0.002 ± 0.001**	0.016 ± 0.001	0.158 ± 0.05	14.91 ± 2	**71.84 ± 5**	4.578 ± 0.5	0.032 ± 0.01	0.874 ± 0.5
***C. cingulata* Tissue**	**0.001 ± 0.0005**	0.006 ± 0.001	0.131 ± 0.05	9.187 ± 2	**29.70 ± 5**	4.638 ± 0.5	0.017 ± 0.01	0.757 ± 0.5
***C. cingulata* Shell**	**0.006 ± 0.001**	0.069 ± 0.001	0.840 ± 0.05	9.500 ± 2	**548.3 ± 5**	6.188 ± 0.5	0.051 ± 0.01	1.766 ± 0.5
Sediment	**0.003 ± 0.001**	0.047 ± 0.001	0.167 ± 0.05	**64.07 ± 2**	23.75 ± 5	0.632 ± 0.5	0.068 ± 0.01	0.515 ± 0.5

DISCUSSION

Trace metals can be divided in to essential elements and non-essential elements. Essential elements occur naturally in all organisms. In high doses essential elements can be poisonous and cause hazardous effects on organisms. The non-essential elements do not have any positive effects on organisms and they are harmful already in low doses. They can inhibit an essential element to bind to enzyme and disturb the normal enzymatic function in the body. Concentration of the heavy metals in sediment, tissues and shells of molluscs from two stations of Uppanar estuary were determined. One station is located in a zone where there is not much industrial or anthropogenic activity (Landing center) whereas the other receives effluents from an industrial area (SIPCOT).

The sediment of two stations had higher concentration of Iron (65.45 ppm) and Magnesium (64.07 ppm). Zinc and Copper were also recorded in considerable amount at all the stations**.** Similar kind of accumulation has been recorded by Ananthan, *et al.,* (2006), in Vellar estuary for the Iron in sediment (285 to 355 µg g^{-1}). Wherein the highest value was recorded during monsoon and lowest during pre monsoon, the value is higher than that of the present study in both the stations. Iron plays an important role as an essential element in all systems from invertebrates to humans but increasing of iron in the environment may result the bioaccumulation in the marine organisms such as bivalves, gastropods.

Sediment cadmium level in the present study were recorded lower values at both stations (0.005 - 0.003 ppm). This can also be attributed to industrial inputs and it is worth mentioning here that cadmium is not an essential element for plants, animals and human beings. The levels of the heavy metals Copper (Cu), Zinc (Zn), Lead (Pb), Cadmium (Cd), Chromium (Cr), Nickel (Ni), Iron (Fe) and Manganese (Mn) were determined in coastal water, sediments and soft tissues of the gastropod limpet, *Patella caerulea* and the bivalve, *Barbatus barbatus*, from seven different stations in the western coast of the Gulf of Suez. The highest accumulated metals were Fe, Zn and Mn in both *P. caerulea* and *B. barbatus*, while the lowest one was Cd. The accumulation of metals was more pronounced in *P. caerulea* than *B. barbatus* (Mohamed and Ahmed, 2006). The concentration of Iron in sediments was higher at station 1 than at station 2 in Uppanar estuary and this could be due to the nature of sediments, its texture and size as the former station are clayey in nature while the later is sandy. The other metals such as Zinc, Manganese, Cobalt, Lead, and Copper, exhibited mediate concentrations. Metals such as cadmium and cobalt were below the detectable limit at all the stations.

The concentration in the sediment had a major influence in the molluscan tissues and shells. Bivalves and gastropods are frequently used as biomonitors for heavy metal pollution in the estuarine environment. In the present investigation, molluscs (*C. cingulata, C. madrasensis* and *M. meretrix)* were subjected to heavy metal accumulation studies. The present study has recorded maximum concentration of Magnesium (13.69 ppm - 548.3 ppm) in the tissues and shells of all the molluscs at both the stations of Uppanar. However, Iron, Lead and Zinc were also present in considerable quantities. Cobalt and Cadmium concentration was the lowest. Cadmium and Cobalt were detected in significant quantities in Station1 and Cadmium was detected in all the samples in Station 2.

In all the cases, the concentration of metals in tissues was higher than the shells. Between the animals, *M. meretrix* showed more accumulation of metals when compared to *C. cingulata* and *C. madrasensis*. But there was significant variation in metal concentration between tissue and shell where tissue concentration was dominating. Comparing the concentration in biota collected from both stations, landing center had more concentration of Magnesium (548.3 ppm) when compared to the other station. This might be due to the fishing and other activities, whereas other station did not show much alarming concentrations. At both stations in the estuary, no significant correlation was obtained between the sediment and the tissue/shell concentration of the metals studied in the present study.

In concordance with the present study, several relevant studies have been made by researchers earlier. The heavy metal accumulation in the gastropod *C. scabridum* from Kuwait coast has been analysed (Bu-olayan and Thomas, 2001) and it was reported that the concentration of the cadmium in the gills was ranging between (7.06 $\mu g/g^{-1}$ and 0.90 $\mu g/g^{-1}$), which is comparatively lower than that of the present study in both the stations at Uppanar.

In the present study, the concentration of heavy metals in the tissue was generally more than that of the shell concentration. The accumulation of Cadmium in the bivalve *B. pharonis* was found as 0.0058-0.0605 $\mu g/g^{-1}$ (Augusto *et al.,* 2006), which is lower than that recorded during the study in Uppanar. In the present study, the molluscan tissues and shells accumulation for Cadmium was below detectable limit. The concentrations of heavy metals (Fe, Mg, Zn and Cu) were estimated in sediment, shell and tissue of the mollusc *Telescopium telescopium* from two stations of Vellar estuary (Kesavan *et al.,* 2009). The concentrations of the heavy metals analyzed exhibited the following decreasing order Fe>Mg >Zn>Cu in sediments and Mg >Fe>Zn>Cu in tissue and shell of the study animal. But during the present analyses it was found in reverse trend for these metals.

Trace metals were determined in the two most abundant species of bivalve molluscs along the Atlantic coast of southern Spain *(Donax trunculus and Chamelea gallina)* and in the sediments where they live. The results show that the area near the mouth of the Huelva estuary is where the highest metal concentrations are found in sediments and in the two bivalve species. The concentration of Cr, Cu, Pb, Zn, As and Hg in *D. trunculus* were significantly higher ($p < 0.05$) than in *C. gallina*; however, *C. gallina* contained more Ni and Cd. In both species the most abundant elements were Cu and Zn, while Hg showed the lowest values. There is a significant correlation ($p < 0.05$) for concentrations of Cu, Pb, Zn and Hg in *D. trunculus* and *C. gallina* relative to their concentrations in surface sediments (Jose *et al.,* 2005).

The bioaccumulation of heavy metals such as Magnesium, Iron, Zinc and Copper concentration in different body parts and shell of *C. melo* and also studied the sediment of its habitat (Shanmugam *et al*., 2007). The bivalves take up metals from solution and suspended material, but oysters (*C. rhizophorae*) are particularly recommended as biomonitors given their strong accumulation patterns for many trace metals, their large size and their local abundance. The intensity of heavy metals (Mg, Fe, Mn, Zn, Cd, Co, Cu, Pb) were estimated in sediment, shell and tissue of the molluscs *Cerithidea cingulata, Crassostrea madrasensis* and *Meretrix meretrix* from two stations of Vellar estuary, southeast coast of India. The concentrations of the heavy metals analyzed exhibited variations in sediments, tissues and shells of the study animal from the 2 stations. Zn and Cu concentration were below the alarming level, whereas Mg (274.0 ± 12) content was higher in shell and tissue (Kesavan *et al*., 2010)

In molluscs, size and weight along with the age is an important factor which determines bioaccumulation and has been reported by several researchers (Jordaens *et al*., 2006). This study revealed that the levels of heavy metals are elevated in some stations that received effluent from human activities and these contributed the levels of metals in the estuary. Sixty percent of metals analyzed measured from surface sediment are from anthropogenic activities. The level of heavy metals needs to be monitored as the development of shrimp ponds and aquacultural activities are in progress in the area.

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