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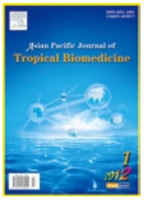

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Heavy metal monitoring using *Nerita crepidularia*–mangrove mollusc from the Vellar estuary, Southeast coast of India

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ABSTRACT

Objective: To estimate levels of the heavy metals such as Fe, Al, Mg, Mn, Cd, Cu, Pb, Zn, Cr and Ni in sediment, shell and soft tissues of the mangrove gastropod *Nerta crepidularia* (*N. crepidularia*) in mangroves at Vellar estuary, Southeast coast of India for the period of January 2007 to December 2007 covering four seasons. **Methods:** To estimate the trace metal content, samples were digested (1g) with conc. HNO_3 and conc. $HClO_4$ as 4:1 and analysed by Optical Emission Spectrophotometer (Optima 2100DV). **Results:** The order of accumulation was Fe> Al> Mg> Mn> Cd> Cu> Cr> Zn> Ni> Pb. The variation in the accumulation of metals in sediment was due to their geographical location .The degree of accumulation of trace metals in the animal tissues was as follows: Fe> Al> Mg> Mn> Cd> Cu> Cr> Zn> Ni> Pb. The higher concentrations of metals in the monsoon season could be due to the heavy inflow of freshwater, which brought lot of effluents from irrigation channels and municipal drainage. The results of ANOVA showed a positive significant relationship between sediment and tissues in their metal concentration. Shells also had some practical advantages, the shell size and weight along with the age are important factors which determined bioaccumulation. The concentrations of Fe, Cd, Ni and Pb in shells were found to be 88.54–176.46, 0.34–0.76, 2.44–14.58, 0–0.22 μg/g, respectively. **Conclusions:** Since *N. crepidularia* accumulates high concentrations of heavy metals, in general, than the ambient environment and Cu & Mn, in particular, it may be used as a biomonitor of certain heavy metals in mangrove environment.

1. Introduction

Pollution in the marine environment has become a new discipline of interest to organisms. Due to the green revolution in India, the usage of pesticides, fungicides and weedicides showed their hands at the maximum and these chemicals containing heavy metals are profoundly leached from the agricultural land drain into the sea through river and estuary. As various species of fish, shellfish serve as important food for human beings it could become a potential hazard to human health[1].Estuaries often act as efficient reservoirs of river borne and marine-derived pollutants[2]. Mangroves found in the tropics between land sea are salt-tolerant plants, which are considered to be important to coastal fisheries both in terms of their role as breeding and nursery ground for fish and as a source of food, so that a complete understanding of their environment and the human impacts upon it is essential. Among pollutants, heavy metals have been of interest because of their toxicity, persistence and prevalence on the environment[3].

Sediments are one of the major sinks of trace metals in the aquatic environment. Partioning among different physicochemical forms plays an important role in determining the biological effects of sediment bound metals and in determining metal exchange between sediments and water. Near-shore sediments have found recent interest because they can be considered as repositories for many chemical species. Organisms can take up contaminants from the water or in particles and accumulate them in the body[4]. Molluscs are common, highly visible, ecologically and commercially important on a global scale as food and as non-food resources. Most studies that provide comparisons among taxonomic groups indicate that bioaccumulation of pollutants in molluscs are in general, greater than that in fish. Their use as biological indicators is particularly appropriate due to their effective accumulation of metals[5]. The use of gastropods as biomonitors of heavy metal pollution has been widely reported in the literature. This is

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due to their characteristics from ecological and biological points of view which are advantageous for biomonitoring.

Nerita crepidularia (*N. crepidularia*) is usually found in the tropical intertidal area including mangrove trees and intertidal mudflats. Although their abundance and distribution are usually found along the Southeast coast of India, Tamil Nadu, its potential as a biomonitor has not been reported in the literature. The objective of the present study was to estimate the heavy metal concentrations such as Fe, Al, Mg, Mn, Cd, Cu, Pb, Zn, Cr and Ni in the soft tissues and shells of *N. crepidularia* along the Southeast coast of India, and exploring the possibilities of using this gastropod as a bioindicator for heavy metal accumulation.

2. Materials and methods

In the present study season – wise accumulation of metals such as Fe, Al, Mg, Mn, Cd, Cu, Pb, Zn, Cr and Ni was estimated in sediment, shell and soft tissues of the mangrove gastropod *N. crepidularia* for the period of January 2007 to December 2007 covering four seasons (January– March, post monsoon; April–June, summer; July–September, premonsoon; October to December, monsoon).

Sediment samples were collected during low tide from the mid tidal water mark (*i.e.* core mangrove areas) of Vellar estuary using a pre–cleaned and acid washed PVC (50 cm) corer and kept immediately in an ice box for further analysis. The sediment samples (about 8 cm) were washed by free metals with double distilled water. Then samples were dried in an oven at 60℃ for about 5–6 hours and then they were ground in a glass mortar to reduce into fine particles. The molluscs (50 snails) collected from the mangroves at each season were collected from the study area by hand picking. The soft tissue was removed from the shells with a plastic knife and dried at 60℃. The dried tissue were reduced into fine powder using pestle and mortar and was then stored in desiccator for further analysis. The shell of individual samples was also finely ground. The resulting powder was selected, using a plastic sieve with 0.2mm opening size and was stored in desiccator for further analysis. To estimate the trace metal content, samples were digested (1 g) with conc. HNO_3 and conc. $HClO_4$ as 4:1 and analysed by Optical Emission Spectrophotometer (Optima 2100DV)[6]. The values were expressed in μg/g. Three samples were analysed for every seasons. The average and mean values were calculated.

Statistically the concentration of heavy metals between the seasons, sediment, tissue and shell was correlated. All the statistical analyses of the data as well as the drawing of curves were made using SPSS V.16 software (SPSS Corporation), Origin V.7.5 (Marcrocal Origin) and Excel 2003. One–way ANOVA test were performed (5% confidence level) between the seasons, metals, sediment, tissue and shell for the heavy metal concentration. The correlation coefficient was also calculated between the sediment, tissues & shell among different seasons.

3. Results

3.1. Sediment

In the present study it is noted that Pb was not recorded in summer but noticed during monsoon and post monsoon.

The concentration of Fe was ranged between 7 424 μg/g and 11 444 μg/g with a mean of (10 028.10± 557.00) μg/g. Maximum concentration (11 444 μg/g) of Fe in sediment was noted during monsoon and minimum (7 424 μg/g) during summer (Figure 1).

The concentration of Zn was found varying between 36.14 μg/g and 56.14 μg/g with a mean of (39.28±0.60) μg/g. The concentration of Zn in the sediment showed its highest value (56.14 μg/g) during monsoon and lowest value (36.14 μg/g) in summer (Figure 2).

The concentration of Mn was ranging between 153.98 μg/g and 217.84 μg/g with a mean of (178.28±1.12) μg/g. Mn concentration in sediments of mangroves showed its peak value (217.84 μg/g) during monsoon and low value during summer (153.98 μg/g) (Figure 3).

The concentration of Cu was fluctuating between 11.28 μg/g and 37.24 μg/g with a mean of (16.28±1.64) μg/g. The highest concentration (37.24 μg/g) of Cu in the sediment was observed during monsoon and lowest concentration (11.28 μg/g) during summer (Figure 4).

The concentration of Al was found varying from 6 292 to 10 506 μg/g with a mean of 8 (542.00±487.58) μg/g. During the study period, the concentration of Al in sediment was found to be high (10 506 μg/g) in monsoonal months and low (6 292 μg/g) in summer months, whereas premonsoon and postmonsoon showed moderate values (Figure 5).

The Mg value was ranging from 2 708 to 4 700 μg/g with a mean of (2 954.61±345.24) μg/g. The maximum Mg (4 700 μg/g) concentration was recorded in monsoon and minimum (2 708) was noted at summer (Figure 6).

The Cd concentration was fluctuating between 2.38 μg/g and 25.98 μg/g with a mean of (9.15±5.83) μg/g. The Cd concentration was recorded high (25.98 μg/g) during monsoon and low (2.38 μg/g) during summer (Figure 7).

The Cr concentration was found lying between 7.10 μg/g and 31.44 μg/g with a mean of (9.44±3.11) μg/g. The maximum concentration of Cd was recorded during monsoon (31.44 μg/g) and minimum during summer (7.10 μg/g) (Figure 8).

The Ni concentration was reported between 15.02 μg/g and 40.42 μg/g with a mean value of (1.64±1.20) μg/g. The Ni concentration was recorded high (15.02 μg/g) during monsoon and low (40.42 μg/g) during summer (Figure 9).

The Pb concentration was ranging between 0 μg/g summer and 2.42 μg/g monsoon with a mean of (0.98±0.41) μg/g (Figure 10).

3.2. Tissues

The Fe concentration was noticed from 1 121.2 to 2 060.4 μg/g with a mean of (1 615.00±464.25) μg/g. Fe concentration in whole body tissue showed its maximum value (2 060.4 μg/g) in December (2007) and minimum in May

Table 1.
Analysis variance (ANOVA) between the seasons for sediment , tissue and shell.

Source of variation		*SS*	*df*	*MS*	*F*	*P*–value	*F* crit
Sediment	Between groups	30 417.80	3	10 139.27	0.245 8	0.863 8	2.866 3
	Within groups	1 484 849.00	36	41 245.81			
Tissue	Between groups	1 530.71	3	510.24	0.281 3	0.838 6	2.866 3
	Within groups	65 307.82	36	1 814.11			
Shell	Between groups	25.21	3	8.40	0.097 4	0.961 0	2.866 3
	Within groups	3 106.16	36	86.28			

Table 2.
Analysis variance (ANOVA) between the sediment and tissues and shell.

Source of variation		*SS*	*df*	*MS*	*F*	*P*–value	*F* crit
Tissue	Between groups	150 539.9	1	150 539.9	7.421 8	0.008 0	3.963 5
	Within groups	1 582 106.0	78	20 283.4			
Shell	Between groups	233 519.4	1	233 519.4	11.995 9	0.000 9	3.963 5
	Within groups	1 518 398	78	19 466.65			

Table 3.
Analysis variance (ANOVA) between the tissues and shell.

Source of variation	*SS*	*df*	*MS*	*F*	*P*–value	*F* crit
Between groups	9 071.37	1	9 071.37	10.112 5	0.002 1	3.963 5
Within groups	69 969.90	78	897.05			

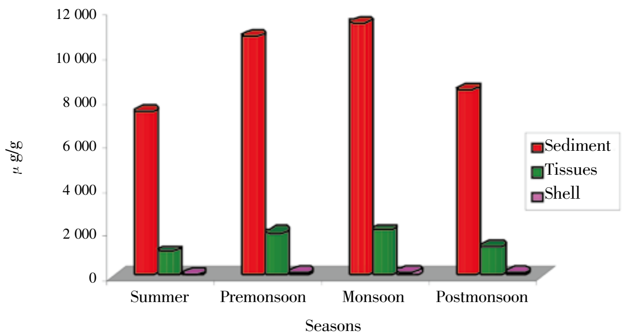

Figure 1. Iron content in different seasons.

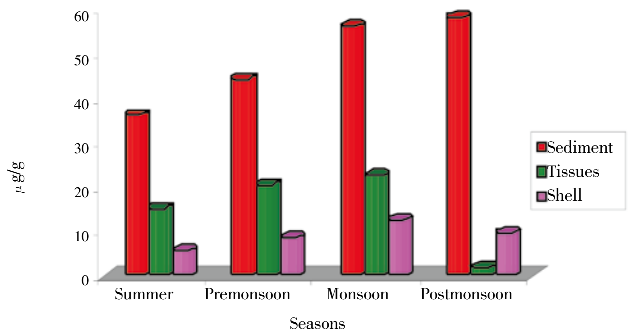

Figure 2. Zinc content in different seasons.

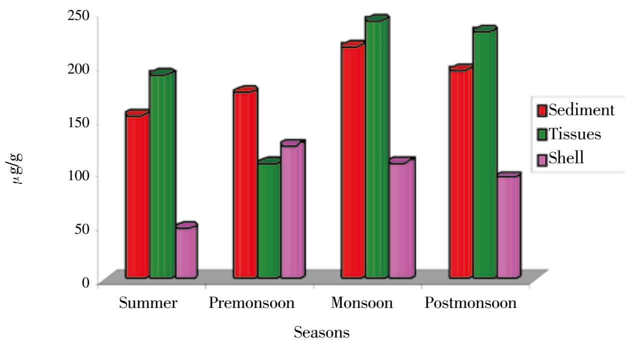

Figure 3. Manganese content in different seasons.

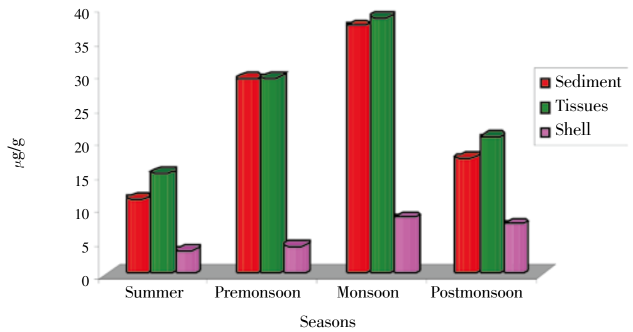

Figure 4. Copper content in different seasons.

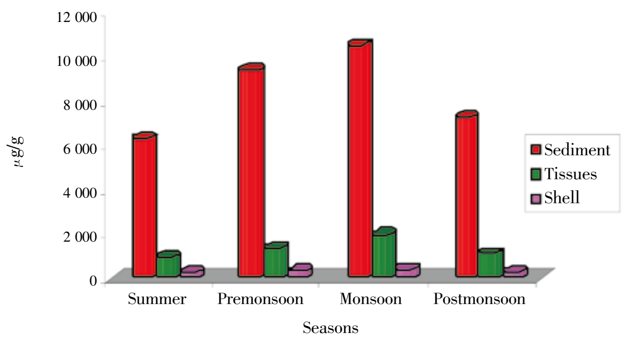

Figure 5. Aluminium content in different seasons.

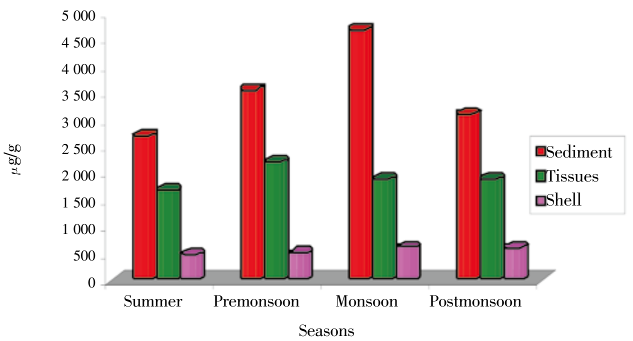

Figure 6. Magnesium content in different seasons.

(2007) (1 121.2 μg/g) (Figure 1).

The concentration of Zn was fluctuating between 15.26 μg/g and 22.50 μg/g with a mean of (15.02±9.29) μg/g. The concentration of zinc in the body tissues of *N. crepidularia* showed the highest value of 22.50 μg/g in December (2007) and lowest value (15.26 μg/g) in May (2007) (Figure 2).

The concentration of Mn was reported ranging from 191.96 to 242.42 μg/g with a mean of (193.82±60.95) μg/g. Tissues of *N. crepidularia* showed the maximum concentration (242.42 μg/g) during December (2007) and minimum (191.96 μg/g) during May (2007) (Figure 3).

The Cu concentration was ranging from 15.16 to 38.56 μg/g with a mean of (25.90±10.30) μg/g. In *N. crepidularia* the values of Cu showed the minimum concentration (15.16 μg/g) in May (2007). The maximum concentration was noted in December (2007) (38.56 μg/g) (Figure 4).

The Al concentration was ranging between 946.60 μg/g and 1 973.20 μg/g with a mean of (1 353.15±447.67) μg/g. The maximum concentration of Al was recorded during monsoon, (December 2007) (1 973.2 μg/g) and minimum during summer, (May 2007) (946.6 μg/g) (Figure 5).

The concentration of Mg was ranging from 1 678 to 2 214 μg/g with a mean of (1 913.00±222.03) μg/g. The maximum (2 214 μg/g) concentration was recorded in December (2007) and minimum (1 678 μg/g) was noted in May (2007) (Figure 6).

The Cd concentration was found varying from 0.48 to 2.44 μg/g with a mean of (1.51±0.98) μg/g. During the study period, the concentration of Cd in tissues was found to be high (2.44 μg/g) in monsoonal months and low (0.48 μg/g) in summer months, whereas premonsoon and postmonsoon showed moderate levels (Figure 7).

The Cr concentration was fluctuating from 1.50 to 2.90 μg/g with a mean of (2.19±0.61) μg/g. The Cr concentration was recorded high (2.90 μg/g) during December (2007) and low (1.50 μg/g) during May (2007) (Figure 8).

The Ni concentration showed the fluctuation from 4.14 to 20.22 μg/g with a mean of (15.38±7.53) μg/g. The highest (20.22 μg/g) concentration of Ni was found in December (2007), then the values were decreasing to the minimum of 4.14 μg/g in May (2007). The premonsoon and postmonsoon showed moderate level of Ni concentration (Figure 9).

The Pb concentration was reported from 0 to 0.44 μg/g with a mean of (1.60±2.20) μg/g. Pb concentration in whole body tissue showed its maximum value (0.022 mg/L) in December (2007). Whereas no Pb was noted during May (2007) (Figure 10).

3.3. Shell

In general, the shell reported only low concentrations when compared with that of the sediment and tissue. In shell, Mg recorded the maximum value of 612.40 μg/g and Pb, the minimum value of 2.44 μg/g. The concentration of other metals analysed are presented in Figure 1-10.

3.4. Statistical analysis

The heavy metal concentration in sediment between seasons varied insignificant (F=0.245 8, $P<0.05$) (Table 1). Likewise, the results of one way ANOVA between the seasons for tissues and shell reported an insignificant variation (F=0.281 3, $P<0.05$; F=0.097 4, $P<0.05$, respectively) (Table 1).

The results of ANOVA of heavy metal concentration in the sediment & tissue, sediment & and tissue & shell showed significant variation (F=7.421 8, 11.995 9 and 10.112 5, all $P<0.05$ respectively) (Table 2&3).

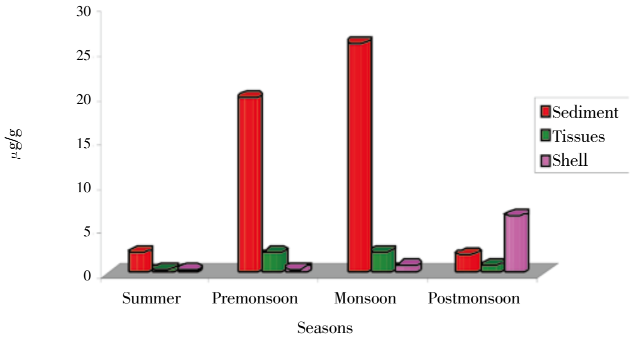

Figure 7. Cadmium content in different seasons.

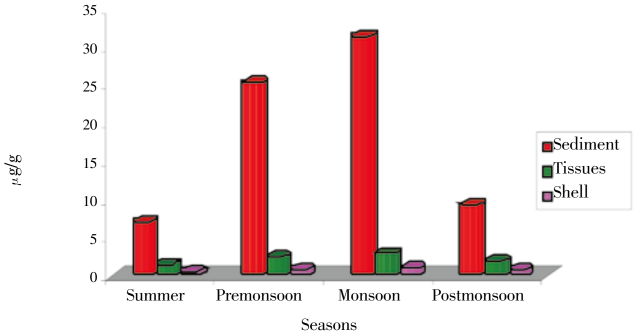

Figure 8. Chromium content in different seasons.

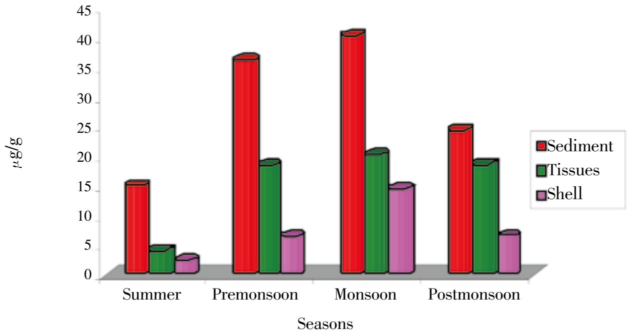

Figure 9. Nickel content in different seasons.

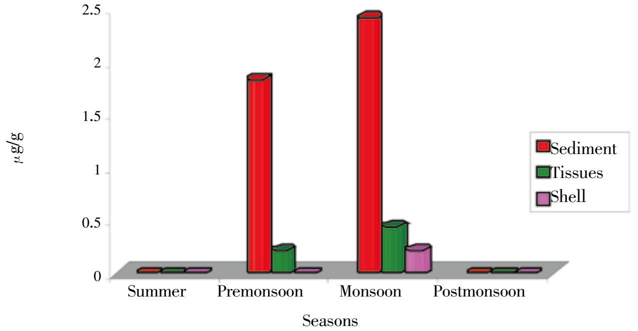

Figure 10. Lead content in different seasons.

In the present study, the relationship obtained between the sediment and tissues was found significant at 0.001 level ($r = 0.850$), between the tissues and shell ($r=0.865$, $P<0.001$) and sediment and shell ($r = 0.562$, $P<0.001$).

4. Discussion

Sediments are one of the major sinks of trace metals in the aquatic environment and may be good indicators of long and medium term metal loads. Likewise, molluscan shell and tissues are also good indicator of metal pollution as they are sessile and sedentary and they reflect the heavy metal concentration of that particular area[7]. As expected, heavy metal concentrations in sediments greatly exceeded those in the surrounding water. In aquatic environments, heavy metals discharged from industrial or sewage effluents or from atmospheric deposition may be rapidly removed from the water column and transported to the bottom sediments[8]. The availability of metals in sediment provides an opportunity for aquatic animals to biomagnify these metals and later remobilized them through the food chain. In the present study, *N. crepidularia* from mangroves at Vellar estuary was studied in shell and body tissue for the level of accumulation of various metals apart from comparing the concentration with that of the sediment.

In the present study, the level of Cr in sediment was ranging between 7.10 μg/g and 31.44 μg/g. The same level of Cr was reported in the sediment of other areas also *viz*., 13.3 to 54.3 μg/g in Brisbane river, Australia[9], 7.6 to 42.5 μg/g in Mazaetlan harbor, Mexico[10], 37.4 to 43.4 μg/g in Guanasara bay, Brazil[11] and 40.0–42.5 μg/g in Deep Bay, Hong Kong[12]. In the present study Cu concentration ranged from 11.28 μg/g to 37.24 μg/g in the sediment of Vellar estuary. At the same time differential levels of Cu was reported by different researchers in sediments from different areas. Some of the worth mentioning studies are as follows: 3.1–30.2 μg/g in Brisbane river, Australia[9]; 41.9–49.8 μg/g in Mai Po, Hong Kong[13]; 80.0 μg/g in Deep Bay, Hong Kong[12]; 7.7–90.9 μg/g in Mazaatlan harbor, Mexico[10]; 79.6–91.7 μg/g in Guanabara Bay, Brazil[11] and (7.06±6.03) μg/g in *S. Buloh* & (32.00±14.33) μg/g in *S. Khatib* Bongsu, Singapore[13].

In the present study Zn concentration (36.14 to 56.14 μg/g) was lower when comparing the following studies: 240.0 μg/g at Deep Bay, Hong Kong[12], 447.5–505.1 μg/g at Guanabara Bay, Brazil[11] and (51.24±39.97) μg/g at *S. Buloh* & (120.23 ±13.90) μg/g at *S. Khatib* Bongsu, Singapore[13]. The higher content of zinc and cu is largely due to the anthropogenic input and it may be said that it is being immobilized through co-precipitation with carbonates and thus cause a lesser hazard to the ecosystem.

Bioaccumulation of iron in Vellar estuarine sediment was studied by Rajan *et al*[14]. The highest concentration of 10 960 μg/g was recorded during summer. Whereas the present study recorded 7 424 μg/g during summer which is lower than that of the earlier studies. Likewise as reported 285.0 μg/g in premonsoon and 355.0 μg/g in monsoon which is lower than that of the present study (11 444.0 μg/g (December 2007–monsoon) and 10 848 μg/g (May 2007–premonsoon season)[15]. The higher values of iron in the present study may be due to the nature of the substratum *i.e.* the clayey substratum of mangrove at Vellar estuary has accumulated more metals than the sandy substratum as reported[16].

The metal concentration in the sediment was in the order of Fe>Zn>Cu in Kodiayakkarai coastal environment. In the present investigation the order of accumulation was Fe> Al> Mg> Mn> Cd> Cu> Cr> Zn> Ni> Pb[17]. The variation in the accumulation of metals in sediment was due to their geographical location[18]. Moreover the difference in the pattern of accumulation could have been influenced by the discharge of varying amounts of sewage and municipal wastes. Moderate enrichment was recorded in the surface layers (0–5 cm) for iron (1 607–1 905 μg/g) in Uppanar river, Cuddalore coast[19]. The present study indicates higher sediment concentration [(10 028.10±557.00) μg/g] than the earlier study. The sediment concentration of cadmium ranged from 0.68 ppm in station 1 and 1.68 ppm in station 2[20]. The higher concentration may be due to the mobilization through organic matter[21] and through clay size fractionation of the sediment and also by the composition of the mineral assemblage with enhance favourable adsorption of metals from water and their incorporation in the sediment[22].

Several studies on Pb in sediments have been conducted in several regions. High levels of Pb, 26–630 μg/g, were detected in North Sea[23] and 0.5–60 μg/g were found in Gulf of Finland[24]. In the present study Pb concentration was found very low (0 to 2.42 μg/g). The metal concentration in sediment is dependent on the nature, adsorption and retaining capacity of the substratum[22]. Overall, heavy metal levels in Vellar estuarine mangrove sediments are lower compared to reported levels for mangroves in Australia, Mexico, Brazil, Hong Kong and Singapore. These variations are considered due grain size, organic carbon content, composition, composition of sediments[25], geological weathering and the presence of well developed mangrove forests which create physicochemical conditions suitable for the accumulation of the measured metals in the mangrove muds[26]. Anthropogenic inputs such as discarded automobiles, transformers, batteries, tires and crude oil spill, atmospheric fallout as well as water-waters disposal.

In the present study the results of ANOVA showed a positive significant relationship between sediment and tissues in their metal concentration. From this, it would be inferred that the available metal concentration in the ambient medium might have influenced the uptake of metal as observed[27].

The use of molluscan shells as sentinels for metal pollution monitoring in marine waters has several advantages over that of the soft tissues. The shells are easy to store and handle and appear to be sensitive to environmental heavy metals over the long term. Since shell growth occurs incrementally they can provide a signal over a discrete time period, unlike the tissues which are strong accumulator of metals and integrate the chemical contamination signal over the life of the organism. Refinement of techniques for determining element using bivalves is important if global monitoring is to become a reality[28]. Pb levels in the nacre of *Mytilus edulis* collected from near a lead Smeiter off the coast of Quebec and Dalhousie, Canada, were only a tenth of the levels in the tissues[29]. Cd, Cu, Mn and Zn were found to be most concentrated in the prismatic calcite layer of the shell of the oyster *Crassostrea virginica*[30].

The composition of the molluscan shell is strongly related to the chemical mineralogy which includes metals accumulated from the environment, and therefore metal concentrations in the shells follow the metal concentrations in their environments[31]. Shells also have some practical advantages over the use of soft tissue as they can reveal less variability, integrate metal concentrations over the life of the organisms, able to give an idea on the metal levels in the past and offer considerable advantages in easy preservation and storage .The shell size and weight along with the age are important factors which determine bioaccumulation[32].

The concentration of iron in the molluscan shells depends to some extent, on the iron concentration in food supply and on the organism's growth rate and they recorded 100 to 309 ppm of iron in *Anadara diluvi* from Port Said Mediterranean Sea[33]. The concentration (ppm) of Zn, Cu, Pb and Cd in bivalve shells were reported as 2.99-3.22, 2.30-2.59, 8.77-9.65, 1.93-2.14 & 20-174, 5.00-89.01, 30.93, 1.8-3.9 from Timsah Lake Suez & Red sea coast respectively[34,35]. The mean concentrations (μg/g dry weight) of Cd, Cu, Fe, Ni, Pb and Zn in the shells of *Nerita lineata* (*N. lineate*) were 3.15, 5.59, 49.78, 24.18, 48.86 and 7.86, respectively from Peninsular, Malaysia[36]. Al, Cu, Zn, Fe and Mn levels were estimated as (80.86±100.48), (3.53±3.29), (24.00±14.63), (211.20±273.71) and (461.52±252.67) μg/g, respectively in shell of *Unio pictorium* mancus from Italy[37]. Iron concentration in *Tridacna maxima* shells varied from 216.4 ppm at Abu-Ghusun Lagoon to 1 286.4 ppm at Safaga Harbor[38]. In the present study the concentration of Fe, Cd, Ni, Pb was found to be 88.54-176.46, 0.34-0.76, 2.44-14.58, 0.00-0.22 μg/g, respectively. The values were relatively lower than that of the above studies except the concentration of Fe. The higher concentrations of Cd, Ni and Pb found in the shell could probably be due to the fact that the crystalline structures of the shell matrix have a higher capacity for incorporation of Cd, Ni and Pb[39].

The Mn content in giant clam shells that ranged from 29.9 ppm at Abu-Ghusun Lagoon to 65.2 ppm at Safaga Harbor[38]; but in the present study the mean concentration of Mn was (75.89±27.81) μg/g which is lower than the previous studies. Highest concentrations of Cd, Ni and Pb in the shells, followed by operculum and the lowest was found in the soft tissues from *N. lineata* from West intertidal area of Peninsular Malaysia[36]. The highest concentration of Zn in the shell of *Tridacna maxima* in Hurghada Harbor (32.6 ppm), while the lowest in Quseir Harbor (11.4 ppm). The Cu content in the giant clam shells ranged from 12.4 ppm at Hurghada Harbor to 81.4 ppm at Abu- Ghusun Lagoon[38] which is higher than that of the present study [5.74-12.62 μg/g (Zn) and 3.62-8.42 μg/g (Cu)]. Several previous studies also showed the fact that some trace metals are incorporated into the shells of molluscs and barnacles through substitution of the calcium ions in the crystalline phase of the shells or are associated with the organic matrix of the shells.

In an environment affected by contaminant, both oyster tissue and shells have an opportunity to adsorb heavy metals from suspended particles. These suspended particles may be either as contaminated sediment or sediment controlled by the behaviour of heavy metals themselves and the physical and chemical conditions of the environment[20]. When Pb and Cd increased in the ambient seawater, they are incorporated into the shells at concentrations higher than normal[40]. The results obtained in the present study clearly indicate the various levels of accumulation of trace metals in the body of *N. crepidularia*. Among the ten metals (Fe, Al, Mg, Mn, Cd, Cu, Cr, Zn, Ni and Pb) analysed, the iron concentration showed higher values than the other metals, the degree of accumulation of trace metals in the animal tissues was as follows:

Fe> Al> Mg> Mn> Cd> Cu> Cr> Zn> Ni> Pb

The higher concentrations of metals (Fe, Al, Mg, Mn, Cd, Cu, Cr, Zn, Ni and Pb) in the monsoon season could be due to the heavy inflow of freshwater, which brings lot of effluents from irrigation channels and municipal drainage. In addition to that, the sea water receives the wastes from the iron ore mining operations at the Servarayan hills, where the river Vellar originates. This may be also one of the factors responsible for high iron concentration in seawater. The lower concentrations in summer may be due to decrease in land drainage caused by the absence of rainfall. This fact was in agreement with the findings[41] and similar pattern in Vellar estuary[27]. This low concentration might also be related to the higher uptake by planktonic organisms. The environmental parameters like salinity, temperature, dissolved oxygen and pH also have some effect on the accumulation of trace metals. Among the above parameters, the salinity played a major role[16]. In the present investigation, high values of all 10 metals were recorded in low saline (monsoon season) and low values during high saline seasons (summer season). The statistical analysis also proved this *i.e.*, significant correlation was observed between the metal concentration in tissue, shell, sediment and seasons.

The results of the present study are comparable to those of the previously reported studies in Neritid gastropods from several geographical regions. In the present study the Cd concentration was found fluctuating from 0.48 to 2.44 μg/g in the body tissues of *N. crepidularia*. In the earlier, Cd concentration was found to be (6.88±1.02) μg/g in *Nerita albicilla* from Taiwan coast[42], 1.78-2.87 μg/g in *Nerita albicilla* from marine environment, Hong Kong[43] and 2.83 μg/g in *N. lineata* from Peninsular, Malaysia[36]. In the present study, Zinc concentration (15.26-22.5 μg/g) was lower when comparing the results for 63.90-81.75 μg/g in Sunderban mangroves[44] in *Erita articulata*; 150-130 μg/g in Taiwan coastal waters[42] in *Nerita albicilla* and 31-680 μg/g in S. Khatib, Singapore[13] in *N. lineata*. In the present study, the concentration of Ni was ranged between 4.14-20.22 μg/g from the body tissues of *N. crepidularia* it was higher accumulation when compare the Ni (2.93-8.34 μg/g) accumulation of *N. lineata* from peninsular Malaysia[36].

In the present study Cu concentration was ranging from 15.16 to 38.56 μg/g from the tissues of *N. crepidularia* that is higher when compared to that of in the Cu *Nerita articulata* (19.74–21.30 μg/g) & *N. lineata* (11.46–25.24 μg/g)[36,44].

The adsorption and consequent settlement by the particulate matter is another reason for these lower concentrations of metals[45]. The process like flocculation due to the salinity increase is also another reason for the low metal concentrations in summer months[46].Further the body tissues reflect the level of heavy metals in water as well as sediment, since they are very good accumulators of them and thus the molluscan animals are acting as very good indicators of heavy metal pollution in any environment[47].

Iron plays an important role as an essential element in all living systems from invertebrates to humans but increasing of iron in the marine environment may reflect the bioaccumulation in the marine organisms such as, bivalves, gastropods, coral reefs and fish. Total iron (Fe) in oceans is estimated to be $4\ 110\times10^6$ metric tones, contributed by geological processes and man–induced activities such as mining[28]. It was also inferred that Fe concentrations in coastal waters is a function of freshwater input and is greatly influenced by riverine waters[48] and is generally abundant in any environment and has several properties similar to those of manganese; for example, its partitioning between water and sediments is largely controlled by the oxygen concentration in the water. Further the highest levels of Fe obtained during an extended biomonitoring study in the Shannon Estuary[49] may indicate a once–off anthropogenic input or release from bedrock such as dredging. However, since the elevated levels were of Fe only, an anthropogenic source seems more likely.

The concentration of iron observed in the tissues of *Anadara rhombea* ranged from 1 310 μg/g to 3 680 μg/g at marine zone and 1 630 μg/g–3 700 μg/g at tidal zone in Vellar estuary[50]. Iron concentration in male and female *Donax cuneatus* ranged between 281 & 2 013 μg/g and 261 & 2 113μg/g from respectively in Porto Novo coast[1]. The range of Fe concentration was 154 to 558 μg/g dw (mean value) in *Megapitaria aqualida* from mine–impacted sites of Mexico[51]. A significant variation in the Fe concentration during their study period (July 1998 to May 1999) in Buena by recording the highest value in October 1998 (328 μg/g) and also in Ponta do Retiro where they recorded the highest Fe concentration of 417 μg/g in the same month[52]. The mean concentration of iron as 566.63 μg/g in the soft tissues of *N. lineata* from peninsular Malaysia[36]. In the present study (*N. crepidularia*), the mean concentration of Fe was found to be (1 615.00±464.24) μg/g. This value is lower when compared to the above mentioned studies. The trace metal of majority of molluscs is related to their size. An increase in metabolic rates in younger individuals may affect metal uptake and elimination differentially[53].

Zn plays an important role in the composition of approximately 90 enzymes in animal metabolism[54]. In the present study, zinc concentration showed the maximum during monsoon and minimum during summer season. The higher concentration of zinc during monsoon season is mainly due to the influence of sewage outfalls and land drainage and also due to the entrance of chemicals from the agricultural lands without and specific industrial influence. The usage of zinc block in the fishing vessels would have resulted in enhanced zinc concentration in coastal waters[55]. In Porto novo also hundreds of vessels have been used for fishing, this may also be another reason for the higher concentration in coastal waters. In the present study, the concentration of zinc in tissues ranged from 15.26 to 22.50 μg/g in the mangroves at Vellar estuary. Whereas the zinc concentration was reported as 950 μg/g–2 760 μg/g at marine zone and from 1 290 μg/g–2 820 μg/g at tidal zone of Vellar estuary in *Anadara rhombea*[50]; 589 to 2 098 μg/g and 487 to 198 μg/g in male and female *Donax cuneatus* in Parangipettai coast[1]; (2.20±0.02) ppm in body tissue of *Cymbium melo* in Cuddalore[56]; 92.75 μg/g in *N. lineata* in Peninsular Malaysia[36]; (10.37±4.60) μg/g in *Patella piperata* in Canary Island, Spain[57]. Comparatively, in the present study, Zn concentration (15.26 to 22.50 μg/g) is lower than that of all the previous studies from Vellar estuary and other areas. In the present study, the zinc concentration also showed its peak during monsoon and minimum during summer. The higher concentration of zinc during monsoon was mainly due to the influence of sewage out falls and land drainage and also due to the entrance of chemicals from the agricultural lands without and specific industrial influence. Nevertheless, Zn and Cu accumulation rates differed greatly between mollusc species/tissues and metals[58]. Metal storage may differ according to the species. Further copper and zinc values are low when compared with that of the previous studies cited above which may be due to low concentrations of these metals in the seawater. The concentration of Zn in *Ostreola equestris* differed significantly ($P<0.05$) between Buena (800 μg/g) and the other two beaches (1 200 μg/g in Ponta do Retiro and 1 400 μg/g in Barra do Furado)[52]. In the present study, Zn concentration of tissues showed negatively correlated ($r=-0.485$) between the salinity. Salinity was thus found to have a profound effect on the accumulation characteristics of metals in the tissues of *N. crepidularia*. This was evidenced statistically with a negative correlation. It was supported in the earlier[1] studied from *Donax cuneatus* in Parangipettai coasts.

Cadmium and some of its compounds are considered carcinogenic and may cause damage to all types of body cells. In the present study, the mean Cd concentration was found to be (1.51±0.98) μg/g. The mean concentration of Cdas1.3 μg/g in *Cladophora glomerata*, 3.9 μg/g in *Ostrea lutaria* and 0.17 μg/g in *Haliotis* from New Zealand waters[59]; Cd concentration ranged from 1.5 to 11.1 μg/g dw from *Megapitaria squalida*[51]. The mean concentration (μg/g dry weight) of Cd was found to be 1.03 in the soft tissues of *N. lineata*[36]. The Cd concentration as 2.65 to 13.50 μg/g at Uppanar, Kaduviar & 10.5 μg/g in Vellar and 2.5 & 12.5 μg/g at Kaduviar from *Crassostrea madrasensis*[47]. In the present study the Cd values are very low when compared to that of the previous studies. The lower level of Cd in *N. crepidularia* may be due to its small size as also reported for *Donax rugosus*[60].

Copper is an essential and potentially toxic element. Ferreira *et al*(2005)[52] recorded a significant spatial difference

($P<0.05$) for Cu in *Ostreola equestris* in Ponta do Retiro (86 μg/g) when compared to Buena (50 μg/g) and Barra do Furado (39 μg/g). The copper concentration found fluctuated from 5.4 to 18.7 μg/g dw in *Megapitaria squalida*[51]; (1.36±0.01) ppm in the body tissues of *Cymbium melo*[56]; 40.0 μg/g in *Cladophora glomerata* and 11.0 μg/g in *Ostrea lutaria*[59]; (2.05±0.91) μg/g in *Patella piperata*[57] and 2.65 μg/g in the soft tissues of *N. lineata*[36]. In the present study the mean concentration of Cu was found to be (25.9±10.3) μg/g which is comparatively lower than that of the earlier studies. Others have found a trend of decreasing tissue metal concentrations with increased mass though this does not always occur[61].

Source of manganese might be due to the land drainage and from effluents through irrigation channels and municipal wastes. These drainage waters which contain high heavy metal concentrations enter the estuaries and mix up with the seawater. In the present study the Mn concentration was ranging from 191.96 to 242.42 μg/g. The concentration of manganese in *Anadara rhombea* and found that ranged between 200 μg/g & 580 μg/g at marine zone and 220 μg/g and 600 μg/g at tidal zone[50].

Lead is number two (after arsenic) on the top 20 list of the most poisoning heavy metals. Its target organs are the bones, brain, blood, kidneys, reproductive and cardiovascular systems and thyroid gland[62]. Lead is leader member of the toxic metals in the marine environment. It is mainly attributed to gasoline fuels containing high ratio of lead tetrachloride. In the present study, Pb concentration (tissues) was reported as 0 to 0.44 μg/g which is comparable with 0.04 μg/g and 0.01 μg/g in *Ostrea equestris* from Barra do Furado and Buena & Ponta do Retiro[52]; 92.72 μg/g in the soft tissues of *N. lineata* [36] from Malaysia.In the present study the Pb mean values was found to be (1.6±2.2) μg/g it was very low when compare the previous studies.

Nickel is a carcinogenic metal and overexposure to it can cause decreased body weight, heart and liver damage and skin irritation in Jordon[62]. In the present study the Ni concentration ranged between 4.14 μg/g and 20.22 μg/g.

Magnesium is a major naturally occurring element and considered as major constituent[63]. The Mg concentration was (151.3±1.9) ppm in the body tissues of *Cymbium melo* from Cuddlore coast[56], but in the present study the mean Mg concentration was found to be (1 913.00±222.03) μg/g. This value was relatively higher in the above studies. This might be due to the sediment nature *i.e.*, the clayey substratum has the ability to accumulate more metals than sandy substratum[64].

Several health problems are related to chromium consumption such as chronic ulceration and perforation of the nasal septum and allergic skin reactions[65]. In the present study the Cr concentration was ranging between 1.5 μg/g and 2.9 μg/g. The Cr concentration varied significantly between Barra do Fura do (0.7 μg/g) and remaining beaches (average of 0.3 μg/g in Ponta do Retiro and Buena)[52], which is much lower than that of the present study on *N. crepidularia*.

Aluminum is regarded as the third most abundant element in the Earth's crust after oxygen and silicon[66] and it is the most commonly used normalizing element due to its abundance, natural origin in most cases and a metal of sediment constitution[67]. In the earlier[37] studied the mean Al concentration was ranged between 252.98±183.77 from *Unio pictorum mancus* from Northern Italian lakes, but in the present study Al concentration was found to be much higher (946.6 to 1973.2 μg/g). 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[68]. The concentrations of the heavy metals analyzed exhibited variations in sediments, tissue and shell of the study animal from the 2 stations. Zn and Cu concentration were below the alarming level, whereas Mg (274.0±12.0) content was higher in shell and tissue[69].

The present exploration was made for a phase of one year (January to December 2007) on *N. crepidularia* in the mangroves at Vellar estuary to know the heavy metal concentration in mangrove sediment and shell & soft tissues and to find out the possibility of using *N. crepidularia* as an indicator organism for heavy metal pollution.

The metal concentration in sediment, tissues and shell revealed that maximum concentration was recorded during monsoon and minimum concentrations during summer season. The higher concentration observed during monsoon could be due to the heavy inflow of fresh water which brings lot of effluents from municipal, domestic and agricultural wastes. Iron ore mining operation at Servarayan hills, where Vellar river originates may be one of the reasons for high iron content in Vellar estuary. The scrap metals from the boat metals & fishing vessels and paint residues (antifouling paints) from local boat jetty may be the source of zinc and copper in the sediment and finally in the shell & tissues of *N. crepidularia* in the present study. The lower concentration of heavy metals in all the three components (sediment, shell and tissue) studied in summer may be due to the decrease in inflow of freshwater into the estuary. Further the adsorption and consequent sedimentation and flocculation due to salinity increase are some of the reasons for low metal concentration in summer. When compared to the other two seasons, the premonsoon showed higher values than postmonsoon. The agricultural activities during the premonsoon season may be another reason for high metal concentration in that season.

In *N. crepidularia* it may be used as an indicator species of metal pollution and therefore can be used as sentinel organism to monitor pollution in the mangroves environment due to the following reasons. Since *N. crepidularia* accumulates high concentrations of heavy metals, in general, than the ambient environment and Cu & Mn, in particular, it may be used as a biomonitor of certain heavy metals in mangrove environment.

Conflict of interest statement

We declare that we have no conflict of interest.

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