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Research Article

Risk assessment for the daily intake of metals fromthe ingestion of mussels (*Meretrix casta* and *Meretrix meretrix*) from Vellar and Coleroon estuaries and Pichavaram mangroves, Southeast coast of India

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Abstract

The level of selected metals such as Cd, Cr, Cu, Ni, Pb and Zn were assessed from two mussels namely, *M. casta* and *M. meretrix* are common bivalves from Vellar and Coleroon estuary and Pichavaram mangroves, Southeast coast of India. The concentration of metals in *M. casta* and *M. meretrix* was ranged between 0.96 - 42.32 and 1.28 - 100.8 (µg g^{-1}) respectively. Target Hazard Quotient analysis of the metals revealed that the level was below to the reference oral dose except Cr and Pb. Among the metals, Cr was highly accumulated in *M. casta* and *M. meretrix*, whereas Pb highly accumulated in *M. meretrix* at all the stations. Since Cr and Pb are heavy toxic chemicals in human and aquatic environment, more attention is very much essential for further remediation and control the source of the chemical to mix up with environment.

Keywords: Metals, Estuary, Lead, Chromium, *Meretrix casta* and *Meretrix meretrix.*

INTRODUCTION

The coastal zone can be considered as a geographic space of interaction between terrestrial and aquatic ecosystems that is of great importance for survival of large variety of plants, animals and aquatic organisms[1,2].The industrialization and agricultural activities contributed to an increasing of discharge of chemical pollutants into the ecosystem in many countries, which lead to increase in metals concentrations in the natural waters that causing damage of fresh and marine habitats [3,4].The accumulation pattern of heavy metals in fish and other aquatic organisms depends on their uptake and elimination rates [5]. Under certain environmental conditions, heavy metals might accumulate up to toxic concentrations and cause ecological damage[6,7]. Metals such as iron, copper, zinc and manganese, are essential metals since they play important roles in biological systems [9], whereas non-essential metals such as Pb and Cd are usually potent toxins and their bioaccumulation leads to intoxication, decreased fertility, cellular and tissue damage, cell death and dysfunction of a variety of organs [10,11].

A recommendable diet should be able of providing sufficient nutrients containing low levels of chemical contaminants. Among these, potentially toxic elements such as cadmium (Cd), and lead (Pb) are widely distributed into the environment [12, 13, 14].In recent years, concern on dietary intake of organic and inorganic environmental pollutants has notably increased [15,16,17,18, 19,20]. Biomonitors, especially shellfish have been extensively used to examine trace metal contamination in coastal systems and to reveal the bioavailability of contaminants, as exemplified by the global ‘‘mussel watch’’ program [21,22,23]. The wide use of shellfish reflects not only the high capacity of these organisms to bioaccumulation of organic and inorganic contaminants and their widespread distribution, but also their importance, because shellfish represent an important source of protein for coastal communities. It has been estimated, for instance, that over 90% of human health exposure to several contaminants occurs through diet primarily seafood and meat [24,25].

Segments of the human population with increased exposure risk include consumers of commercially harvested shellfish, recreational and subsistence fishers and subsistence shellfish consumers, not to mention the children whose diet is based on harvested shellfish. Biomonitors which are taxonomically closely related may preferentially accumulate different metals as well as different species of the same metal in the aquatic environment [26]. Data interpretation, however, should give similar information for different species, if it is compared to what is typical for each species [27]. In addition, as metal concentrations are expressed in µg per g body weight and biomass of a biomonitoring can influence its final metal concentration due to tissue [28]. In view of that, the present study was aimed to obtain quantitative information on heavy metal concentrations in bivalves and to compare total metal content at different stations along southeast coast of India. In addition, risk assessment due to heavy metals in bivalves was also been investigated.

MATERIALS AND METHODS

Study area

Vellar and Coleroon estuarine ecosystem

The study has been conducted in Vellar (Lat. 11° 48' N; Long. 79° 76' E) and Coleroon estuaries (Lat. 11°25'N; Long. 79°48'E) selecting salinity gradient zones. Estuaries serve as both pollution sources for the coastal sea and marginal filter to the polluted runoff from the river drainage basin [29.] (Fig.1).

Pichavaram mangrove ecosystem

The Pichavaram mangrove ecosystem(Lat. 11° 26' N; Long. 79° 77' E) in Tamil Nadu coast is very important in terms of ecological point of view and it is formed out of the network of channels and adjoining creeks through the estuaries of Vellar and Coleroon. This ecosystem fringed with variety of mangrove plants, mud flat shrubs which serve as significant breeding ground for number of marine and brackish water organisms (Fig.1).

Sampling methods

The bivalves were collected in April 2013 from Vellar and Coleroon estuary and Pichavaram mangrove ecosystem. About 20 individuals of each species at each location were collected to prepare a pooled sample to reduce individual variations in heavy metal concentrations. Further, the edible part of the meat from samples were carefully removed by shelling the bivalves with a plastic knife, dried and ground well to obtain fine powder before analysis.

Heavy Metals Analysis

The analysis of metals was done by following the method proposed by Walting [30]. Briefly, two sets of one gram of the dried bivalves such as spiking with known concentration metals and without spiking were taken in a beaker and 20 ml of conc. HNO_3 was added and kept for 24 hrs. Then the sample was evaporated to dry on a hot plate at 120 °C, subsequently 20 ml of 4:1 ratio of nitric acid and perchloric acid mixture was added and the residue was allowed to dry completely and cooled. After that, 10 ml of 10% nitric acid was added and transferred to a 20 ml polytop vial and allowed this mixture to settle the residue for 2 hrs. The supernatant was filtered by using cotton and the filtrate was used for metal analysis by ICP-OES (Perkin Elmer, Optima 2100DV) and quantified against known standards.

RESULT AND DISCUSSION

Heavy metals such as Cu, Zn, Fe and Zn are essential for human; however, these metals may be harmful to human health if their concentration in foods exceeds the limit. Therefore, many national / international organizations worldwide (including WHO, FAO, USEPA, FSANZ, ANZFA, IEFS) set tolerable dietary intake (TDI) limits for the metals depending on body weight and age of the consumers. In the present study, the concentration of heavy metals (Cd, Cu, Cr, Ni, Pb and Zn) in bivalves is presented in Table.1 and 2. Among the metals analyzed, lead (Pb) and cadmium (Cd) were classified as chemical hazards [31, 32, 33].

Traces of cadmium were detected in bivalves, the highest concentration was found in *M. meretrix* (2.52 µg g^{-1}) of Coleroon estuary followed by low level detected in *M. casta* (0.96 µg g^{-1}) from Vellar estuary. The threshold level for acute cadmium toxicity would appear to be a total ingestion of 3–15 mg. Severe toxic symptoms are reported to occur with ingestions of 10–326 mg. Fatal ingestions of Cd, producing shock and acute renal failure, occur by ingestions exceeding 350 mg [34]. The concentration of Cd in all collected bivalve species was lower than that of reported by Hamed and Emara [35] and de Mora et al. [36]. However, the reported Cd concentrations in this study are comparable with that of detected level by Sankar et al.[37].

Copper is an essential part of several enzymes and it is necessary for the synthesis of hemoglobin [38]. In the present study, concentrations of Cu were higher (57.68 µg g^{-1} dry weight) in *M. meretrix* from Vellar estuary and lowest were reported (36.92 µg g^{-1} dry weight) in *M. casta* collected from Pichavaram mangroves. However, concentrations of Cu in all collected samples were much lower than that recorded by Usero et al.[2] in southern Spain (9.2–90 µg g^{-1} dry weight) and De Mora et al.[36] in Gulf of Oman (60.9– 210 µg g^{-1} dry weight).The concentrations of Cu in both the bivalves from Vellar and Coleroon estuary were above the toxic limit[31] (30 µg g^{-1}) which is due to discharges mixed with wastewater of industry, agriculture and sewage.

Chromium is among a group of hazardous metals notified by the USFDA[39]. It has been estimated that the average daily human requires of chromium is 1µg[40] and deficiency of chromium results in impaired growth and disturbances in glucose, lipid, and protein metabolism[41]. In the present study, levels of Cr in both bivalves of all the stations were fluctuated between 38.12 and 100.8 µg g^{-1} dry weight and highest concentration of Cr (100.8 µg g^{-1}) was detected in *M. meretrix* at Coleroon estuary. The concentrations of Cr reported from the present study were comparable with that of reported by De Mora et al.[36] and Cr level reported was relatively higher than the permissible level (13 µg g^{-1}) recommended by FDA[33].

The essential function of nickel (Ni), for example, in enzymes such as urease and hydrogenase, in most plants and some microorganisms is well-known. On the other hand, nickel isa carcinogenic metal and over exposure to it can cause decreased body weight, heart and liver damage and skin irritation[42].The Ni concentration reported in the present study exhibited wide variation where the higher concentration was detected at 21.52 to 50.86 µg g^{-1} dry weight, while the lower in M. *casta* of Pichavaram mangrove (21.52 µg g^{-1} dry weight) and higher (50.86 µg g^{-1} dry weight) in *M. meretrix* of vellar estuary. The results of the present study reveals that the nickel concentrations are relatively lower than that of reported by Usero et al.[2] in southern Spain (66.0 - 92.0 µg g^{-1} dry weight) but higher than that of Siva perumal et al.[43] (0 - 0.89 µg g^{-1} dry weight).

Lead is the second element (after arsenic) on the top 20 list of the most poisoning heavy metals and its target organs are bones, brain, blood, kidneys, reproductive and cardiovascular systems, and thyroid gland [44] . In humans, the main target organ is the central nervous system, particularly young children. An inversely proportional relationship has been demonstrated between blood lead concentrations and intelligence quotient scores. In adults, lead affects the kidneys (increased prevalence of chronic kidney disease) and the cardiovascular system (high systolic blood pressure). The results of the present study evidenced that the accumulation of lead in both mussels fluctuated between 20.36 µg g^{-1} dry weight and 43.48 µg g^{-1} dry weight in *M. meretrix* at coleroon estuary and pichavaram mangroves respectively. The concentration of Pb reported was comparable (0.48–1.18, 0.12–1.09, 0.01–1.18, 0.44–0.67 µg g^{-1} dry weight) with those reported by Favretto et al. [45]; Majori et al.[46]; Widdows et al.[47]; Conti and Cecchetti[50] respectively and lower than that of reported by Usero et al.[2] in southern Spain (0.74–1.92 µg g^{-1} dry weight).

High levels of zinc cause pancreatitis, anemia, muscle pain, and acute renal failure [48]. The level of Zn reported in both mussels were ranged between 34.92 and 78.8 µg g^{-1} dry weight and lowest in *M. casta* at Vellar estuary and highest in *M. meretrix* at Coleroon estuary. The concentration of Zn reported in this study was comparable with those reported by Hamed and Emara[33] (2006) (56.5–191.4 µg g^{-1} dry weight) in Red Sea and Campanella et al.[49] (5–31 µg g^{-1} dry weight) and relatively lower than that of reported by Widdows et al.[47] (82–185 µg g^{-1} dry weight) and Conti and Cecchetti[50] (98–152 µg g^{1} dry weight).

Risk-based consumption limits

Risk factors were calculated according to the guideline of the United State Environmental Protection Agency (US-EPA). Additionally, based on the US-EPA Guidance[51, 54] ingestion doses are equal to the adsorbed contaminant dose and that cooking has no effect on the contaminants [52]. Target hazard quotient (THQ) indicated the ratio between exposure and the reference dose, and calculations were made using the standard assumption for an integrate US-EPA risk analysis. The agency's preferred toxicity values for evaluating the RfD_o for non-carcinogenic effects resulting from exposure in this risk assessment was provided in the EPA's Integrated Risk Information System (IRIS) and Health Effects Assessment (HEAST) and Health Canada[51,53,54] were reported in Table.3 and 4.

Estimated Daily Intake (EDI)

$$= IR \times C \times EF \times ED / BW \times AT$$

EDI = Estimated Daily Intake (mg kg^{-1} day^{-1})
IR = Ingestion rate (kg $day^{-1)}$;
(International adults: 8.01g)
C_{tis} = Con.c (mg kg^{-1} wet weight) of the investigated chemical pollutants in bivalve tissues
EF = Exposure frequency (days $year^{-1}$);
350 days $year^{-1}$for Non carcinogenic effects
ED = Exposure duration (Years);
30 year for Non carcinogenic effects
BW = Body weight (kg); 70kg
AT = Average weight of exposure days;
365 × 30 for Non-carcinogenic effects

As the (RfDo) is a threshold dose or intake, which is conservatively chosen so that if the estimated intake is less than the reference oral dose (HQ < 0.1), there is almost no possibility of an adverse health effect. However if the intake exceeds the reference dose (HQ > 0.1), this does not indicate an adverse health effect is expected, only that a conservative reference oral dose (RfD_o) is exceeded that was mentioned in Table.5 and 6.

$$\mathbf{THQ = EDI / RfD_o}$$

THQ = Target Hazard Quotient
EDI = Estimated Daily Intake (mg kg^{-1} day^{-1})
RfD_o = Oral reference Dose for Non- carcinogenic Toxicity value (mg kg^{-1} day^{-1})

CONCLUSION

The THQ conducted on the metals assessed from bivalves such as *M. casta* and *M. meretrix* from Vellar and Coleroon estuary and Pichavaram mangroves of southeast coast of India was highlighted two major problems: (i) the first problem was related to the scarcity of information on metal concentrations in marine organisms of the selected study animals, and thus no integrated and updated database was available. (ii) The second problem was related to the effects and guidelines and on-site eco-toxicological data were not available meanwhile. In spite of these limitations, the adopted approach in the present study highlighted that no adverse ecological and human health effects are associated with the exposure to metals and thus, a further detailed investigation based on site specific studies would be greatly required in the region, there are no emission inventories of sources or release of inorganic pollutants in the environment, or any official data on stockpiles and disposal. In order to determine and control the sources of the observed contaminants of potential concern, intense localized sampling and analysis of effluents and runoff patterns are very much needed is the specific area.

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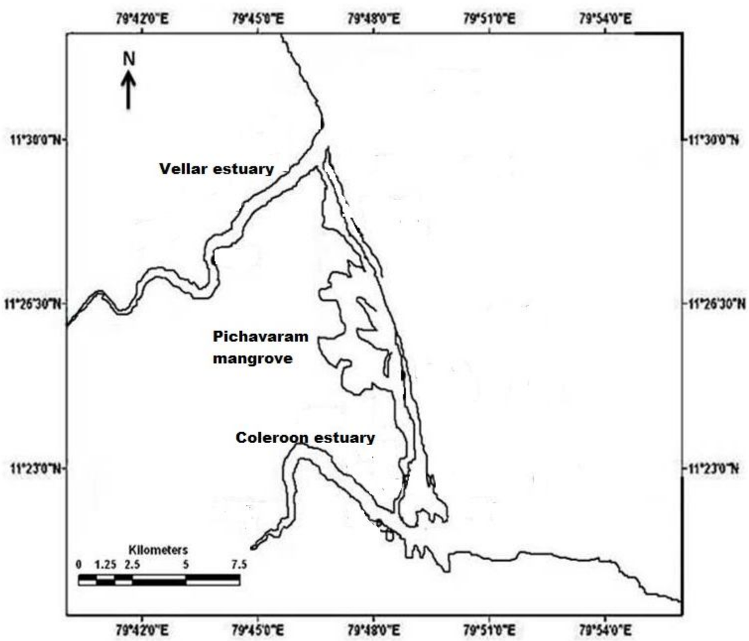

Fig 1: Map showing the sampling sites of Vellar estuary, Coleroon estuary and Pichavaram mangroves.

Table.1. Concentration (µg g^{-1}) of heavy metals in *M. casta*

	Meretrix casta					
Stations	**Cd**	**Cr**	**Cu**	**Ni**	**Pb**	**Zn**
Vellar estuary	0.96	41.64	41.64	23.28	20.44	34.92
Pichavaram mangrove	1.12	38.12	36.92	21.52	21.48	36.24
Coleroon estuary	1.16	42.32	38.28	24.08	20.36	36.32
Max	**1.16**	**42.32**	**41.64**	**24.08**	**21.48**	**36.32**
Min	**0.96**	**38.12**	**36.92**	**21.52**	**20.36**	**34.92**
Mean	**1.08**	**40.69**	**38.95**	**22.96**	**20.76**	**35.83**

Table.2. Concentration (µg g^{-1}) of heavy metals in *M. meretrix*

Stations	**Cd**	**Cr**	**Cu**	**Ni**	**Pb**	**Zn**
Vellar estuary	2.28	98.36	57.68	50.86	42	70.36
Pichavaram mangrove	1.28	57.2	48.04	34.88	43.48	60.84
Coleroon estuary	2.52	100.8	52.608	46.16	39.8	78.8
Max	**2.52**	**100.8**	**57.68**	**50.86**	**43.48**	**78.8**
Min	**1.28**	**57.2**	**48.04**	**34.88**	**39.8**	**60.84**
Mean	**2.03**	**85.45**	**52.78**	**43.97**	**41.76**	**70.00**

Table.3. Estimated daily intake (CDI mg kg^{-1} day^{-1}) of heavy metals in *M. casta*

Stations / **Metals**	**Cd**	**Cr**	**Cu**	**Ni**	**Pb**	**Zn**
Vellar estuary	0.000105	0.004569	0.004569	0.002554	0.002243	0.003832
Pichavaram mangrove	0.000123	0.004183	0.004051	0.002361	0.002357	0.003976
Coleroon estuary	0.000127	0.004644	0.0042	0.002642	0.002234	0.003985
Max	**0.000127**	**0.004644**	**0.004569**	**0.002642**	**0.002357**	**0.003985**
Min	**0.000105**	**0.004183**	**0.004051**	**0.002361**	**0.002234**	**0.003832**
Mean	**0.000119**	**0.004465**	**0.004273**	**0.002519**	**0.002278**	**0.003931**
RfD oral (mg kg^{-1} day^{-1})	**0.001**	**0.003**	**0.04**	**0.02**	**0.0036**	**0.3**

Table.4. Estimated daily intake (CDI mg kg^{-1} day^{-1}) of heavy metals in *M. mereterix*

Stations / **Metals**	**Cd**	**Cr**	**Cu**	**Ni**	**Pb**	**Zn**
Vellar estuary	0.00025	0.010793	0.006329	0.005581	0.004608	0.00772
Pichvaram mangrove	0.00014	0.006276	0.005271	0.003827	0.004771	0.006676
Coleroon estuary	0.000277	0.01106	0.005772	0.005065	0.004367	0.008646
Max	**0.000277**	**0.01106**	**0.006329**	**0.005581**	**0.004771**	**0.008646**
Min	**0.00014**	**0.006276**	**0.005271**	**0.003827**	**0.004367**	**0.006676**
Mean	**0.000222**	**0.009376**	**0.005791**	**0.004824**	**0.004582**	**0.007681**
RfD oral (mg kg^{-1} day^{-1})	**0.001**	**0.003**	**0.04**	**0.02**	**0.0036**	**0.02**

Table.5. Health risk assessment calculation for hazard quotient (HQ) of *M. casta*

Stations	Metals	Cd	Cr	Cu	Ni	Pb	Zn
Vellar estuary		0.11	1.52	0.11	0.13	0.62	0.01
Pichavaram mangrove		0.12	1.39	0.10	0.12	0.65	0.01
Coleroon estuary		0.13	1.55	0.11	0.13	0.62	0.01
Max		**0.13**	**1.55**	**0.11**	**0.13**	**0.65**	**0.01**
Min		**0.11**	**1.39**	**0.10**	**0.12**	**0.62**	**0.01**
Mean		**0.12**	**1.49**	**0.11**	**0.13**	**0.63**	**0.01**

Table.6. Health risk assessment calculation for hazard quotient (HQ) of *M. mereterix*

Stations	Metals	Cd	Cr	Cu	Ni	Pb	Zn
Vellar estuary		0.25	3.60	0.16	0.28	1.28	0.03
Pichavaram mangrove		0.14	2.09	0.13	0.19	1.33	0.02
Coleroon estuary		0.28	3.69	0.14	0.25	1.21	0.03
Max		**0.28**	**3.69**	**0.16**	**0.28**	**1.33**	**0.03**
Min		**0.14**	**2.09**	**0.13**	**0.19**	**1.21**	**0.02**
Mean		**0.22**	**3.13**	**0.14**	**0.24**	**1.27**	**0.03**

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