Bioaccumulation of trace elements in fin and shell fishes of Uppanar and Vellar Estuaries at Southeast coast of India

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Abstract
The study was performed to investigate the concentration of cadmium, cobalt, copper, iron, magnesium, manganese, nickel, lead, zinc, aluminium, chromium and boron in muscle, gill and liver of fin and shell fishes collected from two estuaries (Uppanar and Vellar) of Southeast coast of India. Tissue samples of fin and shell fishes had different level of accumulation: Cd = 0.10-0.72, Co = 0-0.18, Cu = 0.05-0.62, Fe = 1.42-9.85, Mg = 9.74-31.55, Mn = 0.09-11.12, Ni = 0.02-0.30, Pb = 0.14-0.50, Zn = 0.31-2.31, Al = 1.26-3.95, Cr = 0.25-2.91 and B = 0.17-0.51 µg g⁻¹ dry wt. Concentration of toxic metals such as Cd, Mn, Pb and Cr were above the permissible limits, in Uppanar estuary.

Keywords Fin fish; Shell fish; Metal accumulation; Uppanar estuary; Vellar estuary; Health hazard

Introduction
Marine organisms, in general, accumulate contaminants from the environment and therefore have been extensively used in marine pollution monitoring programmes. Recent years have witnessed significant attention being paid to the problems of environmental contamination by a wide variety of chemical pollutants including the heavy metals (Orecchio and Amorello 2010). In many countries, significant alterations in industrial development lead to an increased discharge of chemical effluents into the ecosystem, leading to damage of marine habitats. Metal contamination has been identified as a concern in coastal environment, due to discharges from industrial wastes, agricultural and urban sewage. There are essential and non-essential metals, but when their limits exceed in the body of organisms, they become more toxic. Metals like copper, zinc and iron are essential for fish metabolism, while others such as mercury, cadmium and lead have no known role in biological systems. Excessive pollution in coastal environment could lead to health hazards in man, through consumption of fish. Among animal species, fish are prone to the detrimental effects of these pollutants because of the continuous exposure (Olaifa et al., 2004).

Metals from natural and anthropogenic sources continuously enter the aquatic ecosystem where they pose serious threat because of their toxicity, long persistence, bioaccumulation and biomagnification in the food chain (Papagiannis et al., 2004). Fish being at the higher level of the food chain accumulate large quantities of metals and the accumulation depends upon the intake and elimination from the body (Karade et al., 2004). Pollution monitoring studies in the coastal regions showed that there are some elevated levels of metals. The consequence of heavy metal pollution can be hazardous to man and it often becomes mandatory to check chemical contaminants in foods from the aquatic environment to understand their hazardous levels. The Uppanar estuary located in south east coast of India may receive a huge amount of heavy metals due to the increasing industrial activities in State Industrial Promote Corporation of Tamilnadu region, like pharmaceutical, paper and metal processing plants, boat transports and domestic waste water disposal. Vellar estuarine area, another area located in southeast coast of India not only supports a flourishing tourism industry and a few aquacultural farms and in the area the Cr and Zn concentrations were found to be the highest; followed by Pb<Cd<Co being the lowest in the five species of fish tissue (Lakshmanan et al., 2009). Hence, the present study was taken up to find out the metal
concentrations in fin and shell fishes of commercial importance. Eventhough, there is much more industrial activity near Uppanar estuary than the Vellar estuary, both areas were selected to compare and determine the levels of trace metals (Cd, Co, Cu, Fe, Mg, Mn, Ni, Pb, Zn, Al, Cr and B) in the body organs of fin and shell fishes.

1 Materials and Methods

*Mugil cephalus*, *Penaeus indicus*, *Crassostrea madrasensis* and *Meretrix meretrix* were collected from random catches in the Uppanar (lat. 11°41’N; long. 79°45’E) and Vellar estuaries (lat. 11°29’N; long. 79°46’E) during January – March, 2012 (Figure 1). The fishes were identified using FAO sheet and field guides and their total lengths were measured. For each species, ten representative sample groups with similar length and weight were taken for the rationale study (Table 1). Metal concentrations from the tissues were determined by following the method of Dhaneesh *et al.*, (2012a). After collection, they were immediately brought to the laboratory in an ice box and the fish was dissected to muscle, gills and liver while the shell fishes were dissected to gill and muscle. Muscle, gill and liver were cut into small pieces and dried at 70°C for 15 min. in a microwave oven (Technico, India).

After complete drying, samples were fine powdered using mortar and pestle and weighed to 1 g (± 0.01 g) and were digested in 100 ml glass beaker with concentrated nitric acid (20 ml) overnight. It was then mixed with 10 ml of concentrated nitric and perchloric acid solution (4:1) (v:v) followed by hotplate heating at 120°C up to complete dryness. The residue was then dissolved and diluted with 20 ml of a solution of deionised water and conc. nitric acid (4:1) (v:v) and then this solution was filtered through Whatman filter paper (11 µm) and metal levels were determined by using Inductively Coupled Plasma Optical Emission Spectrometer (Software – WinLab 32) (Perkin Elmer, Optima 2100DV). The precision of the analytical procedure was checked by analysing standard reference materials of commercially available standards (Merck KGCA, 64271 Damstadt, Germany, ICP-Multielement standard solution IV, 23 elements in nitric acid). All acids and chemicals were of analytical reagent grade. Metal concentrations were calculated in micrograms per gram dry weight (µg g⁻¹ d.w.). Two replicate analyses were made in order to have accuracy. All glassware were kept overnight in 10% nitric acid solution and rinsed with deionised water and air dried before use.

Pearson Correlation Coefficient was employed for the better understanding of relationship between the concentrations of various metals with various tissue types using statistical package of SPSS 16.0. One way ANOVA was employed to understand the variation in the concentration of heavy metals with respect to different species and organs.

Table 1. Marine organisms within feasible size range used for the determination of metal accumulation at Uppanar and Vellar estuaries

<table>
<thead>
<tr>
<th>S.NO</th>
<th>Species name</th>
<th>Size range (cm)</th>
<th>Mode of life</th>
<th>Feeding behavior</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.</td>
<td><em>Mugil</em></td>
<td>10.2-16.8</td>
<td>Benthopelagic</td>
<td>Carnivore</td>
</tr>
<tr>
<td>2.</td>
<td><em>Penaeus</em></td>
<td>3.2-5.3</td>
<td>Bethopelagic</td>
<td>Carnivore</td>
</tr>
<tr>
<td>3.</td>
<td><em>Crassostrea</em></td>
<td>-</td>
<td>Damersel</td>
<td>Herbivore</td>
</tr>
<tr>
<td>4.</td>
<td><em>Meretrix</em></td>
<td>-</td>
<td>Damersal</td>
<td>Herbivore</td>
</tr>
</tbody>
</table>

2 Results and Discussion

Concentrations of twelve elements-Cd, Co, Cu, Fe, Mg, Mn, Ni, Pb, Zn, Al, Cr and B concentrations were determined in different parts of the finfish (tissue, gill and liver) and shellfish (tissues and gill). The distributions of metal concentrations in tissue samples of different species, collected from Uppanar...
and Vellar estuaries are show in Table 2. Metal concentrations and the corresponding mean standard deviations measured in muscle tissues, gill, liver are depicted in Figure 2-5. In the present study, metals ranged as Cd: 0.10-0.72 µg g⁻¹, Co: 0.0-0.18 µg g⁻¹, Cu: 0.05-0.62 µg g⁻¹, Fe: 1.42-9.85 µg g⁻¹, Mg: 9.74-31.55 µg g⁻¹, Mn: 0.09-11.12 µg g⁻¹, Ni: 0.02-0.30 µg g⁻¹, Pb: 0.14-0.50 µg g⁻¹, Zn: 0.31-2.31 µg g⁻¹, Al: 1.26-3.95 µg g⁻¹, Cr: 0.25-2.91 µg g⁻¹ and B: 0.17-0.51 µg g⁻¹.

Cadmium is a serious contaminant, a highly toxic element, which is transported to sea through air. The limits of Cd for human consumption of fish is approximately 0.5 µg g⁻¹ dry wt. (CSHPF, 1995). However, in the present study, mean concentrations of cadmium ranged from 0.10 to 0.72 µg g⁻¹ and it was above the maximum permissible limit. Mendil et al., (2010) have reported 0.11 to 0.75 µg g⁻¹ cadmium in fish species from Yesilirmak river and it coincides with the present study.

Concentrations of Cu in the fish samples were between 0.05 and 0.62 µg g⁻¹. The maximum level of copper was observed in gill of Crassostrea madrasensis and minimum in muscle of Mugil cephalus in both stations. Dhaneesh et al., (2012a) report that the values for Cu are 0.98 ± 0.22 µg g⁻¹ and the present results were lower than these values and in similar range as reported by Vijayakumar et al., (2011) (0.42-0.61 µg g⁻¹). As copper is an essential part of several enzymes and necessary for synthesis of hemoglobin, most marine organisms have evolved mechanisms to regulate the concentrations of this metal in their tissues. The concentration of cobalt was found to be 0-0.18 µg g⁻¹ and it was below detectable level (BDL) in the muscle tissues of Mugil cephalus and maximum in gill of Crassostrea madrasensis. Turkmen et al., (2005) report that cobalt concentration varies between 0.73 and 1.91 mg.kg⁻¹ in the muscles of fishes of the Iskenderun Bay and lower Co levels in fish muscles (<0.001-0.002 mg.kg⁻¹) have been reported from the Mediterranean Sea region. These levels are in good agreement with our values. Cobalt is an essential nutrient for man and is an integral part of vitamin B₁₂ and the average daily intake of cobalt, in all forms, ranges 0.30-1.77mg/day.

Concentration of iron levels in tissues and gill was found to be higher as 1.42 and 9.85 µg g⁻¹ in Penaeus indicus and Mugil cephalus respectively Dhaneesh et al., (2012b) report the iron values as 1.6-6.64 µg g⁻¹ and these values are similar to the present study. Iron is an essential element and it is known that adequate iron in a diet is very important for decreasing the incidence of anemia in humans. Iron deficiency occurs when the demand for iron is high, for growth, high menstrual loss and pregnancy. Magnesium showed higher concentration in gill tissues than the muscle (31.55-9.74 µg g⁻¹) and these values are lower than the values reported by Dhaneesh et al., (2012b) (52.63-117.4 µg g⁻¹) and total daily intake of Mg varies from 2.5 to 7 mg in humans. Average manganese contents of the fish samples varied from 0.02-11.12 µg g⁻¹ in tissues. It is above the permissible limit and these values are higher than the previous reports of Biswas et al., (2011). Manganese is an essential element for both animals and plants and its deficiency results in severe skeletal and reproductive abnormalities in mammals. In the present study, concentration of nickel varied from 0.02 to 0.30 µg g⁻¹ and these values were lower than the previous reports of Biswas et al., (2011). Nickel is a hazardous element notified by the WHO (World Health Organization 1994) recommends 100-300 µg nickel for daily intake.

Lead concentrations in Vellar and Uppanar estuaries ranged from 0.14 to 0.50 µg g⁻¹ and these were higher than the value reported by Thiyagarajan et al., (2012) (0.22-0.27 µg g⁻¹). For an average adult (60 kg body weight), the provisional tolerable daily intake for lead, iron, copper and zinc are 0.2 mg, 241µg, 48 mg and 60 mg, respectively (Joint FAO/WHO, 1999). In the present study, the lead values in Uppanar estuary were higher than the permissible limits. It may be due to the high population and industrial effluent in the coastal waters of Uppanar estuary in the Vellar estuary and all the metal values were below within the acceptable level. Zinc concentration in Vellar and uppanar estuaries ranged from 0.31 to 2.31 µg g⁻¹ and these values were lower than the previous reports of Thiyagarajan et al., (2012). Zinc, is an essential micronutrient and as a constituent of many enzymes, it is responsible for certain biological functions, for which relatively higher level is required to maintain them. The recommended daily allowance is 10 mg/day in growing children and 15 mg/day for adults (NAS-NRC, 1974). A deficiency of zinc is marked by
Figure 2 Levels of Cd, Cu, Pb, Cr, Co, B (µg g⁻¹) in muscle, gill, and liver of fin and shell fishes of Uppanar estuary

Figure 4 Levels of Cd, Cu, Pb, Cr, Co, B (µg g⁻¹) in muscle, gill, and liver of fin and shell fishes of Vellar estuary

Figure 3 Levels of Fe, Mg, Al, Mn, Ni, Zn (µg g⁻¹) in muscle, gill and liver of fin and shell fishes of Uppanar estuary

Figure 5 Levels of Fe, Mg, Al, Mn, Ni, Zn (µg g⁻¹) in muscle, gill and liver of fin and shell fishes of Vellar estuary

Retarded growth, loss of taste and hypogonadism, leading to decreased fertility. Aluminium concentration ranged from 1.26 to 3.95 µg g⁻¹ in the Vellar and Uppanar estuaries. These values are higher than the earlier reported values as 0.45-1.50 µg g⁻¹ dry weight in fish species from Iskenderun bay, north east Mediterranean Sea, Turkey (Turkman et al., 2005). Ranau et al., (2001) have reported aluminium levels in the fish samples and they found higher aluminium accumulation in the fish species as 0.02-5.41 µg g⁻¹. Aluminum is not considered to be an essential element in humans. Exposure to aluminium has been implicated in a number of human pathologies including encephalopathy/dialysis dementia, Parkinson disease and Alzheimer’s disease. The permissible aluminium dose for an adult is quite high (60 mg/day) (World Health Organization, 1989).

Cr is an essential mineral for humans and has been related to carbohydrate, lipid, and protein metabolism. The maximum permitted level of Cr for human consumption is 50 µgg⁻¹ as per WHO (1989). In the present study, concentration of chromium was from...
0.25 to 2.91 µg g\(^{-1}\). The Cr level was comparatively higher in Uppanar estuary than the Vellar and it was above the maximum permissible limit. Canli and Atli (2003) have reported higher values (1.24-2.42 µg g\(^{-1}\)) than the present study. Average boron contents of fish samples ranged from 0.17 to 1.87 µg g\(^{-1}\), Yilmaz et al., (2010) have investigated the accumulation of boron in the fishes and they found below the limits of detection in all the tissues samples.

The relationships between metals in fin and shell fishes from Uppanar [Cd (R\(^2\) = 0.9989), Co (R\(^2\) = 0.9865), Cu(R\(^2\) = 0.3971), Fe (R\(^2\) = 0.8243), Mg (R\(^2\) = 0.1655), Mn (R\(^2\) = 0.9998), Ni (R\(^2\) = 0.9841), Pb (R\(^2\) = 0.9331), Zn (R\(^2\) = 0.05), Al (R\(^2\) = 0.0446), Cr (R\(^2\) = 0.997) and B (R\(^2\) = 0.4622) and Vellar [Cd (R\(^2\) = 0.3715), Co (R\(^2\) = 0.9865), Cu (R\(^2\) = 0.6976), Fe (R\(^2\) = 0.4946), Mg (R\(^2\) = 0.0023), Mn (R\(^2\) = 0.9995), Ni (R\(^2\) = 0.9883), Pb (R\(^2\) = 0.382), Zn (R\(^2\) = 0.7119), Al (R\(^2\) = 0.1082), Cr (R\(^2\) = 0.9798) and B (R\(^2\) = 0.2825) were evaluated by the regression method. Results obtained from the present study revealed that the concentrations of Co, Cu, Fe, Mg, Ni, Zn, Al, and B are below the permissible limits suggesting that these metals are at safer level. The concentration of Cd, Mn, Pb and Cr was above the permissible level in Uppanar and it might be due the presence of large scale industries which can significantly contribute to the heavy metal load in to the estuarine system from the SIPCOT (State Industrial Promotion Corporation of Tamil Nadu).

The industrialization has a striking influence on estuaries and coastal waters as these are extensively used as a repository of waste disposal and sewage disposal as a consequence of urbanization and agriculture runoff also add to this woe. Pollution changes the chemistry and upsets ecological relationships in the ocean; eventually it can disrupt the productivity of the oceans. Pollution of our seas and estuaries may cause irreversible and irreparable damage, therefore protecting them from the perils of pollution is now an international concern.

3 Conclusion

In conclusion, among the two areas selected for the study, Uppanar was highly polluted with Cd, Pb, Mn and Cr, higher than the maximum permissible levels. Concentrations are also above the threshold for fin \((Mugil cephalus)\) and shell fishes \((Penaeus indicus, Crassostrea madrasensis\) and Meretrix meretrix\) at present, a potential risk may emerge in the future, depending on the increasing chemicals, beverage manufacturing, domestic waste water disposal, oil, soap, paint production, paper and metal processing plants and boat transports. It is anticipated that, in future, the Uppanar may receive a huge amount of heavy metals due to the increasing industrial activities in SIPCOT region. In the Vellar estuary the run-off from the adjacent landmasses during monsoon is also an important factor behind accumulation of metal levels in the fish tissues. However, there should not be any health threat to the public resulting from the consumption of fish meat in this area. This study has enlightened the fact that persistent pollutants like metals should be regularly monitored and any variation from the normal distributional pattern can furnish an idea about the proper management of the coastal area. A competent monitoring programme is an essential adjunct to any attempt of managing the coastal areas in an ecologically sound and sustainable manner.

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Table 2 Concentration of trace metals accumulation (µg g⁻¹ dry wt.) in the fin and shell fishes in Uppanar and Vellar estuaries

<table>
<thead>
<tr>
<th>Metals</th>
<th>Uppanar estuary</th>
<th>Vellar estuary</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Mugil cephalus</td>
<td>Penaeus indicus</td>
</tr>
<tr>
<td>Cd</td>
<td>0.137±0.020</td>
<td>0.162±0.053</td>
</tr>
<tr>
<td>Co</td>
<td>0.037±0.047</td>
<td>0.316±0.515</td>
</tr>
<tr>
<td>Cu</td>
<td>0.103±0.044</td>
<td>0.165±0.038</td>
</tr>
<tr>
<td>Fe</td>
<td>1.710±0.233</td>
<td>4.134±0.676</td>
</tr>
<tr>
<td>Mn</td>
<td>0.552±0.622</td>
<td>4.072±6.111</td>
</tr>
<tr>
<td>Ni</td>
<td>0.408±0.518</td>
<td>0.673±1.080</td>
</tr>
<tr>
<td>Pb</td>
<td>0.164±0.017</td>
<td>0.221±0.070</td>
</tr>
<tr>
<td>Zn</td>
<td>0.847±0.486</td>
<td>1.394±0.800</td>
</tr>
<tr>
<td>Al</td>
<td>1.739±0.669</td>
<td>2.109±0.451</td>
</tr>
<tr>
<td>Cr</td>
<td>0.327±0.079</td>
<td>0.802±0.354</td>
</tr>
<tr>
<td>B</td>
<td>0.314±0.107</td>
<td>0.408±0.091</td>
</tr>
</tbody>
</table>