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Trace Metal Dynamics in Marine Shrimps from the Southwest Coast of India

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Concentrations of Fe, Co, Ni, Cu, Zn, Cd and Pb in the muscle tissue of marine shrimps, *Penaeus mondon*, *Penaeus japonicus*, *Penaeus indicus*, *Penaeus mergniensis*, *Parapenaeopsis hardwickii* and *Parapenaeopsis stylifera*, collected from the continental shelf waters off Kochi on southwest coast of India were determined. Inter species-specific tissue metals loads were appreciable within the Penaeid shrimps for the region. The ranges of higher concentrations of Fe (756.8 to 1288.5 mg/g), Co (8.8 to 18.6 mg/g), Ni (12.8 to 24.7 mg/g), Cu (32.8 to 46.8 mg/g), Zn (52.6 to 88.7 mg/g), Cd (5.8 to 9.6 mg/g), and Pb (0.8 to 1.9 mg/g) were observed in *P. mondon*, *P. japonicus*, *P. indicus*, *P. mergniensis*, and *P. hardwickii* characterized with an estuarine-sea life history when compared with the lowest concentrations of Fe (725.8 mg/g), Co (8.5 mg/g), Ni (10.6 mg/g), Cu (28.6 mg/g), Zn (44.8 mg/g), Cd (3.6 mg/g), and Pb (0.6 mg/g) found in *P. stylifera* with only a sea life history. This difference is possibly due to the favoring uptake of differently bioavailable metals from the associated food chain bioaccumulation processes within the estuarine and shelf waters off Kochi. The higher bioaccumulation factors 555867, 442857, 171528, 33621, 42933, 48731 and 4750 of the analyzed metals (Fe, Co, Ni, Cu, Zn, Cd & Pb) in the shrimp species *P. mondon* compared with others will support ecologists to use it as a bioindicator for metal pollution.

Keywords: Southwest coast of India, shrimp, food, trace metals, bioaccumulation

Trace metals at elevated concentrations are widespread inorganic contaminants in the aquatic environment. Due to easy accessibility and subsequent high human influence, rivers, estuaries and coastal waters are more vulnerable to their pollution effects. During estuarine mixing, trace metals discharged from industrial or sewage effluents may be rapidly precipitated as suspended matter and removed from the water column via geochemical transportation to bottom sediments (Forstner and Wittmann, 1981; Ouseph, 1992). Consequently, metal concentrations in the sediments are often found to be several orders of magnitude higher than those in ambient seawater (Luoma, 1990). Hence while investigating an aquatic environmental compartment for trace metal concentrations, their toxicity, and bioavailability to aquatic organisms, sediments can act as both carriers and potential sources of contaminants (Salomons and Forstner, 1984). An increase in trace metal concentrations in water is often directly linked to their content in the sediments, and consequently influences the levels of trace metals accumulated in fish (Luoma, 1989). Metals can easily penetrate fish through the gastrointestinal tract, skin, and gills, either directly

assimilated from water, or indirectly with food through the gastrointestinal tract. Although sediment ingestion can be a source of metal uptake, the geochemical controls of sediment on metal bioavailability are not yet fully understood (Jenne and Luoma, 1977; Luoma, 1989; Luoma et al., 1990; Newman and Jagoe, 1994).

The geomorphology (ox-bow shape) of the Cochin Estuary and meandering flow can induce the formation of perennially undulating water bodies or null zones (Soman, 1997). The weak flushing and increased anthropogenic activities during the last few decades have resulted in an entrapment of fine particles bound with trace metals loads in the sediments of the Cochin Estuary and simultaneously increased the metal concentration in the water and particulate matter (Nair et al., 1990; Balachandran et al., 2005) whereas the coastal circulation in the adjacent shelf has reduced its settling (Balachandran et al., 2003; 2006). The metal contamination of suspended and settling particles resuspended from the metal-rich sediments in the Cochin Estuary is a cause for concern as these metals may diffuse into the overlying water column under the prevailing environmental conditions and can undergo bioaccumulation and affect the benthic organisms (Engel et al., 1981; Luoma et al., 1990; Ouseph, 1992; Balachandran et al., 2006).

India, characterized with a long coastline (8129 km), a wide continental shelf (0.5 million km²) and a large Exclusive Economic Zone (2.02 million km²), is the leading seafood producer

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and exporter among the Asian countries. Due to upwelling, the coastal waters of the southwest coast of India are among the most productive regions in the world, supporting abundant invertebrate and fish populations, many of which are harvested as food (Madhupratap et al., 2001). Shrimp populations are a significant component of a larger invertebrate community structure in the Northern Indian Ocean as well as other oceanic regions (Venkatraman, 1960; Suseelan and Nair, 1990; Rogers, 1998). Moreover the vast brackish water along the entire coastline of India, owing to high productivity, is extremely favorable for the growth of many marine fishes, invertebrates and especially shrimps that utilize this biological niche for completing their life-cycle (George, 1968). Considerable variations occur among shrimps, both in the degree to which the brackish water environment is utilized by each species in their life history and in the distribution of parent and juvenile populations in brackish-marine environments (Turner, 1977). Penaeid shrimps such as *P. mondon*, *P. japonicus*, *P. indicus*, *P. mergniensis*, and *P. hardwickii* have a distinctive life history characterized by a more or less predictable length of time spent in the estuarine or brackish and coastal environments, with an exception to this being *P. stylifera* which spends its entire life in the marine environment (Kutkuhn, 1966). In each of the species the parent population breeds in the sea at varying distances from the shoreline and the large number of microscopic, semi-buoyant eggs shed into the sea hatch out into small planktonic larvae. The eggs, larvae and post-larvae of most species are found abundant in the inshore waters in summer monsoon. The younger stages migrate to the estuarine or brackish water environments and once inside the brackish waters, the post-larvae leave at the planktonic stage, settle to the bottom in the shallower regions, grow fairly rapidly to large size from 6 to 9 months, and finally migrate back into the sea where sexual maturity is attained for spawning to complete the life-cycle.

Penaeid shrimp are important resources for worldwide fisheries and aquaculture, and when in larval stages they serve as an important food source to oceanic tuna, flying fishes and a number of other fish species inhabiting in shelf waters supporting commercial fisheries (James et al., 1987; Philip, 1998). The importance of shrimp as sonic scatter sources and as agents in energy transfer to the deep sea is well documented (Omori, 1974; Kirkuchi and Omori, 1985). Detritivores such as post larval and juvenile penaeid shrimps are essential components for converting primary production in estuaries into available forms to higher consumers in the ecosystem. Among the maritime regions of India, the southwest coast is still the major producer of Indian shrimp (Karuppasamy et al., 2006). Traditionally in the Kerala state, off Cochin is well known for shrimp grounds, which were comparable to the world's best grounds in the United States of America and Mexico. There were only few random reports on the metal concentrations in some shrimp species from selected areas adjacent to the Indian Seas available (Zingde et al., 1976; Krishnamurti and Nair, 1999; Hossain and Khan, 2001); however, trace metal bioaccumulation patterns in shrimp from the continental shelf waters of the southwest coast of In-

dia have been poorly investigated. A high degree of variability in the bioaccumulation for different metals, exhibited by marine organisms like fishes and shrimps were used for evaluating the general background as well as toxic levels and for utilizing them as indices of pollution (Zingde et al., 1976; Mokhtar et al., 2009). In this study, we determined the concentration levels as well as bioaccumulation factors of trace metals in various Penaeid shrimp species collected from southwest coastal waters of India at a location off Kochi, and further discussed the controlling factors.

Materials and Methods

As part of the Marine Research-Living Resource Programme (MRLR), trace metal concentrations in the muscle tissue of various Penaeid shrimps like *Penaeus mondon*, *Penaeus japonicus*, *Penaeus indicus*, *Penaeus mergniensis*, *Parapenaeopsis hardwickii* and *Parapenaeopsis stylifera* were determined. The selected species are economically important and are abundant in the study area. Shrimp samples were collected along a transect off Kochi (10° N latitude and 75.60° E longitude.), southwestern India, on Cruise 217 of shipboard Fishery and Oceanographic Research Vessel (FORV) Sagar Sampada in September to October 2003. Sampling was conducted using Expo bottom trawl nets, and samples were subjected to close visual observation under a binocular microscope to ensure the absence of any foreign particles. Each shrimp sample was placed in a small nylon sieve and thoroughly rinsed with Milli-Q water for removing salts. Water adhering to the samples was removed by placing the sieve on good quality contaminant-free Millipore filter paper. Subsequently, dissected muscle of the shrimp samples was dried in an oven at 65° C and stored in a vacuum desiccator.

The dried muscle was powdered and aliquots of approximately 300 mg were digested for 3 hours at 80° C with 0.3 ml HNO₃ (65%, Suprapure, Merck) in tightly closed 2 mL Eppendorf reaction tubes. The digests were diluted to 25 mL with HCl (0.1N) and metals were analyzed using Flame Atomic Absorption Spectrophotometry (PE A Analyst100). The precision and accuracy of analysis were checked by replicate measurements (n = 5) of target metals in a standard reference material of marine biota sample (Defatted Lobster Tomalley; TORT-2, Lobster Hepatopancreas, National Research Council, Canada). The values obtained for the reference material were in excellent agreement with the certified values (Table 1).

Bioaccumulation factors (concentration factors) were calculated for each metal in different shrimps as the ratio of the metal concentration in the organism's body over its concentration in the ambient medium (data from Rejomon, 2005). All metal concentrations in the muscle tissues of various shrimps are reported in µg/g dry weight, since dry weights rather than wet weights provide a more stable basis for comparison.

Pearson's correlation coefficients were used to examine relationships between the elements in muscle of the shrimp species.

Table 1. Accuracy (given as recovery) and precision (given as standard error) of standard Lobster Hepatopancreas (TORT-2), analyzed according to the procedure used for shrimp metal analysis.

Metal	Concentration ($\mu\text{g/g}$; mean \pm s.d.)		Recovery(%)
	Certified	Measured	
Fe	105 \pm 13	106.5 \pm 3.13	101
Co	0.51 \pm 0.09	0.50 \pm 0.08	98
Ni	2.50 \pm 0.20	2.52 \pm 0.22	100
Cu	106 \pm 10	105 \pm 5.03	99
Zn	180 \pm 6	181.2 \pm 4.78	100
Cd	26.7 \pm 0.6	26.9 \pm 0.8	100
Pb	0.35 \pm 0.13	0.36 \pm 0.11	102

The concentrations of the elements in shrimps were compared by statistical means, that is, one-way ANOVA.

Results

Trace Metal Levels in Shrimps

The statistical summary of lengths and weights of and metal concentrations in muscle tissue of various Penaeid shrimps are presented in Table 2. For further calculating the bioaccumulation factors, average concentration of dissolved trace metals in the studied coastal waters are also given in Table 3, which were taken from Rejomon (2005). The corresponding bioaccumulation factors for each species were given in Table 4.

The concentration levels of metals in the various shrimps followed an order: *P. mondon* > *P. japonicus* > *P. indicus* > *P. mergniensis* > *P. hardwickii* > *Pstylifera*. The statistical significance of differences of trace metal levels (Table 5) among various shrimps was tested by one-way analysis of variance (ANOVA). Results indicate that variability in concentrations of metals among the species such as *P. mondon*, *P. japonicus*, *P. hardwickii* and *P. stylifera* were statistically significant for all the metals. However the variability for metals Fe, Co, Ni, Cu & Zn in *P. indicus* and Fe, Ni, Cu, Zn & Cd in *P. mergniensis* when compared with *P. mondon*, *P. japonicus*, *P. hardwickii* and *Pstylifera* were statistically insignificant. The species *P. mondon* showed higher concentration levels for all metals, attributed to high length and weight, but length and weight for other species

Table 3. Average dissolved trace metal concentrations ($\mu\text{g/l}$) in the water column of the coastal areas off Kochi (Rejomon, 2005)

Trace metal	Fe	Co	Ni	Cu	Zn	Cd	Pb
Off Kochi	2.318	0.042	0.144	1.392	2.066	0.197	0.400

appear to not the main factors controlling the levels of trace metal concentrations.

Iron

Fe is a common metal required by both plants and animals. It is an important biochemical ingredient in the production of plankton in the oceans. It is a vital part of the oxygen transport mechanism in the blood (hemoglobin) of all vertebrate and some invertebrate animals. Fe concentrations in muscle of the shrimps collected off Kochi showed large inter-specific variation, ranging from 726 to 1289 $\mu\text{g/g}$ (Table 2). The observed Fe contents were higher than those reported for shrimps from the East Atlantic waters (11 to 34 $\mu\text{g/g}$) (Harding and Goyette, 1989), Hong Kong coastal waters (32 to 781 $\mu\text{g/g}$) (Rainbow, 1986), and Bay of Bengal waters (9.1 to 16 $\mu\text{g/g}$) (Hossain and Khan, 2001).

Cobalt

As an integral part of the vitamin B₁₂ complex, cobalt is essential in trace amounts for humans and animal life. Co contents in muscle tissue of different species of shrimp collected from off Kochi ranged from 8.5 to 19 $\mu\text{g/g}$ (Table 2). Here the measured cobalt showed wide fluctuations except for *P. mergniensis*, *P. hardwickii* and *P. stylifera*, which was found to be almost constant.

Nickel

Ni can accumulate in aquatic life, but its presence is not magnified along food chains. Ni concentrations in muscle tissues ranged from 11 to 25 $\mu\text{g/g}$ (Table 2). The Ni contents in these shrimps collected off Kochi were much higher than the reported ranges of Ni content for shrimps from the near shore waters off Mumbai (0.01 to 0.09 $\mu\text{g/g}$) (Krishnamurti and Nair, 1999), Bay

Table 2. Lengths, weights and trace metal concentrations ($\mu\text{g/g}$) in the muscle tissues of various shrimp species from the coastal waters off Kochi

Shrimp Species	Length (cm)	Weight (g)	Fe (ppm)	Co (ppm)	Ni (ppm)	Cu (ppm)	Zn (ppm)	Cd (ppm)	Pb (ppm)
<i>Penaeus mondon</i>	18.8	10.7	1289	19	25	47	89	9.6	1.9
<i>Penaeus japonicus</i>	13.6	7.8	1226	16	23	43	79	9.5	1.7
<i>Penaeus indicus</i>	12.8	6.6	1044	13	19	40	74	8.7	1.6
<i>Penaeus mergniensis</i>	13.8	7.5	888	9.5	15	37	57	6.5	0.9
<i>Parapenaeopsis hardwickii</i>	15.6	6.8	757	8.8	13	33	53	5.8	0.8
<i>Parapenaeopsis stylifera</i>	14.5	7.5	726	8.5	11	29	45	3.6	0.6

Table 4. Bioaccumulation factors for trace metals in the various shrimp species from the coastal waters off Kochi.

Shrimp Species	Fe	Co	Ni	Cu	Zn	Cd	Pb
<i>Penaeus mondon</i>	555867	442857	171528	33621	42933	48731	4750
<i>Penaeus japonicus</i>	528818	376191	156944	30532	38045	48223	4150
<i>Penaeus indicus</i>	450216	304762	130556	28376	35721	44162	3925
<i>Penaeus mergniensis</i>	382916	226191	101389	26221	27396	32995	2200
<i>Parapenaeopsis hardwickii</i>	326488	209524	88889	23563	25460	29442	1900
<i>Parapenaeopsis stylifera</i>	313115	202381	73611	20546	21684	18274	1375

of Bengal waters (2.9 to 5.9 $\mu\text{g/g}$) (Hossain and Khan, 2001), and Gresik coastal waters off Indonesia (0.01 to 0.05 $\mu\text{g/g}$) (Soegianto and Hamami, 2007). The elevated Ni content in the various shrimp species might be attributed to the extensive upwelling occurring in the area, which brings Ni rich subsurface waters to the surface (Rejomon, 2005).

Copper

Oceans, tidal pools, lakes, rivers, and ponds—all bodies of water that sustain life—have copper present as a vital naturally occurring element for normal growth in living organisms. Copper levels in muscle tissue of the shrimp species collected off Kochi ranged from 29 to 47 $\mu\text{g/g}$ (Table 2). The observed Cu contents agree well with the reported ranges of Cu contents in shrimps collected from the Goa offshore waters (48 $\mu\text{g/g}$) (Zingde et al., 1976), Hong Kong coastal waters (31 to 84 $\mu\text{g/g}$) (Rainbow, 1986), northeast Pacific coastal waters (14 to 20 $\mu\text{g/g}$) (Harding and Goyette, 1989), Malaysian coastal waters (13 to 159 $\mu\text{g/g}$) (Awaluddin et al., 1992; Patimah and Dainal, 1993), near shore waters off Mumbai (11 to 38 $\mu\text{g/g}$) (Krishnamurti and Nair, 1999), Bay of Bengal waters (12 to 21 $\mu\text{g/g}$) (Hossain and Khan, 2001), and Gresik coastal waters off Indonesia (19 to 29 $\mu\text{g/g}$) (Soegianto and Hamami, 2007).

Zinc

Zn is an essential trace element in biological systems. It is involved in nucleic acid synthesis and occurs in many enzymes. However, high concentrations of Zn in the aquatic environment are detrimental to fish and aquatic life. Zn is the next abundant element followed by Fe in the examined shrimp species. Zn contents (Table 2) in shrimps collected off Kochi show wide

fluctuations and range from 44.8 to 88.7 $\mu\text{g/g}$. Our data are well within the ranges of values reported for shrimps from the Hong Kong coastal waters (39 to 146 $\mu\text{g/g}$) (Rainbow, 1986), the northeast Pacific coastal waters (48 to 53 $\mu\text{g/g}$) (Harding and Goyette, 1989), Malaysian coastal waters (68 to 186 $\mu\text{g/g}$) (Patimah and Dainal, 1993), and coastal waters of Mumbai (9.2 to 130.2 $\mu\text{g/g}$) (Krishnamurti and Nair, 1999). However, the ranges of Zn concentrations in shrimps investigated are higher than the reported ranges for shrimps from the Bay of Bengal waters (24.2 to 35.7 $\mu\text{g/g}$) (Hossain and Khan, 2001), and Gresik coastal waters off Indonesia (30.3 to 36.6 $\mu\text{g/g}$) (Soegianto and Hamami, 2007). The occurrence of extensive upwelling in the study area that carries Zn-rich subsurface waters to surface might explain why elevated Zn concentrations are measured in the muscle tissue of adverse shrimps in the present study (Rejomon, 2005).

Cadmium

Cadmium is bioaccumulated in organisms with the main uptake routes, being dissolved Cd from the water column and Cd associated with preys. Cd is bio-persistent and, once absorbed by an organism, could remain resident for many years although it is eventually excreted. Cd contents in muscle tissue of various shrimps in Kochi offshore waters were detectable and ranged from 3.6 to 9.6 $\mu\text{g/g}$ (Table 2). Cd levels detected were consistent with the ranges of values, reported for shrimps of the eastern Atlantic Ocean (1.9 to 7.1 $\mu\text{g/g}$) (Rainbow, 1986), and Malaysian coastal waters (1.6 to 6.1 $\mu\text{g/g}$ & 0.21 to 49.0 $\mu\text{g/g}$) (Awaluddin et al., 1992; Patimah and Dainal, 1993). However, Cd content observed in the present study is relatively higher than the reported ranges of Cd content for shrimps from the Mumbai coastal waters (0.001 to 1.5 $\mu\text{g/g}$) (Krishnamurti and

Table 5. Results of one-way ANOVA for testing the significance of differences (*F*-ratios) in metal concentrations on different shrimp species

Source of Variation	Degree of Freedom	Fe	Co	Ni	Cu	Zn	Cd	Pb
<i>Penaeus mondon</i>	1	14.955*	28.569*	16.785*	19.553*	18.214*	6.954**	13.321*
<i>Penaeus japonicus</i>	1	7.595**	4.787**	6.378**	3.361***	3.724***	6.214**	4.216**
<i>Penaeus indicus</i>	1	0.319	0.073	0.391	0.397	1.322	2.201***	2.506***
<i>Penaeus mergniensis</i>	1	1.075	2.999***	1.467	0.221	1.826	0.632	2.351***
<i>Parapenaeopsis hardwickii</i>	1	7.061**	5.012**	4.494**	3.814***	4.072**	2.434***	4.685**
<i>Parapenaeopsis stylifera</i>	1	9.915*	6.126**	12.759*	20.918*	13.865*	31.541*	12.725*

Significant variance ratios are indicated by at * $p \leq 0.005$, ** $p \leq 0.05$, *** $p \leq 0.10$

Nair, 1999), Bay of Bengal waters (0.2 to 0.3 $\mu\text{g/g}$) (Hossain and Khan, 2001), and Gresik coastal waters off Indonesia (0.01 to 0.05 $\mu\text{g/g}$) (Soegianto and Hamami, 2007).

Indeed, the elevated Cd concentrations in shrimp muscles highlight the exposure to this metal. Cd concentrations in seawater are known to be enhanced in areas of upwelling and Cd has been shown to maintain a positively linear relationship with phosphorus in upwelled waters (Bruland, 1983). This Cd could be readily bioaccumulated in the lower portion of the food chain and biomagnified by trophic transfers to high levels in the shrimp. The apparent enrichment of Cd in muscle tissue is likely due to food chain transfer of high levels of Cd brought into surface waters through the strong upwelling of nutrient-rich waters that was occurring during the summer monsoon (Rejomon, 2005). The increase of Cd bioavailability in the southwest coast of India by upwelled deep waters might be at the origin of an increase of Cd concentrations in the marine biota on a local scale.

Lead

Pb is one of the most serious environmental pollutants and is a non-essential trace element toxic to biota at elevated concentrations. Pb is the least abundant element examined in the shrimps muscle tissues. Pb concentrations (Table 2) of shrimps were detectable and ranged from 0.6 to 1.9 $\mu\text{g/g}$. Pb levels measured in our study are well within the ranges of Pb contents reported for shrimps from the northeast Pacific coastal waters (0.7 to 1.3 $\mu\text{g/g}$) (Harding and Goyette, 1989), and Bay of Bengal waters (0.8 to 1.3 $\mu\text{g/g}$) (Hossain and Khan, 2001). However our Pb concentrations are higher than the reported data (0.01 to 0.09 $\mu\text{g/g}$) for shrimps from the shelf waters off Mumbai (Krishnamurti and Nair 1999).

Discussion

The different concentration levels of metals in the penaeid shrimps indicate a distinct bioaccumulation strategy for each species and suggest that trace metal absorption from the prey (planktonic and benthic organisms) is not uniform in the habitat of the southwest coast of India. Further the results indicated that the concentration of trace metals like Fe, Ni, Zn and Cd in shrimps are higher than those reported from other areas. This may be due to variable bioavailability of metals in the estuarine and coastal environments. For example, *P. stylifera* which spends its entire life-cycle in the marine environment is exposed to a relatively low degree of bioavailable metals (mostly natural contribution from upwelling), whereas *P. mondon* spends part of its life-cycle in the estuarine environment and, as a consequence, is exposed to a higher degree of bioavailable metals (mainly anthropogenic contribution from industrial effluents) other than marine environment. Therefore the two species showed a low and high affinity for metal bioaccumulation, respectively. Additionally, the differences in life history patterns influence exposure of bioavailable metals (including trophic levels and geo-

graphic distribution of life stages) to shrimps in a different way and hence a different enrichment pattern for various species, as discussed below.

Factors Influencing Trace Metal Levels in Shrimps

Enhanced Concentration of Metals in the Estuarine and Shelf Waters

Metals bound in the sediment via physical adsorption and chemisorption is in a balance with sediment interstitial waters, and hence can easily diffuse to the water column of the estuarine and shelf environments (Tessier et al., 1985; O'Day et al., 2000). The southwest coast of India (estuarine and shelf waters off Cochin) has seasonally high concentration of trace metals (Fe, Co, Ni, Cu, Zn, Cd, and Pb) in the water column as a result of diagenetic remobilization from the contaminated sediments and desorption from resuspended sediments (Harper et al., 1998; Balachandran et al., 2003). During the summer monsoon period, the estuarine and shelf waters are highly productive and the underlying sediments are correspondingly rich in organic matter. The prevalence of reducing condition due to high benthic oxygen demand in the organic rich sediments favors formation of metal sulphides (Rajamani, 1994; Vigneault et al., 2001). The remobilization of metals can still occur into the overlying water body because of the resuspension of anoxic sediments by undercurrents and the subsequent re-dissolution of the previously deposited metal sulphide species due to oxidation, following with mixing up into the surface waters by tidal currents and upwelling (Harper et al., 1998; Rejomon, 2005; Martin et al., 2010). Owing to the physical processes, the enrichment of trace metals in the estuarine and shelf waters increases both the concentration of trace metals and the biological productivity that concentrates them (Madhupratap, 1987). Increased ambient metal concentrations are absorbed by the primary as well as secondary producers (Luoma et al., 1998; Rejomon et al., 2008) thereby increasing metal bioavailability to the food chain within the estuarine and shelf waters. The enhanced productivity facilitates growth and trophic transfers of both benthic and pelagic microorganisms, amplifying biomagnification, leading to different metal enrichment patterns in shrimps.

Absorption and Bioavailability of Metals

Uptake of metals by shrimps depends on their bioavailability (Carvalho et al., 1999). In general, the absorption and bioavailability of metals are affected by the physiology of aquatic organism and ecology, and various environmental factors that govern dissolved metals in the water column (Jenne and Luoma, 1977; Engel et al., 1981; Chen et al., 2000). During the summer monsoon period the enhanced concentrations of metals and high biological production in the estuarine and shelf waters play a prominent role in structuring of phytoplankton and zooplankton species in the surface layer and consequently affect to the recruitment of pelagic and demersal invertebrates due to higher food availability (Madhupratap et al., 1990; Muraliedharan et al., 2007). Among the environmental hazards associated

with the discharge of industrial effluents to estuaries are the metals, which are released during remineralization of organic matter (Forstner and Wittmann, 1981; Nair and Ramachandran, 2002; Doong et al. 2008). These metals could become bioavailable on a local scale (Balachandran et al., 2006) probably due to formation of weak metal-dissolved organic complexes (Chakraborty et al., 2009) and are potentially trapped by the biota (Hudson, 1998). Organic matter discharged from wastewater effluents into the estuaries can increase bioavailable metal ions by lowering the pH and forming metal chelates that are subsequently absorbed by the planktonic and benthic food webs (Forstner et al., 1989; Phinney and Bruland, 1997; Sedlak et al., 1997; Simpson et al., 1998). The demersal species are expected to contain higher metal concentrations than pelagic species, which may be related to the greater exposure to metal enriched bottom sediments and its interaction with benthic organisms (Campbell et al., 1988). Even though most of the shrimps are occasionally pelagic in their search for food, they are mainly bottom dwelling inhabitants (Dall et al., 1990; Bailey-Brock and Moss, 1992), and thus have a direct contact between gills and the near bottom waters with an increased propensity for uptake of trace metals through the gills (Campbell et al., 1988). Exposure to metals for the shrimp and other invertebrate species is mainly through their food (diet) and to a limited extent by direct absorption from the water column (Luoma, 1995; Wang and Fisher, 1999). Accordingly, the different enrichment pattern of Fe, Co, Ni, Cu, Zn, Cd, and Pb in the shrimps off Cochin was likely due to different bioavailability of these elements arising from the food chain as a consequence of varying prey availability in the estuarine and shelf waters during the high-production summer.

The feeding habit, life cycle and physiological efficiency of shrimp species to metabolize various metals are the major factors that contribute to the level of metals (Zhou et al., 2008). Penaeid shrimps are known to ingest a variety of whatever plant or animal material that is readily available and are described as omnivorous scavengers (filter-feeders), opportunistic omnivores, detritus feeders, carnivores, and predators (Eriksen and Belk, 1999; Roberts, 2005). The apparent different accumulation strategies of the six species for the metals (Fe, Co, Ni, Cu, Zn, Cd, and Pb): *P. mondon* > *P. japonicus* > *P. indicus* > *P. mergniensis* > *P. hardwickii* > *P. stylifera* may be due to variable bioavailability resulting from a variable competence for metal absorption from their feeding food habits at the ecological regimes of the estuarine and shelf waters. Moreover, the possible differences in retention time of the various species in polluted estuarine waters can be accounted for the differences in the metal accumulation patterns. The enrichment of trace metals in the species *P. mondon*, *P. japonicus*, *P. indicus*, *P. mergniensis*, and *P. hardwickii* (characterized with a more or less predictable length of time spent in the estuarine environments during the early life-stages) when compared with the species *P. stylifera* (characterized with the entire life in the marine environments) is likely to be due to food chain absorption of high degree of bioavailable metals at the early life-stages by filtering microalgae from the water at proto-

zoal stages, and capturing zooplankton at mysis and postlarval stages in the estuarine waters (Holthuis, 1980; Kumlu, 1999). Higher metal concentrations in *P. indicus* than *P. mergniensis* even though they feeds on vegetable material and crustaceans (Gopalakrishnan, 1952; Chong and Sasekumar, 1981), is tentatively attributed to variable bioavailability resulting from a variable competence for metal absorption from their feeding food habits at these ecological regimes. The high metal content noted in the highly mobile species *P. mondon* that effectively swims throughout the water column horizontally and vertically indicates that it is more of a predator rather than a scavenger or detritus feeder (Grey et al., 1983; Devi, 1988; Bauer, 2006; DFO, 2006). Thus, the high enrichment of Fe, Co, Ni, Cu, Zn, Cd and Pb in the shrimp *P. mondon* when compared with other species is likely to be due to the resulting high bioavailability of these metals from extensive feeding on a variety of crustaceans and mollusks (by 85% ingested food), and the remaining 15% consisting of vegetable matter, polychaetes, small fish, debris and sand (George et al., 1985; Solis, 1988; Dall et al., 1990) and by a high metabolism.

Bioaccumulation of Trace Metals in Shrimps

Metal concentrations at any trophic level may result from a combination of bioconcentration (uptake from the water) and bioaccumulation (uptake from the diet) processes particular to each metal and taxa. The bioaccumulation factor (concentration factor) was evaluated in relation to the concentration of the soluble trace metal in the seawater. In general the non-essential element Pb is bioaccumulated least efficiently, while the essential element Fe is bioaccumulated with a higher efficiency in all shrimp species (Table 4). The shrimps showed a higher range of bioaccumulation factors for the metals like Fe (313115–555867), Co (202381–442857), Ni (73611–171528), Cu (20546–33621), Zn (21684–42933), Cd (18274–48731) and Pb (1375–4750). These values were quite comparable with the ranges of bioaccumulation factors reported for metals like Fe (189104–322808), Co (134917–423037), Ni (44126–96891), Cu (1567–2600), Zn (14428–40812), Cd (8516–29407) and Pb (575–1404) in fish of the southwest coast of India (Rejomon et al., 2009). The degree of metal bioaccumulation in shrimps may depend on absorption and elimination processes, exposure metal concentrations in the surrounding medium, environmental factors (e.g., the level of dissolved organic matter); it may varies greatly with the individual species (Bradley and Sprague, 1985; Hare and Tessier, 1996; Wang and Fisher, 1999a, 1999b; Hashmi et al., 2002).

Among the shrimp species examined, *P. mondon* and *P. stylifera* had the largest and the narrowest ranges of bioaccumulation factors for the analyzed metals, respectively. Irrespective of food sources, metal bioaccumulation varies with bioavailability, uptake rate, the organism's threshold limit, and its physiological efficiency to excrete excess metals (Davies and Dobbs, 1984; Phillips, 1995). Hence, various species of shrimps at the same location showed different accumulation metal levels.

Table 6. Pearson product-moment correlation coefficients between length, weight and trace metals in the muscle tissue of shrimps

Metal	Length	Weight	Fe	Co	Ni	Cu	Zn	Cd	Pb
Length	1.00								
Weight	0.85**	1.00							
Fe	0.31*	0.65**	1.00						
Co	0.47*	0.75**	0.98**	1.00					
Ni	0.35*	0.65**	0.98**	0.98**	1.00				
Cu	0.38*	0.66**	0.97**	0.95**	0.98**	1.00			
Zn	0.36*	0.63**	0.98**	0.97**	0.98**	0.98**	1.00		
Cd	0.17	0.44*	0.95**	0.89**	0.96**	0.96**	0.96**	1.00	
Pb	0.28*	0.56**	0.97**	0.95**	0.98**	0.96**	0.98**	0.96**	1.00

Significant levels are indicated by * at $p < 0.05$ and ** at $p < 0.001$

For many aquatic invertebrates, the trophic transfer of metals from prey to predator accounts for a major portion of the total metal accumulation (Reinfelder et al., 1998; Wang and Fisher, 1999). Additionally, the relatively higher level of metal concentrations in shrimps may also be due to ineffective elimination of trace metals by biota (Krishnamurti and Nair, 1999). Because shrimps often have higher bioaccumulation factors of trace metals than fishes, they, especially *P. mondon*, which has the highest factor, hence could serve as an indicator for biomonitoring metal pollution.

In addition, the concentration factors of the analyzed metals in *P. mondon*, *P. japonicus*, *P. indicus*, *P. mergniensis*, and *P. hardwickii* followed the order: Fe > Co > Ni > Cd > Zn > Cu > Pb, whereas in the species *P. stylifera* the order is: Fe > Co > Ni > Zn > Cu > Cd > Pb. However, metal contents in all shrimp species followed the same order: Fe > Zn > Cu > Ni > Co > Cd > Pb, probably due to the same metabolic requirements of these metals in the individuals.

Interestingly, when considering all the metal contents, and bioaccumulation and concentration factors for each metal examined in the study, both Fe and Pb rank the highest and lowest ones, respectively, regardless of the shrimp species. This particularly highlights the importance of bioavailability of metals in seawater and prey to the shrimps.

Inter-Elemental Relationships in Shrimps

Possible association among the elements (inter-element correlations) in the muscle tissue of six shrimp species were assessed by means of Pearson's correlation coefficients; the results are given in Table 6. In the present study, the essential element Fe showed high correlations with six other elements, Co, Ni, Cu, Zn, Cd and Pb. Co also showed high correlations with Ni, Cu, Zn, Cd and Pb. Additionally, Ni had good correlations with Cu, Zn, Cd and Pb. Cu had significant correlations with Zn, Cd and Pb. Zn also showed high correlations with the toxic elements, Cd and Pb; another correlation between the toxic metals Cd and Pb was also noted. Pourang et al. (2005) also found similar inter-elemental correlation between the essential and non-essential elements in the fish and shrimp tissues from the Persian Gulf waters. The significant correlations found among most of the

elements may reflect a common source of trace metals. Besides, the correlations between the different elements in shrimps may result from the similar accumulation behavior and their interactions (Pourang et al., 2004; Kojadinovic et al., 2007; Rejomon et al., 2009). These inter-elemental relationships for essential and non-essential elements noted in the muscle tissue have been regarded as indicative of similar biogeochemical pathways for metal accumulation in shrimps.

Hazard Level

Regarding the risk of elevated metal amounts to marine organisms, much attention has been paid to Cu, Zn, Cd and Pb. Among these metals, Pb and Cd are classified as toxic metals causing chemical hazards, and therefore maximum residual levels have been prescribed for human consumption by various agencies of food standards (Food and Agriculture Organisation [FAO], 1983; European Commission [EC], 2001). For comparing with food standards of metals, the metal concentrations in the shrimp tissue usually presented in dry weight (Table 7) need to be converted to wet weight-based concentrations by dividing them by a factor of 4–6 (Khan et al., 1987; Parsons, 1999; Romeo et al., 1999; California Environmental Protection Agency, 2002; Lewis et al., 2002); here the factor (5) was adopted. Because the food standards of trace metals in shrimps are unavailable over the world, therefore we compared the trace metals of shrimp with those of fish. The food standards for fish set by the FAO (1983) are wet weight-based concentrations of 40.0, 30.0, 0.5 and 0.5 $\mu\text{g/g}$ for Zn, Cu, Cd and Pb, respectively. The derived wet weight based- concentrations of Zn, Cu, and Pb in our shrimps were well below the thresholds of concern, whereas for Cd it was slightly above the acceptable level. When further comparing with the European food standards for fish (i.e., wet weight based concentrations 0.2–0.4 $\mu\text{g/g}$ for Pb and 0.05–0.10 $\mu\text{g/g}$ for Cd) (EC, 2001), results also showed that Pb was lower than the guidelines for the edible part of the examined shrimp whereas Cd was slightly above the acceptable limits for human consumption. On the basis of the recommended daily dietary allowances (RDA) for safe consumption of the fish muscle, the allowed intake was regulated at 50–350 mg of metal per 100 g serving of the muscle (United States National Research Council, 1980). From this

Table 7. Comparison of trace metal concentrations in the muscle of different shrimp species (Concentrations are in ppm, dry weight) from the Indian coastal waters as well as maximum allowable concentrations of trace metals in sea food (*Concentrations are in ppm, wet weight) for human consumption

Geographical areas/Standards	Shrimp Species	Fe	Co	Ni	Cu	Zn	Cd	Pb	References
Cochin coastal waters	<i>Penaid shrimps</i>	725.8–1288.5	8.5–18.6	10.6–24.7	28.6–46.8	44.8–88.7	3.6–9.6	0.6–1.9	Present study
Goa coastal waters	<i>Penaeus mondon</i>				48.3	70.0			Zingde et al. (1976)
Mumbai coastal waters	<i>Penaeus indicus</i>			0.01–0.08	16.9–37.4	41.9–72.0	0.04–0.10	0.02–0.09	Krishnamurti & Nair (1999)
Mumbai coastal waters	<i>Parapenaeopsis hardwickii</i>			0.008–0.03	22.6–30.5	64.7–130.2	0.03–1.2	0.01–0.07	Krishnamurti and Nair(1999)
Mumbai coastal waters	<i>Parapenaeopsis sculptilis</i>			0.004–0.09	10.5–37.8	9.2–85.0	0.001–1.5	0.01–0.07	Krishnamurti and Nair(1999)
Bay of Bengal waters	<i>Penaeus mondon</i>	9.1–15.7		2.9–5.9	12.2–21.3	24.2–35.7	0.2–0.3	0.8–1.3	Hossain & Khan, (20010
Gresik coastal waters, Indonesia	<i>Penaid shrimps</i>			0.01–0.05	19.5–28.9	30.3–36.6	0.01–0.05		Soegianto & Hamami (20070
Hong Kong coastal waters	<i>Penaid shrimps</i>	32.0–781.0			31.0–84.0	39.0–146.0			Rainbow, 1986
Pacific coastal waters	<i>Penaid shrimps</i>				14.0–20.0	48.0–53.0	0.1	0.7–1.3	Harding & Goyette (1989)
Atlantic ocean waters	<i>Penaid shrimps</i>	11.0–34.0			17.0–99.0	51.0–83.0	1.9–7.1	1.5	Rainbow (1986)
Malaysian coastal waters	<i>Penaeus mondon</i>				12.8–159.0		1.6–6.1	4.6–32.0	Awaluddin et al. (1992)
Malaysian coastal waters	<i>Penaeus mondon</i>				32.0–99.0	68.0–186.0	0.21–49.0	1.68–54.0	Patimah & Dalhal (1993)
*FAO					30	40	0.5	0.5	FAO (1983)
*EC							0.05–0.10	0.2–0.4	EC (2001)

investigation, the examined shrimps were not associated with vulnerable metal contents in their muscle and were safe within the limits for human consumption although Cd enrichment in the shrimp from the food web of the southwest coast of India is a matter of concern.

Conclusion

The origins of locally elevated metal levels in the shrimps along the southwest Indian coast likely reflect the associated food chain bioaccumulation of metals from the habitat of the estuarine and shelf waters. Differential inter-specific accumulation strategies of metals are evident from the bioaccumulation factors in the shrimps with a maximum in the species *P. mondon* and a minimum in *P. stylifera*, indicating that shrimps could serve as an indicator of metal pollution. This study provides baseline data for trace metals in marine shrimps along the southwest coast of India for assessing future change, if any, in this environment.

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