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Short Communication

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Heavy metal distribution in mangrove sediment cores from selected sites along western coast of India

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Abstract: Sediment cores were collected from four different mangrove areas of northern Kerala and southern Karnataka, western coast of India. The cores were analysed for the concentration of five heavy metals (Pb, Ni, Zn, Cu Fe) using Atomic Absorption Spectrometry. The levels of heavy metals in the present study from all the four sediment cores were in the order Fe > Pb > Zn > Ni > Cu and the mean concentrations of each elements in different cores were comparable. According to Sediment Quality Guidelines (SQG), the mangrove sediments analysed here were moderately contaminated with Ni and heavily contaminated with Pb. The increased concentration of Ni and Pb in the sediments might be due to their atmospheric deposition or water discharge from different far away sources since the areas selected for study were not disturbed by direct anthropogenic impacts. Elevated levels of Fe which is considered to be a common phenomenon in mangrove sediments have also been found in the present study. Heavy metal levels in sediments showed statistically significant correlations with pH, calcium carbonate and organic matter. This suggests the influence of physico-chemical parameters on the adsorption, deposition and persistence of heavy metals in mangrove sediments. The heavy metal concentration and the pollution status of the mangroves of west coast, especially the areas selected in this work are less studied before. Hence the data provide from the present baseline study would be further helpful in remediation and management of mangrove ecosystem.

Keywords: Conservation, correlation, heavy metals, mangroves, physico-chemical parameters, pollution, sediment cores.

Following industrial revolution, heavy metals are being continuously introduced to estuarine and coastal ecosystem. The potential sources of heavy metal inputs to the ecosystems include river discharge, sewage runoff from land, industrial effluents and atmospheric deposition. Natural deposition from erosion of rocks, wind-blown dust, volcanic activity and forest fires also contribute to the heavy metal influx (Gopinath et al. 2010). Toxicity of heavy metals to man and other organisms and their accumulation in the food chain of the ecosystem is of serious concern. Hence, study on concentration of metals in water and sediments have gained interest in recent years.

Sediments are considered to be the most sensitive indicators of contamination in marine environment. They carry historical record of the temporal changes that occurred over a period of time (Sundararajan & Natesan 2010). The concentration of metals in sediments is considered to reflect the mineralogy, origin and texture of the sediments. Details on the sedimentary characteristics are provided by the behaviour and stability of heavy metals in the sedimentary column (Gregory et al. 2002). Mangroves, which develop in intertidal coastal zones, are constantly influenced by terrestrial, fresh water and marine environments. High

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accretion rates and stabilized vegetative nature make this ecosystem an archive of environmental changes and pollution (Daoust et al. 1996). The dynamics of mangrove sediments are regulated by internal and external factors. Internal factors include flocculation, dissolution and mixing *etc* while external factors include river input, agricultural runoff, and pollution. Mangrove sediments due to their inherent physical and chemical properties can accumulate and concentrate metals which are discharged to the marine environment (Defew et al. 2005). Thus, they are capable of retaining water-borne heavy metals and act as sink for heavy metals (Tam & Yao 1998; Kamaruzzaman et al. 2011).

Mangrove ecosystems are facing destruction at an alarming rate all over the world especially in India. A better understanding about the pollution status of these ecosystems is of great importance for protection and management. Analysis of sediment cores is found to be very useful in understanding the chronological history of heavy metal status and associated factors affecting it. It helps in analysing whether the buried contaminants are migrating, accumulating or degenerating (Al-Masri 2002; Harikumar et al. 2009). Research on core sediments have been conducted around the world as they help in establishing the effect of natural and anthropogenic processes on depositional systems like mangroves (Mounier et al. 2001; Machado et al. 2002). Studies have also been done on Indian mangrove sediments over past few years but the main focus were on mangroves of East coast of India (Sarkar et al. 2002; Alagarsamy 2006; Saha et al. 2006; Janaki-Raman et al. 2007). Hence, heavy metal sediment profile data of mangroves of Western coast of India are scanty (Ramanathan et al. 1997; Fernandes et al. 2011; Fernandes & Nayak 2012). The concentration of heavy metals in the deposited sediments is believed to serve as a measure of health status for mangrove ecosystem. The present study was therefore conducted (1) to establish vertical depth profile of concentration level and distribution patterns of five selected heavy metals (Pb, Ni, Zn, Cu and Fe) in the sediment cores collected from different mangrove areas of Northern Kerala and Southern Karnataka, Western coast of India, (2) to analyse the relationships between major physico-chemical parameters and heavy metal distribution in the sediment cores. These heavy metals were particularly selected due to their significant impacts on mangrove health and pollution status of the ecosystem. The presence of these metals in high amounts suggests the input of metals from faraway places through various water bodies in to the mangrove sediments.

Materials and Methods

Sediment cores were collected from mangroves of Chitrapu ($13^0$04.606'N & $74^0$46'49.830'E) of Karnataka, Kumbla ($12^0$35.995'N & $74^0$56.255'E), Kanhangad ($12^0$20.357'N & $75^0$03.909'E) and Manjeshwar ($12^0$42.695'N & $74^0$53.235'E) of Kerala (Fig. 1). Cores collected from Chithrapu, Kumbla, Kanhangad and Manjeshwar were of depth 98cm, 118cm, 83cm and 63cm respectively. Parallel sediment cores were taken from each of these sites during periods of low tides. The cores were brought to lab, cut and sub - sampled by slicing at every 2.5cm intervals. The sediment subsamples from identical depths of replicate cores were pooled together and homogenized for further procedures. The subsamples at every 5cm depth intervals were taken for heavy metal analyses of Chithrapu and Kumbla cores as they were longer cores. A depth interval of 12.5cm was chosen for Kanhangad and Manjeshwar cores as they were comparatively short length cores. The depth intervals were selected and standardised based on trial and error methods with different subsamples to get the most appropriate results.

Preweighed sediment samples were oven dried, pulverized and subjected to acid digestion according to established protocols (APHA 1995; Thomas & Fernandez 1997) with suitable modifications. Digested samples were filtered using Whatman No. 42 filter paper and made up to 50ml using double distilled water for further analysis. Along with reagent blanks and suitable standards, the digested samples were analysed for five heavy metals using Flame Atomic Absorption Spectrophotometry (GBC PAL 3000 Autosampler, Australia). pH of the sediment samples down the core were measured in supernatant suspension of a 1:5 soil liquid mixture potentiometrically using pH meter (Trivedi & Goel 1986). Modified Walkley Black method (Trivedi & Goel 1986) was used for calculating the percentage organic matter present in the sediment samples down the core. Estimation of Calcium Carbonate was done by Acid soluble weight loss method (Campillo et al. 1992) and the percentage was calculated. Salinity of the core samples was measured using salinity meter following Trivedi & Goel (1986) and was expressed in per mille (‰). Statistical analysis was performed to understand the correlation between the distribution of heavy metals and physico-chemical parameters within the core sediments. SPSS Version 20 was the software used for analyses and Pearson correlation (two-tailed *p*-value) between the parameters was studied.

Figure 1. Sites of collection of mangrove sediment cores (a) outline of Western coast of India showing study sites (Black dots indicate area of collection). Google earth images showing (b) Chithrapu (c) Kumbla (d) Kanhangad (e) Manjeshwar (White arrows show sites of collection). Scale bar = 1km

Results

The maximum, minimum and mean concentrations of heavy metals (expressed in mg/kg) along the length of mangrove sediment cores from all the four study sites were summarized in Table 1. Detailed depth profile graphs of heavy metals in sediment cores from each sampling sites were constructed (Figs. 2–5) which provided an elaborate picture on the pattern of heavy metal distribution in sediments over a period of time. The concentration of heavy metals analyzed in the present study from all the four sediment cores were in the order Fe > Pb > Zn > Ni > Cu. The mean concentrations of each of the five heavy metals in sediment cores under study were almost comparable. Cores from mangroves of Kumbla had high mean concentrations of Ni and Zn (53.70±78.86, 86.80±107.30, in mg/kg). The cores from Manjeshwar showed highest mean concentration of Cu (12.10±3.25 mg/kg) while those from Kanhangad showed highest mean concentration of Pb (167.39±81.14 mg/kg).

Pearson correlation analyses of physico-chemical parameters and heavy metals within the cores were studied. In our study, Chithrapu cores showed statistically significant positive correlation between

Table 1. Table comprising the mean distribution of five heavy metals in mangrove sediment cores from (A) Chithrapu (B) Kumbla (C) Manjeshwar (D) Kanhangad (Max - Maximum concentration, Min - Minimum concentration, Mean - Mean concentration, S.D - Standard deviation from mean concentration)

	Pb (mg/ kg)	Ni (mg/ kg)	Zn (mg/ kg)	Cu (mg/ kg)	Fe (mg/ kg)	Pb (mg/ kg)
A	Max	181.75	34.75	175.70	17.75	12087.50
	Min	75.50	2.00	33.25	0.00	870.00
	Mean	142.38	15.20	63.76	5.46	5273.88
	S.D	24.813	9.897	35.568	4.349	4310.99
B	Max	187.50	333.25	429.50	28.00	73075.00
	Min	89.25	4.00	14.25	2.25	182.50
	Mean	143.16	53.70	86.84	10.59	29823.19
	S.D	20.104	78.864	107.315	6.347	24511.69
C	Max	150.75	41.00	104.5	17.75	35462.00
	Min	97.50	26.25	64.50	10.00	6862.50
	Mean	120.45	33.55	84.05	12.10	20642.40
	S.D	22.345	5.909	15.741	3.253	13930.33
D	Max	325.5	34.75	111.00	27.75	19737.00
	Min	97.50	0.00	40.75	4.00	6845.00
	Mean	167.39	20.39	64.36	10.14	11277.29
	S.D	81.142	13.68	24.918	8.114	4248.244

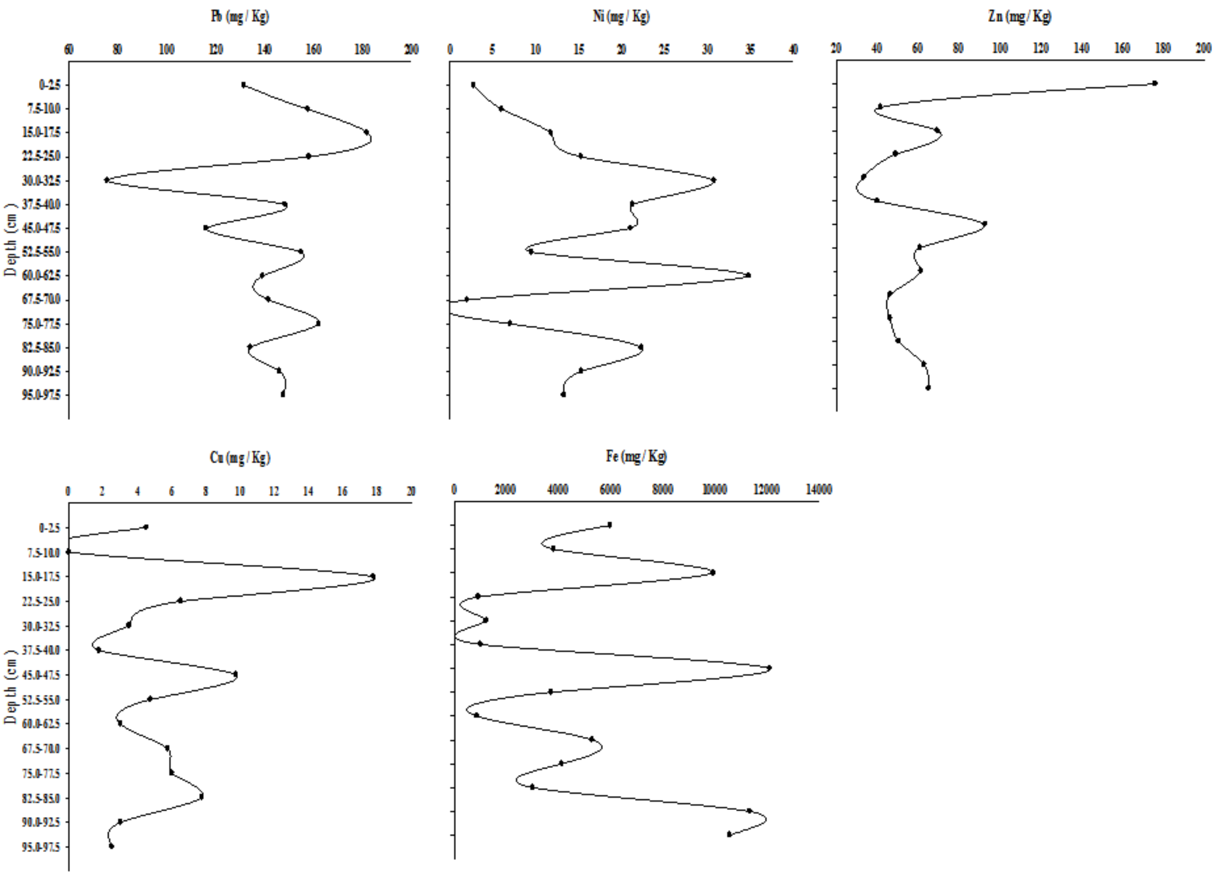

Figure 2. Down core depth profile of heavy metals in mangrove core sediments of Chithrapu

Calcium carbonate (%) and Zn concentration (r = 0.570; P > 0.05). Kumbla cores showed statistically significant positive correlation between Pb concentration with pH (r = 0.511; P > 0.05) and Salinity (r = 0.577; P > 0.05), and also Zn with organic matter (r = 0.643; P > 0.01). Correlation statistics of Chithrapu and Kumbla cores were summarized in Table 2. No significant correlations were seen between the heavy metal concentrations and physico-chemical parameters in Kanhangad and Manjeshwar cores. The pattern of heavy metal distribution varied between consecutive depths in the cores and these variations did not show any kind of significant relationships with the depth.

Discussion

The approximate age of the mangrove sediment cores was estimated using Lead-210 analyses and was found to be around 40 years old. Hence, the vertical depth profile data on heavy metal levels gives us an account of the ecological behaviour and pollution status of the mangroves over a period of time. The distribution of heavy metals in all the four cores analyzed was similar and their concentrations followed the pattern Fe > Pb > Zn > Ni > Cu. Standard Sediment Quality Guidelines (SQG) have been used around the world to assess the extent of contamination by heavy metals and other pollutants in aquatic ecosystems (Burton 2002; Merchanda et al. 2006; Kumar & Edward 2009). Sediments can be classified as non-polluted, moderately polluted and heavily polluted, based on SQG of USEPA (Harikumar et al. 2009). The mean concentrations of heavy metals in the cores of present study were compared with Sediment Quality Guidelines and other similar studies around the world (Table 3). The mean concentration ranges of Cu and Zn in the sediment cores studied here were well below the limits of tolerance and hence the sediments were not polluted with them. The mean concentrations of Ni were slightly higher than the accepted levels in the sediments and they were in the moderately polluted status. Nickel is a trace metal which is needed at very low levels and causes dangerous effects if the tolerance limits are exceeded. The common sources of Ni are steel and other metal products, power plants and trash incinerators, that get accumulated in plants and animals

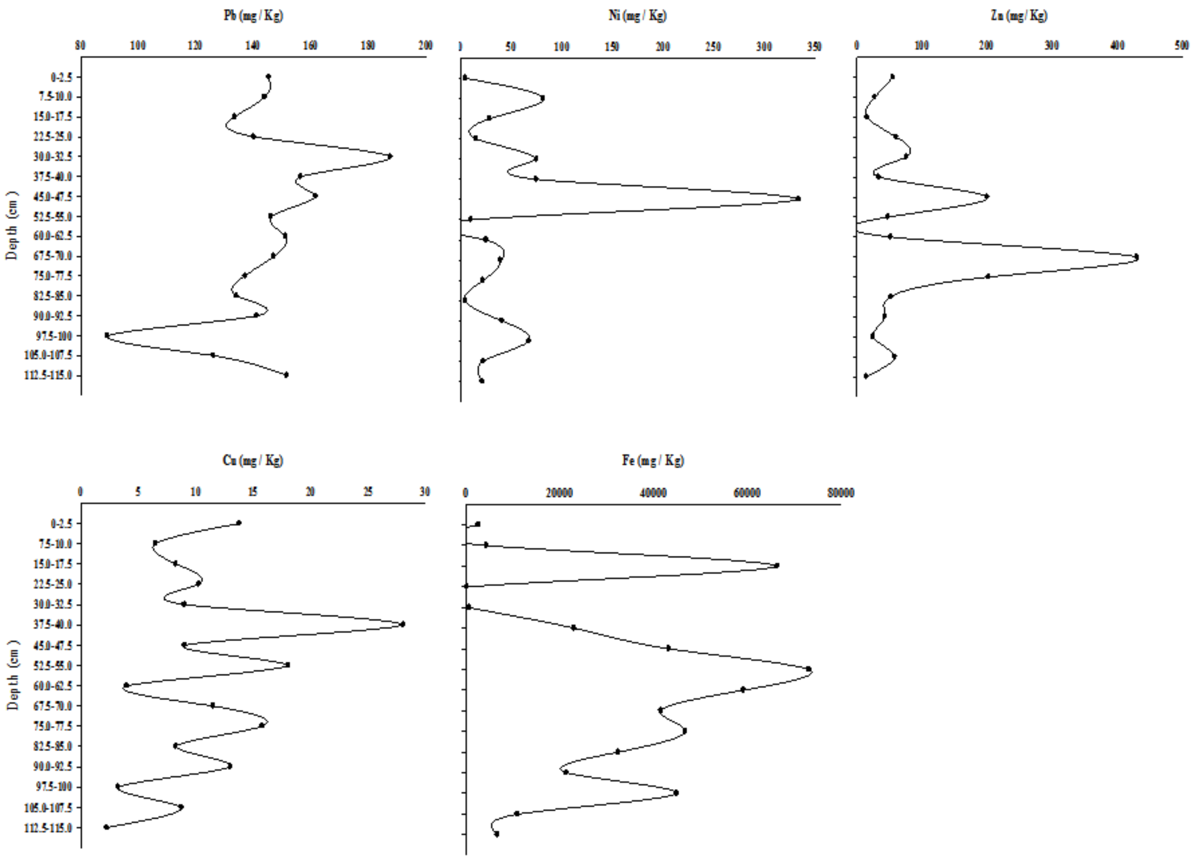

Figure 3. Down core depth profile of heavy metals in mangrove core sediments of Kumbla

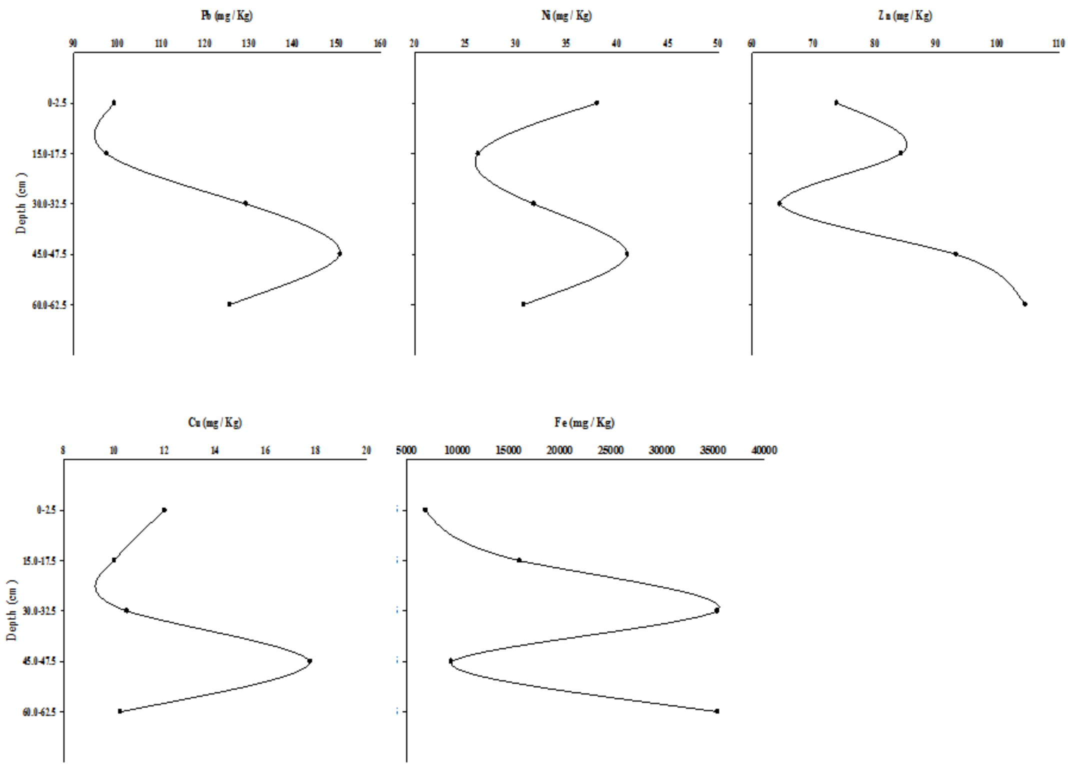

Figure 4. Down core depth profile of heavy metals in mangrove core sediments of Manjeshwar

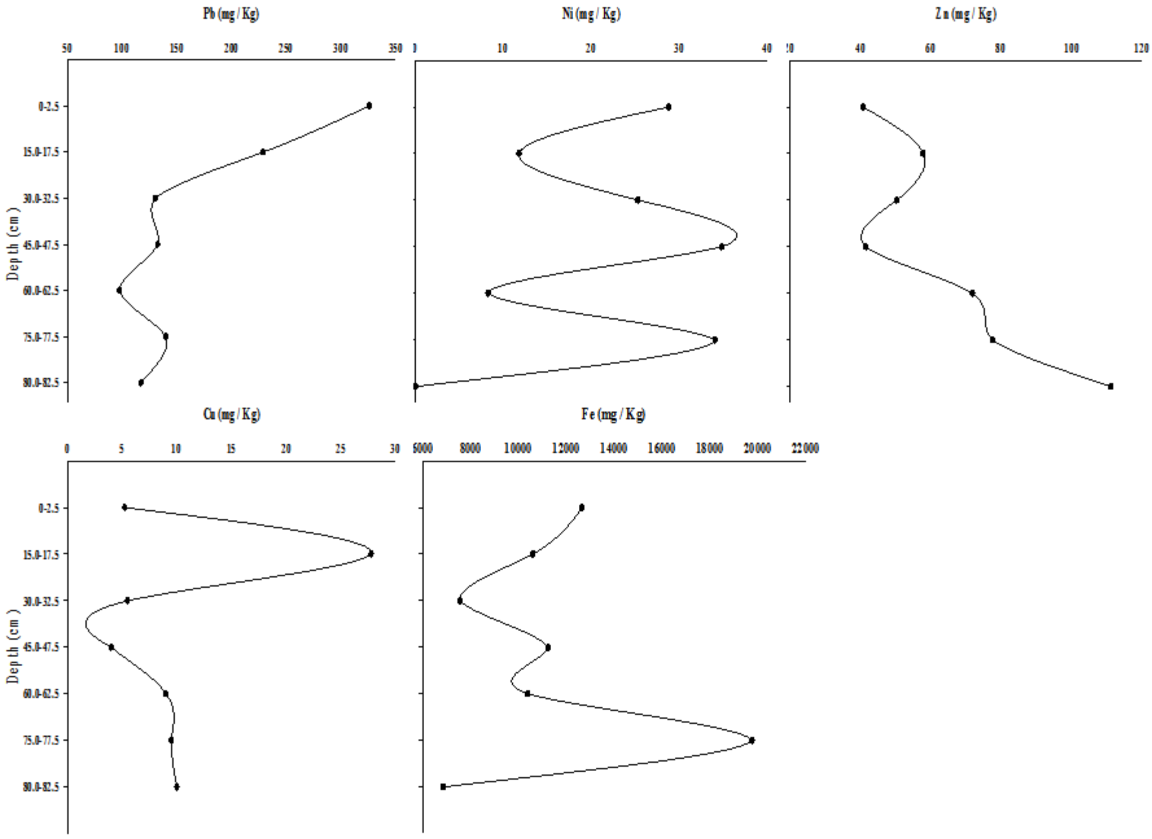

Figure 5. Down core depth profile of heavy metals in mangrove core sediments of Kanhangad

(Pacyna et al. 1991). The increased concentration of Ni in the sediments of the cores studied here might be due to its atmospheric deposition or water discharge from far away sources. Ni of terrigenous origin also could contribute to its elevation in sediments of mangroves. Chithrapu and Kanhangad core sediments found to have lower Ni concentrations. The Ni concentration of Chithrapu sediments was in no pollution range while that of Kanhangad was on the margin of moderate pollution. An increase in concentrations of metals above the normal range was observed at certain depths in the cores. Ni concentration was enormously high (333.25mg/kg) at depth 45–47.5 cm while Zn concentrations also showed higher values at depths 45–47.5 cm (200.50mg/kg), 67.5–70 cm (429.50mg/kg) and 75–77.5 cm (202.00mg/kg) sediment cores of Kumbla. However, the mean concentrations of these metals in the cores were comparable with the normal concentration ranges. This increase in the metal concentrations at particular depths in the core might be due to the high influx and deposition of that particular metal during those periods.

Very high concentration of Pb was seen in all the cores analyzed which was much higher than the SQG levels. Hence, the sediments were considered to be heavily polluted with Pb. The common sources of Pb are leaded petroleum, paints, brackish aquaculture and construction activities. Even though the areas selected for study were not affected much by industrial or such kind of human activities, the atmospheric deposition and water discharge of Pb from other areas are possible. This might have resulted in the higher concentration of Pb in the core sediments. Precipitation of iron as iron sulphides is considered to be a common phenomenon in all mangrove ecosystems (Thomas & Fernandez 1997) and increased concentration of Fe has been reported in sediments from mangroves in studies all over the world (Nair et al. 1987; Hirner et al. 1990) Alongi (2010) have mentioned the beneficiary effect of high sediment Fe concentration for the growth of mangrove plants. Thus, tolerance limit of Fe especially in mangrove sediments is very high compared to other metals due to their high inherent iron levels. The increased concentrations of Ni (moderately polluted), Pb (heavily polluted) and Fe observed in the mangrove sediments of the present study area are of concern. The high metal values in the mangrove sediments could be due to natural or

Table 2. Pearson correlation analyses of physico - chemical parameters and heavy metals within core sediments of (A) Chithrapu (B) Kumbla. Statistically significant correlations of interest are shown in bold fonts. {*Correlation is significant at the 0.05 level (2-tailed); **Correlation is significant at the 0.01 level (2-tailed)}.

A	pH	Organic Matter	Calcium carbonate	Salinity	Pb	Ni	Zn	Cu	Fe
pH	1								
Organic Matter	.000	1							
Calcium carbonate	.370	.049	1						
Salinity	**.614***	-.148	**.761****	1					
Pb	.511*	.063	.468	.577*	1				
Ni	.333	.380	.240	.091	.270	1			
Zn	-.001	**.643****	.021	-.130	.168	.250	1		
Cu	.486	-.009	.243	.280	.246	-.042	.114	1	
Fe	-.115	.412	-.127	.020	.251	.067	.192	.071	1

B	pH	Organic Matter	Calcium carbonate	Salinity	Pb	Ni	Zn	Cu	Fe
pH	1								
Organic Matter	-.290	1							
Calcium carbonate	.338	-.614*	1						
Salinity	.057	-.521	.644*	1					
Pb	-.046	.024	.033	.195	1				
Ni	.181	.031	-.323	-.278	-.517	1			
Zn	.120	-.007	**.570***	.337	-.059	-.308	1		
Cu	.054	-.050	-.125	-.161	.279	.069	.149	1	
Fe	.222	-.204	.411	.249	.145	.294	.366	.384	1

Table 3. Mean concentrations of heavy metals in the mangrove sediment cores studied and their comparison with the SQG. Levels of Fe were not available. Numbers in bold indicate heavily polluted concentrations in the cores.

Metals (mg/ kg)	SQG Not polluted	SQG Moderate polluted	SQG Heavily polluted	Mean concentrations in cores (mg/ kg)			
				Chithrapu	Kumbla	Manjeshwar	Kanhangad
Cu	<25	25-50	>50	5.46	10.59	12.10	10.14
Ni	<23	20-50	>50	15.20	**53.70**	33.55	20.39
Pb	<40	40-60	>60	**142.38**	**143.16**	**120.45**	**167.39**
Zn	<90	90-200	>200	63.76	86.84	84.05	64.36
Fe	-	-	-	5273.88	29823.19	20642.40	11277.29

anthropological impacts. Exact causes for the elevation in the levels of these heavy metals need to be identified as they are highly toxic to both flora and fauna of the ecosystem.

Many factors influence the concentration of heavy metal in sediments. Physico-chemical parameters like organic matter, pH, carbonate content and cation exchange capacity, etc., are some among them. Heavy metal levels in sediments in the present study showed some statistically significant correlations with pH, calcium carbonate and organic matter. pH is believed to have role in the mobility and distribution of heavy metals in sediments by acting on its adsorption properties. Studies have shown that with an increase in pH from 4–7; the adsorption capacity of Pb by soil is also increased (Odat & Alshammari 2011). pH of soil also regulates the action of carbonates on heavy metals (Adriano 1986). This suggests a relationship of heavy metal concentrations in sediments with pH and carbonate content. These statistically significant correlations in Chithrapu and Kumbla cores substantiate the concept of heavy metal uptake under the influence of physico- chemical parameters like pH, carbonates and organic matter. There was no significant correlation seen between heavy metal levels and physico-chemical parameters in Manjeshwar and Kanhangad cores. The changes in the physico-chemical parameters like pH, organic matter and salinity *etc* that the sediments undergo after accretion (post depositional changes) can disturb the pattern of distribution of heavy metals in the cores. The mangroves studied showed the presence of burrowing animals like fiddler crabs and some annelids, which are involved in the sediment mixing by the process of bioturbation. Apart from this, disturbance like Tsunamis, heavy showers and flooding of rivers, can be considered as possible ecological perturbations (Kamaruzzaman 2011). Any or all of these factors can be the reason for lack of similar pattern of heavy metal accumulation with respect to the depth and parameters studied between the cores.

The mangrove ecosystems which were selected for sediment coring constituted of healthy plants and were devoid of direct anthropological activities. Still there are chances of heavy metal transport from far away regions through various water bodies in to sea water and there by absorption in to these mangrove sediments. The present study showed elevated concentrations of Ni and Pb in the mangrove sediments. According to SQG, the mangroves sediments analysed here were found to be moderately contaminiated with Ni and heavily contaminated with Pb. The present work on heavy metal status of mangrove sediments can be considered as a baseline study since there is lack of previous established data on these aspects from the sites selected. Tracing the actual sources of these metals is important in understanding the pollution status and also in remediation purpose. This study would also help in gaining insight in to the ecological history of mangroves which further can be employed in conservation and management processes.

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