Preliminary Survey on the Heavy Metal Pollution in Punnakayal Estury of Tuticorin Coast, Tamil Nadu

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Abstract: In Punnakayal estuary preliminary heavy metal analysis was done in water sediment and mangrove leaf samples of two stations for one year. The metal concentrations of the samples were high during monsoon and were higher in sediments.

Key words: Punnakayal • Gulf of Mannar • Mangrove • Pollution • Heavy metal

INTRODUCTION

specialized mangroves ecosystems developed along the estuarine areas of tropical and subtropical regions of the world, mainly in the intertidal zone. Wide varieties of plant and animal species with special adaptations are suited to this ecosystem. Mangrove ecosystem is considered as the most productive and providing significant functions in the coastal zones as buffer against erosion, storm surge and tsunamis. The carbon fixed in mangroves is highly important in the coastal food webs and the litter from mangroves and the subsequent formation of detritus and its tidal export have also profound effect on promoting biodiversity richness. The scientific report on India's mangrove forest was published during early 1800s in which the flora of the Indian mangrove ecosystem was described [1]. The total area of India's mangrove forest was estimated in 1980s was about 6740 km² covering 7% of the world mangroves and 8% of the Indian coastline. The human impact on the mangrove forest started in India during the early 19th century [2] and due to this the total mangrove area decreased to 4474 km² in a decade later activities [3]. Increasing human population growth, expending aquaculture practices, habitat destruction, deforestation, industrial growth and mounting toxic pollution are drastically affecting the mangrove ecosystems of Tamil Nadu, South India.

The mangrove ecosystem harbors flora and fauna, although possessing enormous ecological and commercial important. But nowadays the mangrove ecosystem also affected by effluent discharges, urban and agricultural runoff and solid waste dumping due to their proximity to urban development. Among the main anthropogenic impacts in mangrove ecosystems from these sources are heavy metals, due to their affinity and immobilization within anaerobic sediments [4]. Metals may also be transported to estuarine waters when accumulated by mangroves and concentrated in exported leaf detritus, which is an important food source for higher organisms in estuarine food chains [5]. Heavy metals are the most serious pollutants within the natural environment due to their toxicity, persistence and bioaccumulation problems [6, 7]. Mangrove mud's have an extraordinary capacity to accumulate materials discharged to the near shore marine environment [4]. Mangrove sediments are anaerobic and reduced, as well as being rich in sulphide and organic matter therefore favours the retention of water-borne heavy metals [8, 9] and the subsequent oxidation of sulphides between tides allows metal mobilization and bioavailability [10]. Concentrations of heavy metals in sediments usually exceed those of the overlying water by 3-5 orders of magnitude [11] and with such high concentrations, the bioavailability of even a minute fraction of the total sediment metal content assumes considerable importance with respect to bioaccumulation within both animal and plant species living in the mangrove environment. Since heavy metals cannot be degraded biologically, they are transferred and concentrated into plant tissues from soils and pose long-term damaging effects on plants. High inputs of heavy metals from untreated domestic sewage, storm water road run-off and diffuse inputs from shipping and agricultural activities [12].

Punnakayl estuary is the only estuary in Tuticorin coast of Gulf of mannar. Tambraparani river rises in Agasthiyamalai of the western ghats and flows through Srivaikundam and Thiruchendur taluks and joins with the sea at Punnakayl. The mangrove tree Avicennia sp. is dominant in this ecosystem and they are short due to high saline condition and lack of fresh water inflow except during the monsoon [13]. Around 80 families depend this mangrove ecosystem for their livelihood. About 50 men are involved in fishing activities in the mangrove area and about 30 women are involved in collecting firewood from the mangrove ecosystem so the mangrove areas are degraded due to this activities and the destruction activity is estimated to be at a higher rate [14]. The aim of this study was to determine the current levels of eight commonly polluting metals such as manganese (Mn). copper (Cu), zinc (Zn), nickel (Ni), lead (Pb), iron (Fe), mercury (Hg), chromium (Cr) and cadmium (Cd); and to determine the degree of spatial variation of metals in waters, sediments and mangrove leafs within the study area.

MATERIALS AND METHODS

Area of Investigation: The survey was conducted from February 2008 to January 2009 at punnakayal estuary. Two stations were selected for the collection of samples for metal analysis. Station 1 (08°38'119"N 78°07'110"E) is the river side of punnakayal estuary and Avicennia sp. is dominant in this site and Rhizophora sp. were also planted in this area by the forest department. High level fishing activity is going on in this station.

Station 2 (08°38'163"N 78°07'362"E) is the river mouth of punnakayal estuary. It is the road way of fishing boat and fishing activity also high in this station. Tidal level also high and Avicennia sp. is dominant in this station.

Collection and Analysis of the Samples: The water, sediment and mangrove leaf samples were collected monthly from both the stations. Water samples were collected in sterile acid washed plastic bottle and labeled and stored in clean acid washed container until transportation to the laboratory. The sediment samples were collected using a grab. The collected samples were stored in clean acid washed plastic containers until transportation to the laboratory. Each sample were wet sieved through a 1 mm bronze mesh with double distilled water and the samples were collected in acid washed glass jar and labeled. Sediment samples were dried in an oven at 60 ± 50 °C for 24 hours to eliminate the water content and were homogenized by mortal and pestle and stored in acid washed plastic bags for metal analysis.

Twenty leaves were collected from Avicennia sp. without destroying the plants. Leaves were collected from 1m tall with a girth at a breast height of greater than 2.5 cm and that were of similar health condition. Leaves were sub sampled (n= 3), washed in distilled water, oven dried at 60°C for 24 hours and homogenized 14. The concentration of metal in water samples were estimated by the method described by Danielsson [15, 16]. The water samples were digested with 20 ml of 5:1 mixture of concentrated nitric acid and perchloric acid in triplicate [17]. The digestion process was continued till the brown fumes of nitrogen peroxide cease to appear.



Fig. 1: Map showing the sampling stations

The process was continued until the sample evaporated to near dryness. Then it was cooled, filtered through Whatman no: 1 paper and made up to 25 ml with deionised double distilled crystal clear water. The samples were transferred to polythene bottle and analyzed for various metals using an Atomic Absorption Spectroscopy (AAS; ELICO-SD 164, India).

The powdered sediment samples were sieved through a $102~\mu$ mess size sieve and 1.0~gm of sieved sediment samples were taken for digestion. 9 ml of concentrated nitric acid and 1 ml perchloric acid were added and heated on a hot plate until the solution become clear. Then the samples were filtered through Whatmann no.1 filter paper and diluted with distilled water and made up to 25~ml in a volumetric flask. The made up samples were stored in prewashed polythene bottle for metal analysis using an Atomic Absorption Spectroscopy (AAS; ELICO-SD 164, India).

250 mg of oven dried leaf tissue was digested in concentrated nitric acid (100° C) and hydrogen peroxide (65°C) following the method of Krishnamurthy [18]. Samples were made up to 40 ml, filtered with 0.45 μm and metal analysis was carried out using air / acetylene Atomic Atomic Absorption Spectroscopy (AAS; ELICO-SD 164, India) (or) 0.3 gm of oven dried leaf tissue was digested using concentrated nitric acid and 0.5 ml of hydrogen per oxide on a hot plate. After the digestion, sample was filtered with 0.45 μm filter paper and filtrate was made up to 25 ml with double distilled water [18]. Trace metal extraction of soil and sediments by nitric acid-hydrogen peroxide.

Blanks were also prepared by the addition of same quantity of reagents without the samples and digested and made up to 25 ml for water and sediment, 40 ml for leaf samples.

RESULTS

Heavy Metals in Water Sample (mg/L): The heavy metal concentrations in the water samples of station 1 and 2 were presented in table 1. Heavy metal concentrations were slightly varied in the water samples of both the stations. Zinc concentration was mostly varied between the stations in October, December, January and March. High concentration of lead was found in August, September and October in station 2 where as it was very high in station 1 during November, to January. The iron concentration was high in all the months at station 1 than station 2. Chromium was below detectable range during the month of February in both the stations and during October and November it was high in station 1 than station 2. Mercury was shown below detectable range in both the stations in all the months. Nickel was below detectable level in February and April to June in both the stations and it was high in station 1 than station 2 in the rest of the months. Manganese was high in station 2 than station 1 and cadmium was in below detectable range in May month of both the stations.

Heavy Metals in Sediment Sample (mg/gm dry weight): The heavy metal concentrations in the sediment samples of station 1 and 2 were tabulated in table 2.

-	Zn		Pb		Fe		Cr		Hg	Hg		Ni		Mn		Cd	
Months	S 1	S 2	S 1	S 2	S 1	S 2	S 1	S2	S 1	S 2	S 1	S 2	S 1	S 2	S 1	S 2	
Feb. 08	BDR	BDR	0.051	0.048	0.71	0.45	BDR	BDR	BDR	BDR	BDR	BDR	0.270	0.140	0.05	0.03	
Mar. 08	0.38	BDR	0.030	0.026	0.99	0.81	0.150	0.130	BDR	BDR	0.18	0.08	0.180	0.150	0.05	0.08	
Apr. 08	BDR	BDR	0.020	0.020	0.78	0.75	0.080	0.080	BDR	BDR	BDR	BDR	0.150	0.170	0.03	0.01	
May. 08	BDR	BDR	0.020	0.010	0.11	0.10	0.061	0.040	BDR	BDR	BDR	BDR	0.180	0.180	BDR	BDR	
Jun. 08	0.25	0.25	0.033	0.030	0.61	0.65	0.068	0.064	BDR	BDR	BDR	BDR	BDR	BDR	0.02	0.01	
July. 08	0.31	0.21	0.035	0.011	1.80	0.80	0.066	0.058	BDR	BDR	0.18	0.10	0.173	0.183	0.03	0.03	
Aug. 08	0.25	0.28	0.051	0.070	1.50	0.35	0.088	0.071	BDR	BDR	0.18	0.08	0.200	0.250	0.04	0.04	
Sep. 08	0.28	0.25	0.050	0.065	1.60	0.38	0.100	0.091	BDR	BDR	0.21	0.12	0.130	0.180	0.05	0.04	
Oct. 08	0.38	0.28	0.070	0.080	2.88	3.18	0.160	0.110	BDR	BDR	0.13	0.13	0.130	0.180	0.03	0.02	
Nov. 08	0.45	0.41	0.081	0.068	2.55	2.48	0.230	0.180	BDR	BDR	0.23	0.18	0.180	0.190	0.01	0.01	
Dec. 08	0.31	0.18	0.058	0.033	3.31	1.23	0.110	0.080	BDR	BDR	0.07	0.09	0.150	0.180	0.03	0.02	
Jan. 09	0.35	0.21	0.038	0.015	2.81	0.80	0.080	0.100	BDR	BDR	0.01	0.11	0.210	0.150	0.04	0.01	

Table 2: Heavy metals in the sediment (mg/gm Dry weight) of Punnakayal station 1 and 2

	Zn		Pb		Fe		Cr		Hg		Ni		Mn		Cd	
Months	S 1	S 2	S 1	S 2	S 1	S 2	S 1	S2	S 1	S 2	S 1	S 2	S 1	S 2	S 1	S 2
Feb. 08	BDR	BDR	0.058	0.056	0.66	0.66	BDR	BDR	BDR	BDR	BDR	BDR	0.27	0.300	0.01	0.02
Mar. 08	BDR	BDR	0.038	0.033	1.58	1.65	0.190	0.100	BDR	BDR	0.192	0.186	0.20	0.150	BDR	BDR
Apr. 08	0.23	BDR	0.028	0.028	0.81	0.88	0.100	0.100	BDR	BDR	BDR	BDR	0.18	0.180	0.02	0.02
May. 08	0.60	BDR	0.020	0.020	0.53	0.35	0.073	0.060	BDR	BDR	BDR	BDR	0.21	0.150	0.01	0.01
Jun. 08	0.21	0.06	0.045	0.030	0.96	0.81	0.083	0.060	BDR	BDR	0.025	BDR	0.09	BDR	0.01	BDR
July. 08	0.22	0.08	0.041	0.028	2.00	1.00	0.085	0.080	BDR	BDR	0.180	0.080	0.59	0.193	0.03	0.03
Aug. 08	0.35	0.25	0.088	0.068	1.80	0.61	0.100	0.080	BDR	BDR	0.220	0.091	0.33	0.130	0.05	0.04
Sep. 08	0.19	0.18	0.071	0.070	1.80	0.68	0.130	0.070	BDR	BDR	0.230	0.138	0.31	0.190	0.05	0.04
Oct. 08	0.31	0.25	0.096	0.078	3.10	3.15	0.180	0.180	BDR	BDR	0.280	0.140	0.33	0.198	0.06	0.05
Nov. 08	0.43	0.31	0.088	0.088	3.50	3.00	0.260	0.180	BDR	BDR	0.300	0.183	0.35	0.230	0.08	0.05
Dec. 08	0.21	0.18	0.060	0.045	2.80	2.10	0.130	0.110	BDR	BDR	0.100	0.070	0.20	0.150	0.03	0.02
Jan. 09	0.28	0.21	0.040	0.018	2.00	0.81	0.100	0.110	BDR	BDR	0.100	0.051	0.21	0.110	0.02	0.01

Table 3: Heavy metals in the mangrove leaves (mg/gm Dry weight) of Punnakayal station 1 and 2

	Zn		Pb		Fe		Cr		Hg		Ni		Mn		Cd	
Months	S 1	S 2	S 1	S 2	S 1	S 2	S 1	S2	S 1	S 2	S 1	S 2	S 1	S 2	S 1	S 2
Feb. 08	BDR	BDR	0.041	0.033	0.38	0.33	BDR	BDR	BDR	BDR	BDR	BDR	0.110	0.180	0.08	0.01
Mar. 08	BDR	BDR	0.023	0.018	1.18	0.28	0.060	0.01	BDR	BDR	0.050	BDR	0.090	0.098	BDR	BDR
Apr. 08	0.18	BDR	BDR	BDR	0.58	0.28	0.060	0.04	BDR	BDR	BDR	BDR	0.085	0.078	BDR	0.01
May. 08	0.04	BDR	0.020	BDR	0.44	0.11	0.020	BDR	BDR	BDR	BDR	BDR	0.078	0.043	BDR	BDR
Jun. 08	0.03	0.06	0.021	0.010	0.50	0.18	0.030	0.03	BDR	BDR	0.018	BDR	0.060	0.040	BDR	BDR
July. 08	0.08	0.08	0.023	0.018	1.35	0.88	0.080	0.04	BDR	BDR	BDR	BDR	0.380	0.155	0.03	0.01
Aug. 08	0.33	0.05	0.050	0.038	1.11	0.51	0.065	0.04	BDR	BDR	0.080	0.068	0.210	0.210	0.01	0.03
Sep. 08	0.10	0.10	0.068	0.041	1.00	0.78	0.080	0.06	BDR	BDR	0.090	0.087	0.110	0.210	0.03	0.02
Oct. 08	0.48	0.12	0.068	0.049	2.81	2.51	0.100	0.09	BDR	BDR	0.100	0.091	0.100	0.250	0.03	BDR
Nov. 08	0.55	0.18	0.081	0.041	2.73	2.11	0.180	0.08	BDR	BDR	0.130	0.090	0.130	0.290	0.03	BDR
Dec. 08	0.21	0.10	0.058	0.019	2.59	1.11	0.050	0.08	BDR	BDR	0.760	0.030	0.090	0.110	0.01	0.01
Jan. 09	0.21	0.10	0.031	0.013	2.10	0.73	0.060	0.03	BDR	BDR	0.044	0.020	0.088	0.087	0.01	0.01

S1 - Station 1, S2 - Station 2

Heavy metal concentrations were varied between the water and sediment. Zinc concentration was high in all the months at station 1 especially during April to July. Lead, chromium and nickel concentrations were high in station 1 than station 2 in all the months. The iron concentration was very high in station 1 during July and January. Mercury was below detectable range in all the months in both the stations. Manganese was recorded high in station 2 than station1 and it varied more in both the stations but cadmium was not significantly varied in both the stations.

Heavy Metals in Leaf Sample (mg/gm dry weight): The heavy metal concentrations in the mangrove leaves of station 1 and 2 were given in table 2. Heavy metal concentrations were lower in leaves than water and

sediments. The Zinc concentration was high in all the months except June in station 1 and below detectable range was observed at both stations during February and March. Lead concentration was found below detectable range in station 2 during May and it was very high in November and December in station 1. Iron concentration was very high in station 1 during March, August, December and January. Chromium was also high in all the months in station 1 than station 2. Mercury was in below detectable range in all the months of both the stations. Nickel was found below detectable range in both the stations during February, April, May and July months and high concentrations were observed in March, July and December at station 1. Manganese was very high in station 2 during February and September to November. Cadmium was not significantly varied in both the stations.

Table 4: Student't' test analysis of Heavy metals between water and Sediment samples of station 1

Parameters	Df	t Stat	P Value	Remarks
Zn	22	-0.08787	0.930772	p>0.05
pb	22	-1.24987	0.224478	p>0.05
Fe	22	-0.38315	0.705289	p>0.05
Cr	22	-0.77505	0.446558	p>0.05
Ni	22	-0.85928	0.399454	p>0.05
Mn	22	-2.68768	0.013445	P<0.05
Cd	22	0.099504	0.921639	p>0.05

Note: p>0.05-Not significant; p<0.05- Significant

Table 5: Student't' test analysis of Heavy metals between water and Sediments samples of station 2

Parameters	Df	t Stat	P Value	Remarks
Zn	22	0.876557	0.390199	p>0.05
pb	22	-0.70719	0.486876	p>0.05
Fe	22	-0.81263	0.42514	p>0.05
Cr	22	-0.53911	0.595225	p>0.05
Ni	22	-0.15131	0.881106	p>0.05
Mn	21	-0.26664	0.792347	p>0.05
Cd	22	0.103292	0.918667	p>0.05

Note: p>0.05-Not significant; p<0.05- Significant

Table 6: Student't' test analysis of Heavy metals between Sediment and leafs samples of station 1

Parameters	Df	t Stat	P Value	Remarks
Zn	22	0.954369	0.350267	p>0.05
pb	22	1.552146	0.134895	p>0.05
Fe	22	1.028714	0.314792	p>0.05
Cr	22	2.315170	0.030319	P<0.05
Ni	22	0.428925	0.672149	p>0.05
Mn	22	3.264338	0.003550	P<0.05
Cd	22	1.204523	0.241187	p>0.05

Note: p>0.05-Not significant; p<0.05- Significant

Table 7: Student't' test analysis of Heavy metals between Sediment and leaf samples of station 2

Parameters	Df	t Stat	P Value	Remarks
Zn	22	1.626023	0.118185	p>0.05
pb	22	2.767997	0.011221	P<0.05
Fe	22	1.390649	0.178240	p>0.05
Cr	22	3.100027	0.005225	P<0.05
Ni	22	1.961061	0.062652	p>0.05
Mn	22	0.606439	0.550430	p>0.05
Cd	22	2.724414	0.012380	P<0.05

Note: p>0.05-Not significant; p<0.05- Significant

The metal concentrations of station 1 and 2 were statistically analyzed between water and sediment samples and the results were presented in tables 4 and 5.

The student "t" test showed no significant difference (P>0.05) between the metals concentrations of water and sediment samples for all the metals except manganese of Station-1 (Table 4). There is no significant variation between water and sediment samples in station 2 of all the metal concentrations (Table 5).

The sediment and leaf samples were statistically analyzed and the results were shown in Tables 6 and 7. Heavy metal concentration were not significantly (P>0.05) varied between metal concentrations of sediment and leaf samples except chromium and manganese of station 1 (Table 6). In station 2, the heavy metal concentrations were not significantly (p>0.05) varied for zinc, iron, nickel and manganese and significantly (p<0.05) varied for lead, chromium and cadmium (Table 7).

DISCUSSION

The metal concentration was high in sediment than water and leaf of both the stations. Sediments have a mixture of minerals as well as organic compound which is the one of the major factor for absorption of heavy metals [19-22]. Organic matter normally present in sediment than water and it has favorable binding site for heavy metals and humic substance. The organic matter plays a vital role in the sorption of metal on marine sediments due to the presence of charge surfaces [23].

The metals concentration was high in sediment of station 1 because high level of anthropogenic activity such as sewage disposal, human excreta and activities of fishing by trawler boat (such as antifouling paint, oil dropping and garbage waste) were mixed in station 1 and this influences the metal level. Silt and clay nature of sediment was found in station 1 and sediment nature is also one of the important factors for metal accumulation. Fine sediment has much binding site than other sediment. The smaller particulars have a greater specific surface area than the larger ones. Fine particles contain mainly clay minerals, which have more metal binding sites. The silt and clay fractions usually contain higher metal concentrations than the sand fraction [24]. Fine sediment particles were found in both the stations, so all metals were found to be in high levels. This report was supported by [25]. Silt and clay sediment had high amount of metals than sand fraction at Tuticorin harbour and concentration of metals in sediments depends on several factors such as local conditions, particle size and organic matter content [26]. Fine grained sediments (clays, clayey silts) are characterized by higher metal concentration in sediment [27]. Except Manganese, all metals concentration

was high in station 1. This was mainly due to domestic wastes, land and agricultural drainage, boating activities, antifouling paints from boats, oil dropping from boats and fishing activity and this result is sustained by Asoke Kumar [28]. Iron was high where as mercury was in below detectable range in both the stations. The same trend was observed in Bay of Bengal along Tamil Nadu coast [29].

The metals concentrations in both the stations were high in monsoon and low in summer seasons. High concentrations of zinc were observed during monsoon and it was due to the effect of increased land runoffs. Large part of anthropogenic discharge of heavy metals into the environment becomes part of the suspended matter in rivers. This suspended matter can act as a scavenger for heavy metal in water [30]. Similar observations were made in Godavari estuarine sediments [31] and Pondicherry harbour [32]. Relatively higher concentrations of iron during monsoon may be due to the higher inputs of land runoff and influx of metal rich fresh water. The increased particulate matter along with suspended sediment load brought in by the river would also be a possible reason for the abnormal high values during monsoon [32]. High content of iron during monsoon and post-monsoon seasons in Tuticorin coast, which is attributed to the increased land runoffs [33]. In Godavari estuarine sediments had higher concentrations of heavy metals during monsoon season and this may be due to high fresh water interfere [31]. High concentration of manganese was reported during monsoon and postmonsoon seasons in Tuticorin coast [33]. High level cadmium was recorded in monsoon season and minimum in summer season. Similar result was observed in pondicherry harbour [32] and this was attributed to the land runoff and influx of metal rich water. Seasonal variation in the concentrations of chromium registered highest values during the monsoon period followed by those in pre-monsoon and post-monsoon seasons. High concentrations observed during monsoon can be attributed to the land runoff and influx of metal rich fresh water. The increased particulate matter along with suspended sediment load brought in by the river would also be a possible reason for the abnormally higher values during monsoon [32]. Similarly higher concentrations of chromium in sediments was observed in Godavari estuary during monsoon season [31].

Avicennia sp. is dominant in punnakayal mangrove area. They are in stunted growth, may be heavy metal accumulation is the one of the factor. When plants absorbed and accumulated heavy metals, the vessels

became constricted and deposit the unknown substance blocked the vascular system and retarded the water transportation [34]. High heavy metal concentrations from waste water that significantly reduced leaf number and stem basal diameter [35]. Old leaves were yellow and shed off whilst young leaves continued to survive. Toxic heavy metals accumulate in mangrove ecosystems due to urban development and sources of metal contamination range from domestic garbage dumps to agricultural runoff [36]. High level of Cu, Fe, Mn, and Pb concentrations indicating more polluted mangrove ecosystem of Tamil Nadu, South India [14, 37]. Heavy metal concentrations in plant tissues are triggered by metabolic requirements for essential micro nutrients such as Cu and Zn, while the non-essential Pb tends to be toxic to some species [38]. Copper and zinc are essential micro nutrients required in chloroplast reactions, enzyme systems, protein synthesis, growth hormones and carbohydrate metabolism [39].

The preliminary survey reveals that Punnakayal estuarine sediment accumulated more heavy metals than water. This heavy metal pollution also affects the biodiversity of this estuary.

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