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RESEARCH ARTICLE

Physico-Chemical Charecteristics and Distribution of Heavy Metals along the Punnakayal Estuary, SE Coast, India

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ABSTRACT

Punnakayal is a mangrove ecosystem located in the south east coast of India where Thamirabarani River joins the Arabian Sea. Mangroves especially *Avicennia sp.* are abundant and domestic waste, fish waste, human and animal excreta are disposed in this region. The present investigation was carried out to find out the changes in physicochemical including heavy-metal concentration in the Punnakayal estuary of Tuticorin coast. Among the various parameters, salinity and electrical conductivity are low; turbidity was high during monsoon due to the river run off. Nutrients and heavy metal are found to be well within limits of nominal coastal ecosystems.

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INTRODUCTION

Punnakayal is situated in the confluence of a tributary of the perennial River Thamirabarani, 30 Km south of the industrial hub of Tamilnadu, Tuticorin, Southeast coast of India. The coastal ecosystem includes an estuary, mangrove patches and mud flats (Plate 1). The total estimated mangrove area is around 7 sq.km out of which 3 sq.km is denuded and in 1 sq.km restoration has been attempted. The area surrounding this mangrove thicket support rich fishery; an abode for resident and migratory birds and it is the livelihood dependence of about 80 fishermen families living in village Punnakayal (Jayaseeli and Murugan, 2002). This mangrove supports rich biodiversity and fishery which is not documented till the present study was made. Largely in the global scenario, anthropological pressures and natural calamities threaten fragile coastal ecosystems. Punnakayal mangroves ecosystem lies in close proximity of a massive Titanium Dioxide (TiO_2) plant, caustic soda plant, fish curing centers and salt pan drains in to the area. This mangrove patch is also severely denuded by cutting the mangroves for fire-wood and building bunds for salt pans. Moreover, hypersalination due to salt-pan brine release also inhibits/curtails the growth of the mangroves of this area. Such pressures would ultimately lead to massive degradation and destruction of habitats.
The specific objectives of the study are to determine the physico-chemical parameters including heavy metal distribution in the Punnakayal mangrove area.

MATERIALS AND METHODS

Various environmental parameters such as temperature, salinity, pH, electrical conductivity, turbidity, TSS, dissolved oxygen and biological oxygen demand etc. were recorded from 6 stations fixed in the Punnakayal estuary. Water temperature was measured with a mercury thermometer with ± 0.02˚C accuracy. Salinity was measured with a refractometer (Erma, Japan). pH of water was measured with a calibrated pH pen (pH Scan1 Tester-Eutech Instruments, Singapore) and sediment pH was measured by soil pH Tester (Model DM-13, Takemura Electric Works Ltd., Tokyo, Japan). Electrical conductivity was measured by EC meter and the turbidity was measured using (ELICO PE 138). Total suspended solid was estimated by filtered method (APHA, 1989). Biological oxygen demand was estimated with the help of Light and dark bottle method (APHA, 1989). Water samples were collected for dissolved oxygen estimation by using BOD bottles. Dissolved oxygen was immediately fixed and brought to the laboratory and analysed using the modified Winkler's method (Strickland and Parsons, 1972). Water nutrients *viz.,* silicate, phosphate, nitrate and nitrite were analyzed by using the methods of Strickland and Parsons (1972). Calcium and magnesium were measured by complexometric titration Murphy and Riley (1962). The organic carbon content in the sediment samples was determined by using the standard method of Wakeel and Riley (1956). Trace elements or Heavy Metals like Copper(Cu), Nickel(Ni), Chromium(Cr), Cadmium(Cd), Lead(Pb), Zinc(Zn), Manganese(Mn), Iron(Fe) and Mercury(Hg) were analysed for water samples and sediments. All above metals were analysed in Atomic Absorption Spectrophotometer (GBC 132 plus) and Mercury was analysed by cold vapour technique using Mercury Analyser attached to the AAS.

RESULT AND DISCUSSION

Physico-chemical properties of Punnakayal mangrove area

During the study period the observed variations were minimal between the stations but prominent monthly variations could be observed. The variations in physico-chemical parameters have been represented in Figures 1-5. The water temperature is influenced by atmospheric temperature. Thus, the temperature steadily increased from January to April-May with its peak in May. The differences in temperature are mainly between the summer and

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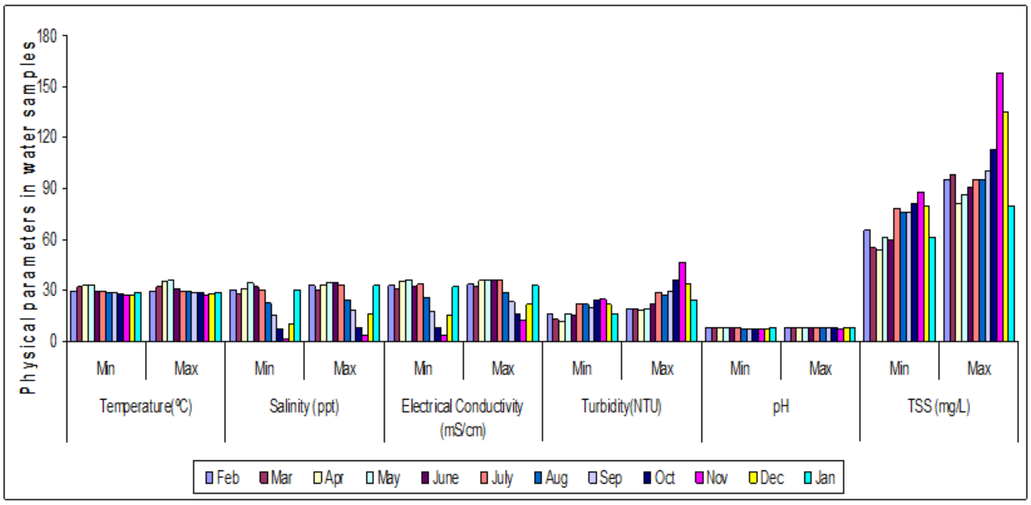

Figure 1: Physical characteristics of water at Punnakayal during February 2010 to January 2011

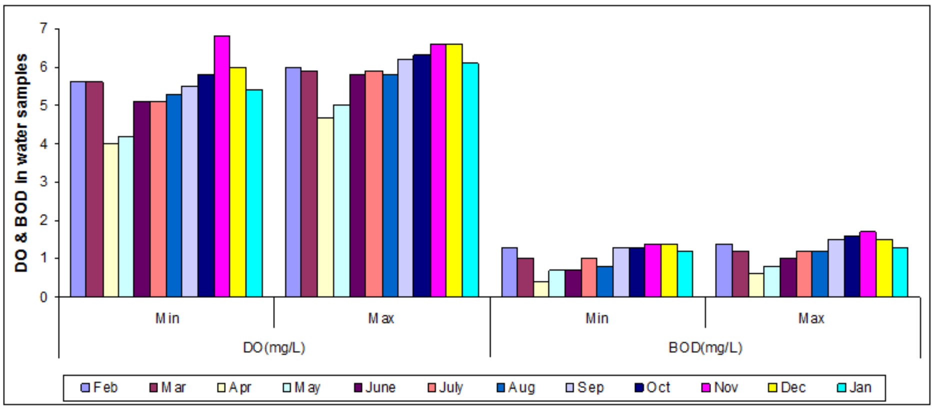

Figure 2: DO & BOD in Punnakayal waters during February 2010 to January 2011

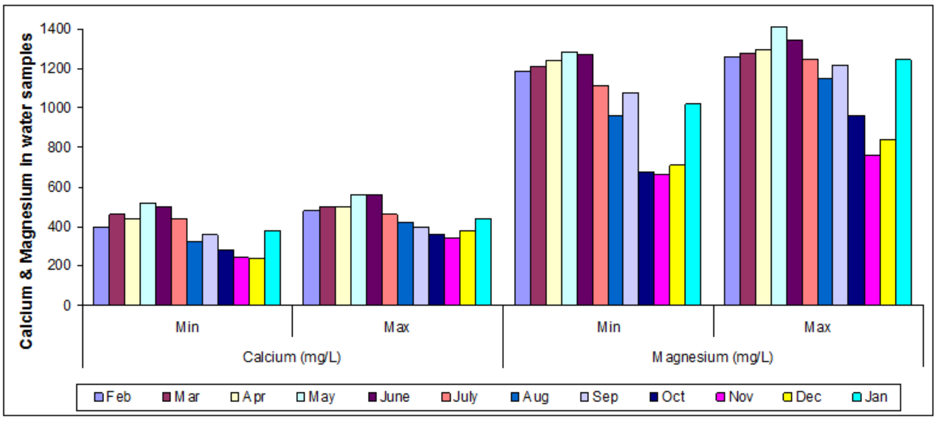

Figure 3: Calcium and Magnesium in Punnakayal waters during February 2010 to January 2011

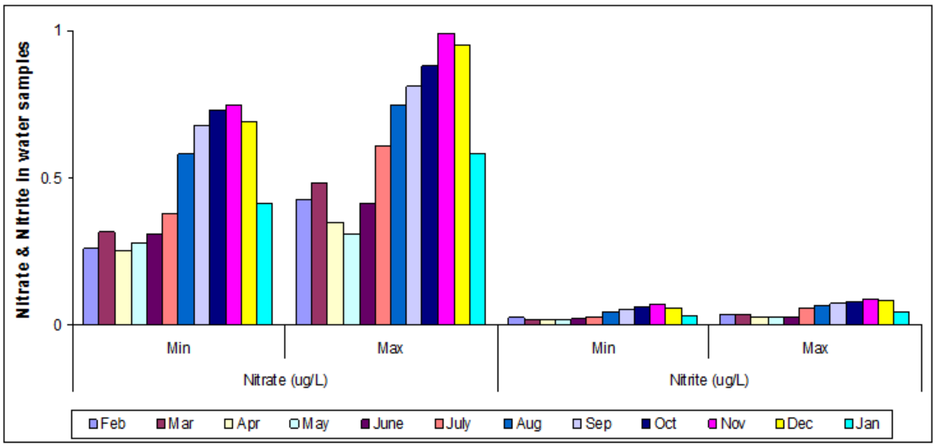

Figure 4: Nitrate and Nitrite in Punnakayal waters during February 2010 to January 2011

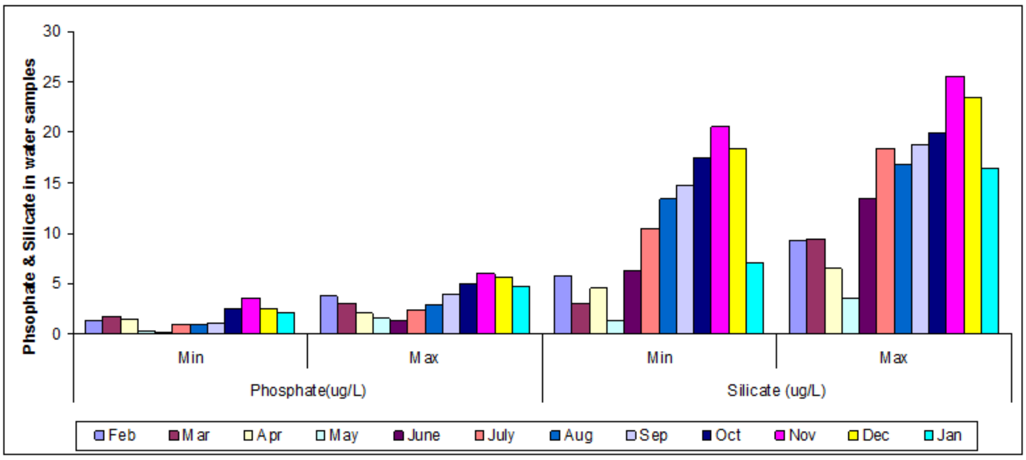

Figure 5: Phosphate and Silicate in Punnakayal waters during February 2010 to January 2011

monsoon months. Thangaraj *et al.* (1979) and Rajapandian *et al.* (1990) observed maximum temperature during summer and minimum temperature during monsoon seasons at all the stations. This could be attributed due to high solar radiation (Prabu *et al.*, 2008). Lower temperature was observed due to cloudy sky and rainfall (Kannan and Kannan, 1996). Similar observations have been reported by Thangaraj (1985), Gothandaraman (1993) and Seenivasan (1998) from Vellar estuary, Mani (1989), Vasantha (1989), Kaliyaperumal (1992) and Karuppusamy (1997) from Pichavaram mangroves water. Saraswati (1993) from Arasalar and Kaveri estuarine complex and Kannan and Kannan (1996) from Palk Bay. Salinity maintained a range between 30-33ppt except for the monsoon months (Oct-Dec) and during the late premonsoon month of September, the range was between 1-16 ppt. The general range was 1-34 ppt. This salinity range is in order of the general salinities of the Gulf of Mannar (Kumaraguru, 2006). This region is dominated by the sea. Jayaraman (1951) reported that salinity increased from April to August and declined during November and December in Madras coastal waters. Jayaraman (1954) recorded low salinity of 23.9 ppt during December and January. The variations in salinity in the study site was mainly influenced by the rainfall and entry of freshwater as reported earlier for Gulf of Kachchh by Saravanakumar *et al.* (2008) and for Godavari estuary by Raut *et al.* (2005). La Fond (1954) explains that the low salinity is caused by discharge of Ganges and Brahmaputra river's fresh water and it dilutes the surface waters. During summer higher salinity could be attributed to faster evaporation.

The Electrical conductivity followed a similar trend from January to July (36.1 mS/cm) and started to decrease from August to December with a record low during November (2.8 mS/cm). The pH range has also dropped in par with the salinity range starting from July, steadily decreasing till December and again elevating from January months (7.8 to 8.1). The same result was recorded during monsoon in Punnakayal estuary of Tuticorin (Mala and Vaitheeswaran, 2009). Turbidity followed a similar trend from February to June (11 NTU) and then there is a gradual increase reaching its peak during November (46.5 NTU). The gradual increase is due to the premonsoonal showers and the peak at November is due to the land runoff due to monsoon. Easterson *et al*. (2000) had reported turbidity between 13.4 to 45.6 NTU from Tuticorin coastal waters which was similar to the present study. Punnakayal mangrove found at the end of Thamiraparani River, during October, November and December was loaded with huge load of agriculture waste, domestic waste, etc. this type of waste may induce turbidity level in water. Heavy land runoff influences the turbidity of these stations during monsoon. The same trend was observed by Igbinosa *et al.* (2009) in South Africa. Slightly lower values (4.0 ppm) for dissolved oxygen could be observed during peak summer (April and May) and comparatively higher values (6.8 ppm) could be observed during November. The former observation is due to the summer heating and the later is due to the enrichment of the area with oxygen-rich fresh water. Comparatively lower BOD values were observed during Apr-Jun (0.4 to 1.0 ppm). James and Najmuddin (1986) reported steady increase of the dissolved oxygen content from October to December and slow decrease in dissolved oxygen content from January to March and this drop in oxygen content may be due to the absence of strong winds thus decreasing the solubility of atmospheric gases associated with increasing surface water temperature as summer approaches by the end of March. Das *et al.* (1997) and Saravanakumar *et al.* (2007) mainly attributed seasonal variation of dissolved oxygen to freshwater influx and ferruginous impact of sediments. According to Kannan and Kannan (1996) seagrass environment showed lower dissolved oxygen concentration during the summer season and it may be due to the higher surface water temperature recorded during this season in Gulf of Mannar. However the subsequent dissolution of oxygen might have been reduced due to higher surface water temperature recorded during this season. Magnesium (675 to 1399 ppm) and calcium (238 to 560 ppm) followed a similar trend of distribution. Both the elements were in the higher range during February to June and there is gradual decrease from July-December with lowest values during November. This is due to the dilution of the calcium-magnesium rich seawater by the fresh water. In monsoon season, (October, November and December) high rainfall was observed and land runoff also occurred during these months. These factors could be the reason for change of the calcium and magnesium in water column.

The same trend has been reported in Yamuna River in Haryana by Ravindra *et al.* (2003) that Magnesium level was low in winter season. Jeyaprakash *et al.* (2005) suggested that calcium and magnesium level could be reduced due to input of fresh water in the estuary. Narvekar and Zingde (1987) have also reported a similar result. Nitrate was higher than nitrite at all the stations sampled. This is indicative of fairly good oxygen rich environment. However, nitrate and nitrite content was lower during the months of January-June (0.25 and 0.015 ppb respectively). From July, the nitrite and nitrate content has gradually increased with its peak concentration during November with 0.99 and 0.108 ppb respectively. The increased nitrates level is due to fresh water inflow, mangrove leaves litter fall decomposition and terrestrial run-off during the monsoon season (Karuppasamy and Perumal, 2000). The higher nitrite value in monsoon season could be due to the increased phytoplankton excretion, oxidation of ammonia, reduction of nitrate, the recycling of nitrogen and bacterial decomposition of planktonic detritus (Swami *et al*., 1996; Govindasamy *et al*., 2000; Asha and Diwakar, 2007). Further, denitrification and air-sea interaction exchange of chemicals are also responsible for these increased values (Mathew and Pillai, 1990; Choudhury and Panigrahy, 1991). Higher salinity reduced the nitrite level in water during summer (Saravanakumar *et al*., 2008). Mani and Krishnamurthy (1989); Murugan and Ayyakkannu (1991) suggested that the less fresh water inflow and higher salinity reduced the nitrite level in water. Nitrite concentration was found to be much lower than that of nitrate and the same trend of fluctuation was noticed in Muthupettai mangrove by Ashokkumar *et al.* (2011). The phosphates have shown a decreasing trend from January to June and from July the concentration showed an increasing trend with the peak value during November (6.0 ppb). Two currents in Gulf of Mannar are reasons for fluctuation of nutrients, northeast monsoon current increased the phosphate level and south west monsoon reduced the phosphate level in water. This result has been supported by Jayaraman (1954). The low values of phosphate observed may be due to the utilization by phytoplankton and other primary producers. Phosphate levels were observed to be high during monsoon period Sarkar *et al.* (2007).

The low phosphate value could be attributed to the high utilization of phosphate by phytoplankton and also the variation may also be due to the processes like adsorption and desorption of phosphate and buffering action of sediment under varying environmental conditions (Rajasegar, 2003). Silicate concentration was also found to be minimal from January to May with a record low during May (1.4 ppb). The concentration of silicates in the waters around Punnakayal was maximum during November with a gradual increase from May. In October, November and December months, higher rainfall influenced land runoff and high fresh water inflow increased silicate concentration in Gulf of Mannar. It has been reported that the silicate from the bottom sediment might get exchanged with overlying water in this mangrove environment (Govindasamy and Kannan, 1996; Rajasegar, 2003). The low value observed in summer and post monsoonal seasons could be attributed to uptake of silicates by phytoplankton for their biological processess (Prabu *et al*., 2008; Saravanakumar *et al*., 2008). Paramasivam and Kannan (2005) and Ajithkumar *et al.* (2006) have reported silicate level up to (128.6 µM) from this mangrove environment. Gopinathan and Rodrigo (1991) have reported that higher plankton induced negligible amount of silicate in Tuticorin water. The above are the observations on the trend of variations in the physico-chemical parameters in the Punnakayal region during the study period. Simillar observations on physicochemical parameters were conducted by Gandhi (1998) who observed the physicochemical parameters in the range observed during the present study. Mahalakshmi (2002) has made similar observations as of the present study in the case of nutrient distribution. The observations made during the present study clearly indicate that there is marked difference in the water-quality based on seasons. Also the freshwater inflow has a major influence on the nutrient load of the region. The elevated nutrient levels during the monsoon period of the present study are indicative that the river collects rich load of nutrients from the land and drains into the Punnakayal estuarine area.

Distribution of Heavy metals

Studies on the distribution of heavy-metals are inevitable to assess the pollution load in an area. Particularly the Punnakayal area warrants such investigation due to its closeness to industries and the possible load of metals in the atmosphere which could enter the area due to precipitation. The variations in heavy metal concentrations in the water and sediment during different seasons at various stations have been represented in Figures 6 and 7. Zinc showed discontinuous distribution through various seasons (BDL to 0.630 ppm). The concentration of zinc was in the lower ranges during January to May. However station 1 showed higher concentration of the metal during March (0.600 ppm). According to Boxall *et al*. (2000) zinc can enter the aquatic environment from a number of sources including industrial discharges, sewage effluent and river runoff. Asha *et al*. (2010) reported lower zinc level in Tuticorin coastal waters than the present study. The concentration of the metal showed an increasing trend from June and attained its peak concentration during November. Sediment concentration (BDL to 0.710 ppm) was not found to vary much but still on the higher side during the months of June to November. Ashokkumar *et al*. (2009) recorded higher zinc concentration in Muthupettai Mangroves, Southeast Coast of India than the present investigation. Ashokkumar *et al*. (2009) suggested domestic, municipal, industrial and agricultural waste was the reason for the addition of zinc in the marine environment. Lead showed discontinuous distribution (0.100 to 0.980 ppm) it was minimum during April-July and at all other period of the year lead was comparatively on the higher side. The stations 1-4 situated in the mangrove area showed higher concentrations of lead. The concentration of lead was higher during rainy season due to the increase of total organic carbon content (Tukura *et al.,* 2007). According to Mitra. (1998), the lead concentration increased during monsoon due to huge load of freshwater runoff. Batley and Brockbank (1990) suggested that multifarious sources like domestic sewage, power-plant operation, major storm events or dumping of sediments dredging along the international shipping zones induced increase in the lead level in the marine environment. Sediment concentrations were largely stable except for April-May when it was comparatively lower. Stations 2 and 3 located in the mangrove area mainly had higher loads of lead (0.880 and 0.913 ppm). The presently recorded concentration of lead is slightly higher than a earlier study in Tuticorin coast by Jonathan and Ram Mohan (2003). Sujatha *et al*. (2008) reported higher lead concentration than the present study in Nagapattinam coast. Lead concentration in seawater can be ascribed to the influence of external discrete sources like sewage, agricultural runoff and leaching from antifouling paints used in fisherman boats and also from leakage of leaded diesel and petrol from boats (Abu-Hilal 1987).

Iron in the water column (BDL to 2.88 ppm) was comparatively in the lower ranges from February to September whereas, elevated levels could be recorded during Oct-Dec. There is a gradual increase in sediment iron concentration from July to November (0.330 to 4.82 mg/gm) and it started to decrease from the month of December. Sewage waste, domestic and river flow and land runoff implement the iron content in water and sediment. Organic content and sediment texture is a reason for metal load in the marine environment. Higher iron concentration than the present study has been recorded by Jonathan and Ram Mohan (2003) in the coastal sediments of Tuticorin. The concentration of chromium (BDL to 0.880 ppm) gradually increased from January to November and started to decline in December. The same trend has been observed in Manakudy estuary, southwest coast of India (Kumar and Edward, 2009). River water loaded with coir retting pits and domestic sewage from fishermen settlement has been attributed to chromium concentration in water and sediment of Manakudy estuary. The Stations 1-4 had comparatively higher concentration of the metal. Chromium concentration in the sediments was low in the months of January to May (BDL to 0.350 ppm), moderate during June to September gradually increasing in October and reached its maximum during November (0.230 to 0.910 ppm). Concentration then in according to the results of Izmir bay by Turkoglu *et al.*(1992) studied chromium concentration in the Izmir bay and reported that precipitation processes of high organic and inorganic suspended matter adsorb chromium and other heavy metals from water and increases the sediment concentration of these metals whereas, the comparative water concentration is low. Maanan *et al*. (2004) reported that the effluent of metal finishing industry and corrosion of building materials play an important role in increasing the chromium concentration in the marine environment.

Similar concentration was reported in sediments of Albanian coast (Celo *et al.,* 1999). Nickel concentration (0.100 to 0.330 ppm) was in the stable range from July-January. Comparatively higher concentrations were recorded during March and nickel concentration was below detection during May and June and during April it was recorded only at station 3. Sediment concentration (BDL to 0.577 mg/gm) also followed a trend as the water column with seasonal variations. Manganese in the water column (BDL to 0.420 ppm) has shown minimal and more or less constant concentration through various stations and seasons. Comparatively much higher

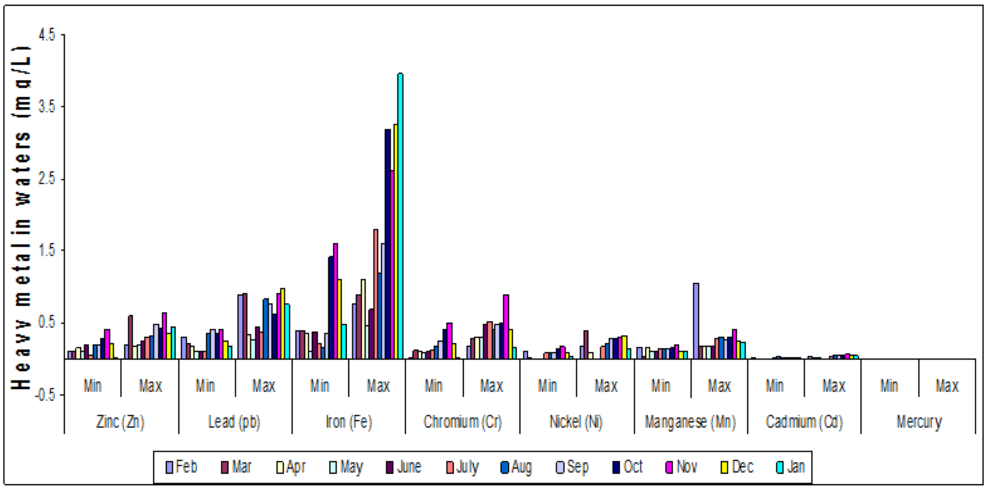

Figure 6: Concentration of heavy metals in the water column at different stations at Punnakayal during February 2010 to January 2011

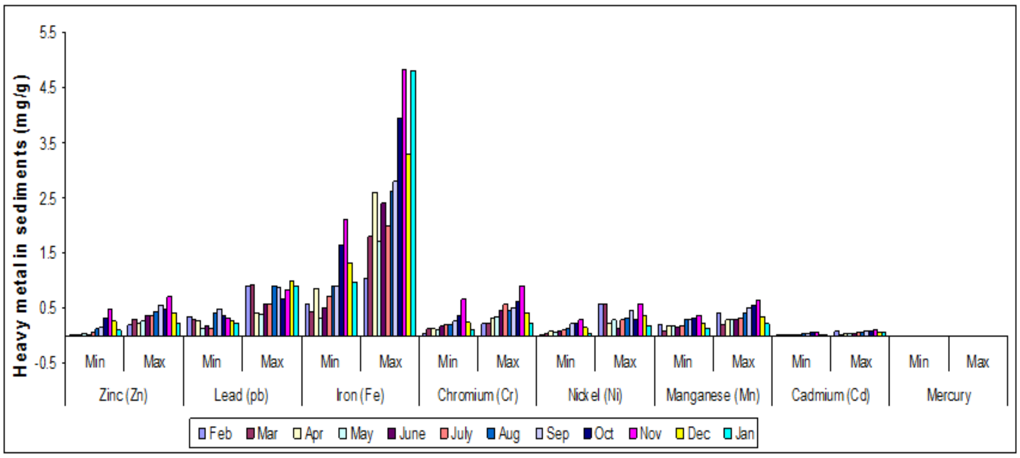

Figure 7: Concentration of heavy metals in sediments at different stations at Punnakayal during February 2010 to January 2011

concentration of manganese was detected in the sediments (BDL to 630 ppm). Also, the concentration in water and sediment was higher during the months of August to November. Ganesan and Kannan, 1995 suggested that the industrial pollution induces the nickel and manganese level in coastal environment of Tuticorin. Higher concentration of cadmium (BDL to 0.060 ppm) in the water column was present during the months of July-December. During February-June the concentration was comparatively lower. The concentration of cadmium in the sediment (BDL to 0.110 ppm) was comparatively low from February to June and there was gradual increase from July to November exhibiting the peak concentration during November.

According to Fianko *et al*. (2007) cadmium may enter the marine environment due to the geology of the catchment soil and runoffs from phosphate fertilized agricultural soils, disposed off nickel-cadmium based batteries and cadmium-plated items. High concentration of industrial discharge, domestic waste, sewage disposal, river runoff, stripping and painting of ships are the other sources for cadmium metal in marine environment (Fatoki and Mathabatha., 2001). Cadmium is a natural constituent of rock phosphates and deposits in some regions of the world and certain regions contain markedly elevated natural levels of this metal. Analyzing the trend of results obtained during the present study, it is evident that the monsoon plays a prominent role in the distribution of heavy metals. There is marked seasonal changes in the metal concentration. It is observed that the stations with mangroves hold comparatively higher concentration of heavy metals. The levels indicate that the Punnakayal area is not polluted with heavy metals. However, frequent studies are warranted in this account.

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