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RESEARCH ARTICLE

DISTRIBUTION OF HEAVY METAL CONCENTRATIONS IN SURFACE WATERS FROM ENNORE ESTUARY, TAMIL NADU, INDIA

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ABSTRACT

An investigation was carried out in the Ennore creek for the assessment of heavy metal pollution pertaining to cadmium, copper, lead and zinc during January – December 2008 in all the four stations of post-monsoon, summer, pre-monsoon and monsoon. The data processed revealed that the concentrations of cadmium, copper, lead and zinc were above the permissible limits of coastal standards. The station wise levels of heavy metal concentration were in the order of station 4> station3> station 2> station 1. The seasonal variation of heavy metals observed in the Ennore estuary was maximum during summer and monsoon. Strong correlation (P<0.001, α=0.05) was found among the stations in the Ennore creek during post-monsoon, summer, pre-monsoon and monsoon. Among the stations in the Ennore estuary stations 3 and 4 were highly polluted and stations 1 and 2 was less polluted with cadmium, copper, lead and zinc.

INTRODUCTION

Humanity's ability to mine and use metals has played a major role in development of modern human society. Anthropogenic activities in and around the coastal areas largely contribute the sources of metals to the aquatic environment (Botte *et al.,* 2007). Estuaries determine the fate of metals of continental origin to the ocean (Ridgway and Shimmield, 2002). The net flux of riverine inputs to the open ocean depends on their biogeochemical behaviours in the estuary (Morillo *et al.,* 2008). Many metals have a wide range of uses, but these have come at a significant environmental price, have serious negative environmental consequences, yet our dependence on them continues to result in large inputs into our environment (Ahiamadjie *et al.,* 2011). Heavy metals are an important category of pollutants and as such have major detrimental impact on human and environmental health (Ogundiran and Afolabi, 2008). Antifouling boat paint particles are readily transported into the local aquatic environment with wash water and runoff or as airborne dust. Their potential for long-range

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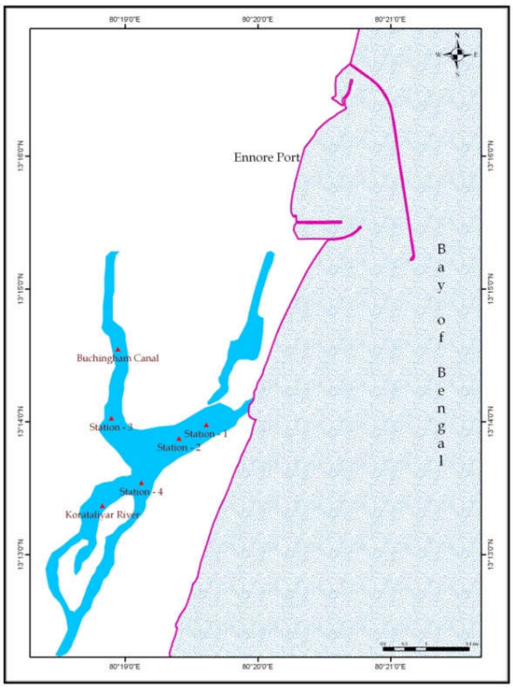
transport, coupled with a relatively high surface area and erodibility, suggests that antifouling boat paint particles may pose chemical and biological impacts (Singh and Turner, 2009). Elevated concentrations of biocidal components, including copper and zinc and various organic boosters, are frequently reported (Helland and Bakke, 2002). Ennore creek carries high load of heavy metals (Kannan *et al.,* 2007). Buckingham canal and Korataliyar river are no longer able to receive and assimilate effluents because they have fallen below minimum levels of flow. The treated effluents of the Madras Refinery Ltd, through the Buckingham canal and the Madras Fertilizers Ltd, through the Red Hills surplus channel, reach the Ennore backwater (Sreenivasan and Franklin, 1975). The coastal erosion has become a perennial problem for the people living along the Royapuram-Ennore coast (Ramakrishnan, 2002). In the present investigation the Ennore estuary will be assessed for the concentrations of cadmium, copper, lead and zinc with the coastal standards (NEB, 1994) in the stations 1, 2, 3 and 4 during post-monsoon, summer, pre-monsoon and monsoon.

MATERIALS AND METHODS

Study area and sampling methods

The southeast coast of India is an important stretch of coastline, where many major rivers drain into the Bay of Bengal and they are also richer in marine fauna than the western coast of India. Ennore creek (13°13'54.48" N, 80°19' 26.60" E) (Fig 1) is located in the northeast coast of metropolitan Chennai city, Tamil Nadu, India. Most of the area consists of alluvial tracts and beach dunes, tidal flats and creek in the eastern part. Ennore comprises of lagoons, with salt marshes and backwaters, which are submerged under water during high tide and form an arm of the sea opening in to the Bay of Bengal. The total area of the creek is 2.25 sq km and is nearly 400 m wide. Its channels connect it to the Pulicat lake to the north and to the Kortalaiyar river in the south (Kannan *et al.,* 2007). Ennore coast receives untreated sewage from Royapuram sewage outfall, untreated / treated industrial effluents from Manali Industrial Belt, which houses many chemical industries. The dredging activities in Ennore area result in changes in the landscape, sediment transport, and dust pollution to the coast by quarrying process (Palanisamy *et al.,* 2006).

Water samples were collected during the period of January-December 2008. The study period was alienated into post-monsoon (January, February and March), summer (April, May and June), pre-monsoon (July, August and September), monsoon (October, November and December). In Ennore estuary, station 1 was located in the bar mouth region (13°14'02.31" N, 80°19'49.47" E), station 2 was located the creek (13°13'52.54" N, 80°19' 24.26" E) between the stations 1, 3 and 4, station 3, the Buckingham canal (north towards Pulicat lake) (13°14'02.72" N, 80°18' 54.18" E) and station 4 right down the railway bridge (13°13'30.39" N, 80°19' 02.30" E). The water samples for trace metal analysis were collected using 1 L polyethylene-tereftalate (PET) bottles and immediately acidified with 3ml of concentrated nitric acid.



*Map was designed from ARCGIS 9.0

Fig. 1. Ennore creek

The preserved samples were transported to the laboratory and were filtered through Millipore vacuum pump using 0.45 µm filter paper (HA-Millipore). The filtered samples were acidified using 1.5 ml of suprapure nitric acid analytical quality (Merck) until the pH was adjusted to 2. These samples were stored at 4°C until analysis. Sampling was carried out monthly in duplicates. Prior to use all the glassware's employed for sampling, filtration and sample storage were carefully cleaned with 10 percent nitric acid (Suprapure, Merck) and rinsed with single followed by double distilled water and dried in oven following internationally recommended protocols (APHA, 1998).

The dissolved metals in seawater was extracted according to the method described by Koirtyohann and Wen (1973) and updated by El-Moselhy and Gabal (2004). Metal concentrations were determined with a Varian SpectraAA 220FS Atomic Absorption Spectrophotometer (AAS) with an air/acetylene flame. Appropriate internal standards (Merck Chemicals, Germany) were used to calibrate the instrument. Analytical grade reagents were used to make up the relevant blanks.

RESULTS

Concentrations of cadmium, copper, lead and zinc in the surface waters of the Ennore creek varied significantly ($P<0.001$) with respect to seasons and stations. In post-monsoon, heavy metals concentrations in station 4 were high (Fig 2, 3, 4 and 5). The concentrations of heavy metals were above the coastal standards of National Environment Board (NEB). During monsoon the heavy metal concentrations measured were high when compared with post-monsoon, summer and pre-monsoon. Measured concentrations for cadmium, copper, lead and zinc was high in stations 3 and 4 when compared with stations 1 and 2, the correlation was significant at $P<0.001$ ($\alpha=0.05$) (Fig 6). The maximum levels of cadmium was found in station 4 (0.024 mg/l) during monsoon; the copper concentration was 0.156 mg/l in postmonsoon. Lead and zinc of 0.226 and 0.372 mg/l were the maximum concentrations found in station 4 during monsoon

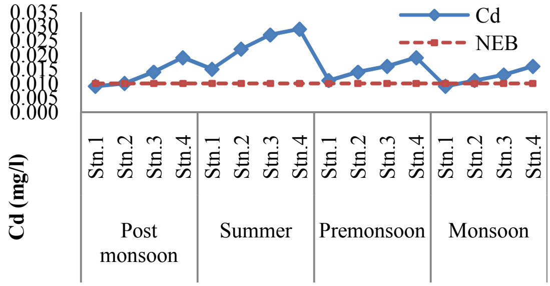


Fig. 2. Seasonal changes in the dissolved cadmium in the surface waters of Ennore creek in stations1, 2, 3 and 4; National Environmental Board, 1994 (NEB).

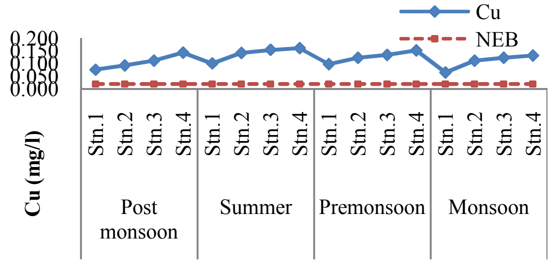


Fig. 3. Seasonal changes in the dissolved copper in the surface waters of Ennore creek in stations1, 2, 3 and 4; National Environmental Board, 1994 (NEB)

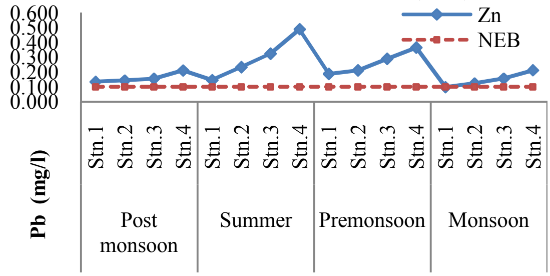


Fig.4. Seasonal changes in the dissolved lead in the surface waters of Ennore creek in stations1, 2, 3 and 4; National Environmental Board, 1994 (NEB)

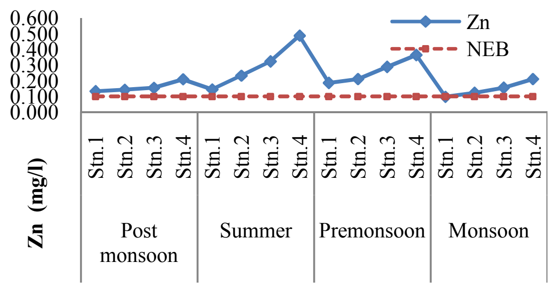


Fig.5. Seasonal changes in the dissolved zinc in the surface waters of Ennore creek in stations1, 2, 3 and 4; National Environmental Board, 1994 (NEB)

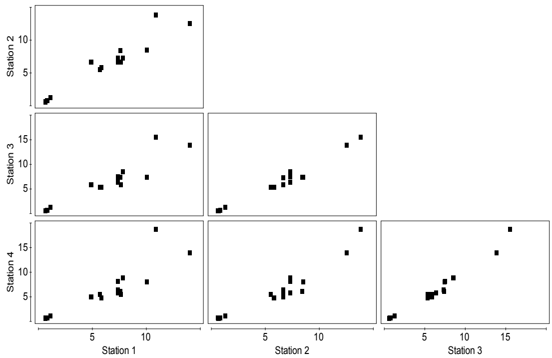


Fig.6. Draftsman plot of the dissolved heavy metals in the surface waters showing the strong correlation between the stations 1, 2, 3 and 4 (r^2= 0.91, 0.90, 0.96 and 0.87) (α=0.05) at *P*<0.0001 (2-tailed) in Ennore creek during postmonsoon, summer, premonsoon and monsoon

The chief source of cadmium, copper, lead and zinc was found in stations 3 and 4 which was distributed throughout the creek correlating with a high significance in all the stations. Above all, the zinc concentrations were high followed by lead, copper and cadmium in all the stations irrespective of seasons. The concentrations of heavy metals in the Ennore estuary was high in station 4 followed by station 3, 2 and 1in the order of station 4>station 3> station 2> station1.

DISSCUSION

The present investigation reveals that the heavy metals in the surface waters are above the permissible limits throughout the creek. The stations 3 and 4 are the prime source of heavy metals. Metal concentrations measured during monsoon were higher than in summer. Al-Saadi *et al.* (2002) investigated concentrations of heavy metals in water of Habbaniya lake (Iraq) and found the following concentrations: 15-86 mg/l of zinc, 5-50 mg/l of copper, 0.3-33.3 mg/l of lead and metals displayed the highest values during the spring, except zinc during the winter. When compared with the present study in Ennore creek the heavy metal concentrations were lower than the heavy metal concentrations in Habbaniya lake (Iraq). Karadede and Unlu (2000) reported concentrations of heavy metals at Ataturk Dam lake (Turkey) in mg/l: as 0.025-0.220 of copper, 0.006-0.197 of zinc. Studies in Pulicat Lake (Tamil nadu, India) recorded an elevated level of heavy metal concentrations, especially iron, cadmium and mercury (Kannan and Krishnamoorthy, 2006). Cadmium concentration in water samples was 0.01 mg/l during premonsoon and postmonsoon. Rajathy and Azariah (1996) reported that the levels of zinc and copper in water and sediment samples showed seasonal fluctuations in the Ennore and Adayar estuary. In addition, the high values of metals in stations 3 and 4 were due to the low salinity and input of sedimentary fluxes (Cutter, 1991). Seasonal variation in metal distribution is influenced by strong hydrodynamic and physiochemical conditions (Padmini and Kavitha, 2005a & b). Dissolved cadmium values reported by Botte *et al.* (2007) varied from 0.0001 to 0.0024 mg/l at Puerto Galvan. Recordable levels of cadmium have been measured in the study area, which is agreed with previous reports from Andrade *et al.* (2000) findings of <0.0005-0.001 mg/l Cd and by Ferrer *et al.* (2000) who reported 0.0001 mg/l of cadmium at Bahia Blanca Estuary (Argentina).

The levels of dissolved cadmium in Ennore water presented in this study was in the same magnitude to those reported by Mucha *et al.* (2004) at Douro estuary, in Portugal, as well as those concerned with Jayaprakash *et al.* (2005), considered as anthropically stressed but not strongly polluted systems. The values presented here were much lower than those reported for strongly polluted estuaries, such as the Rio Tinto Ria Huelva on southwest Spain (Achterberg *et al.,* 2003), where the values were 0.045 mg/l Cd. Goody *et al.* (2002) reported cadmium concentrations from the Chalk system, at Salisbury (Wiltshire, UK) of 0.0001-0.0027 mg/l, values were lower when compared with the present study. Man *et al.* (2004) reported for the Mai Po and Inner Deep Bay, on Western Hong Kong, in China (0.0005-0.004 mg/l). All the above literature values are lower, when compared with the concentrations of cadmium in Ennore creek, which clearly indicates that the creek is polluted with cadmium. The evident values of dissolved lead, sustained that a continuous source of this metal occurs within the studied region. Dissolved lead concentrations from Ennore creek as presented in this study were higher when compared with those reported by Vazquez *et al.* (1998) for Alvarado Lagoon water, in Mexico (0.009-0.063 mg/l), also reported by Nayar *et al.* (2004) for the Ponggol estuary, in Singapore (0.020 mg/l). Goody *et al.* (2002) reported 0.0002-0.0038 mg/l Pb in UK and Man *et al.* (2004) reported 0.0002-0.185 mg/l Pb in China. Present day concentrations of heavy metals are several orders of higher magnitude (Man *et al.,* 2004). Hall *et al.* (1996) reported lead concentration in the dissolved phase was between 0.0009-0.0002 mg/l in Scottish estuaries with low anthropogenic input. Dissolved lead concentration in station 1 decreased with increasing salinity, but in station 4 with lower salinities a complex behaviour exists with high concentration of lead in the downstream direction (Dauby *et al.,* 1994). This increase in dissolved lead in Ennore creek is linked to the high industrialization of the surrounding areas, which produces large amounts of lead-based effluents. Moreover, the shallow depth of the river favours re-suspension of sedimentary matter that can contain a significant amount of lead, easily scavenged at the sediment-water interface (Dauby *et al.,* 1994). In the present study conducted in Ennore creek the lead concentrations were higher than the literature values and the permissible limits in all four stations during postmonsoon, summer, premonsoon and monsoon, indicating pollution of lead in the aquatic environment. Brigden *et al.* (2005) reported lead concentrations between 17 and 247 times higher, exceeding guidelines of WHO (2004) by 190 to 2400 times in India (New Delhi and Bangalore). Dissolved metals are considered to be the most mobile, thus reactive in bioavailable fractions of the aquatic system and are cause for concern (Wong *et al.,* 2007). Dahuri and Pahlevi (1994) reported copper concentrations at Asahan and Deli Serdang in North Sumatra and Lhokseumawe in North Aceh (Indonesia), ranging from 0.071 to 0.107 mg/l, which are comparatively lower in Ennore creek. Station 4 had higher concentration of heavy metals than station 1, which may be closely associated with suspended particulate matter (SPM) inputs either land-originated or mainly linked with re-suspension and remobilization processes with an active role of benthic biogenic particles (Wang and Liu, 2003). The present study indicates that the zinc concentrations were above the national environmental board (NEB, 1994) guidelines. The concentration of dissolved zinc shows a semi-conservative behaviour at high saline in station 1 and low saline ends in station 4, where high values of zinc exist in stations 4 and 3. The results indicate that zinc is a non-soluble element even at low salinities and shows adsorption which is a major process controlling the distribution of dissolved zinc in Ennore creek (Benoit *et al.,* 1994). The high average in station 4 and the high values in the mixing zone in Ennore creek reveal that riverine inputs have increased the zinc concentrations along the station 4. Studies in other areas indicate that zinc may be complexed with organic ligands in surface waters and this phenomenon is also reflected in station 4 (Tanizaki *et al.,* 1992).

The results of the heavy metal investigation in the landfill sites reported by Ogundiran and Afolabi (2008) showed that zinc was the most predominant metal in the landfill ranging from 0.264-0.947 mg/l higher than the present study in Ennore creek, while cadmium concentration was the lowest ranging from 0.001-0.022 mg/l. The concentration of lead, cadmium and zinc in the station 4 was the highest when compared with other sites, indicating that the site could be a possible source of cadmium and lead. Concentration of dissolved trace metals is inversely related to the salinity of the anoxic zone where dissolved oxygen values are also low (Hatje *et al.,* 2001a & b). Rukah and Al-Kafahi (2001) reported 0.012 mg/l Cd, 0.190 mg/l Pb, 95 mg /l Zn and 0.210 mg/l Cu in El Akader of Jordan. The high concentrations of heavy metals in sediments may not necessarily indicate anthropogenic contamination, because of different background levels in parent materials and sediment properties (Esen, 2010). All studied metals were low in summer and high in monsoon.

Conclusion

The assessment of heavy metal concentrations in Ennore creek using the coastal standards (NEB) produced valuable results. The concentrations were above the permissible limits during post-monsoon, summer, pre-monsoon and monsoon. Cadmium, copper, lead and zinc were high during the summer and monsoon in stations 1, 2, 3 and 4. The concentrations of heavy metals were high in stations 3 and 4 and less in stations 1 and 2 in the present study. In the present study the concentrations of zinc was high followed by copper, lead and cadmium. Anthropogenic (Industrial and domestic sewage) inputs from point and non-point sources would had governed the higher values of cadmium, copper, lead and zinc in the surface waters of Ennore estuary. The higher concentrations were above the permissible levels indicating that the estuary was not stable and suitable for the propagation of aquatic organisms especially the juvenile stages.

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