HISTORICAL POLLUTION TRENDS IN COASTAL ENVIRONMENTS OF INDIA

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Abstract. Seventeen sediment cores were collected from different coastal ecosystems of Tamil Nadu, India that include coastal lagoon (Pulicat), polluted rivers in Chennai (Adyar and Cooum), Coral reef (Gulf of Mannar) and a perennial river (Tamiraparani). Radiometric dating has been used to determine the modern sedimentation rates in these ecosystems. The Pulicat Lake and the polluted rivers (Adyar and Cooum) yield an average sediment accumulation rate of 12.34 and 7.85 mm yr^{-1}, respectively. In the Gulf of Mannar coral reef, the sedimentation rate averages 17.37 mm yr^{-1}, while the rate in Tamiraparani River is 11.00 mm yr^{-1}. In the Tamiraparani River basin, the deposition rates were an order of magnitude higher when compared to the erosion rates, which may be due to bank erosion and the intense human activity. In general high rates of sedimentation observed in the coastal ecosystems not only reflect the capacity of the coastal regions as sinks for trace metals but also denote increased input of pollutants into the coastal environments in the recent past. The deposition rates of heavy metals – Fe, Mn, Zn, Cu, Cr and Ni in the depth profiles have been computed using sedimentation rates and their distribution is discussed. It can be seen that the mean deposition rates of all the measured elements in the Tamil Nadu coastal ecosystems are high compared with rates determined for the sediments of the deltaic regions of India and the Bay of Bengal.

Keywords: coastal ecosystems, India, ^{210}Pb dating, trace metal contamination, time span

1. Introduction

The sediment quality of the coastal ecosystems of India is affected by industrial and agricultural activities in the urban and rural areas, respectively. Semi-enclosed areas such as coastal lagoons, estuaries etc., are particularly sensitive to the effects of human activity because they are not flushed as frequently as open coasts. Coastal marine sediments are sinks for many inorganic and organic pollutants transported from terrestrial sources.

Studies of variations in metal concentrations in dated sediment cores have been used to understand long-term trends in anthropogenic metal inputs to coastal environments (Appleby and Oldfield, 1992; French *et al.*, 1994). Such reconstruction studies are very useful to improve our management strategies and to assess the recent trend of pollutant inputs. For such a study, it is essential to establish reliable chronologies of sedimentation for the past 100 to 200 yr. Although several methods exist for estimating sediment accumulation rates in aqueous systems the sedimentary record in dynamic coastal environments is often obliterated by

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post-depositional mixing of particles and by physical and biological (such as '*bioturbation*') mechanisms. The use of short-lived radioisotopes such as ^{210}Pb is particularly useful in determining the ages of relatively recent lacustrine and coastal marine sediments. This dating method has been applied increasingly to studies concerned with the impact of human activity on the aquatic environment by several researchers (Krishnaswami *et al.*, 1971; Ramesh *et al.*, 1988; Ramanathan, 1992). The objective of this study is to determine the sedimentation rates and to quantify deposition rates of trace metals in the sediment cores from diverse coastal ecosystems of Tamil Nadu during the last 20 to 120 yr.

2. Materials and Methods

2.1. STUDY AREA

Tamil Nadu is the southern most State on the east coast of India with a coastline of ~1000 km (Figure 1). This coast has extensive areas of estuaries, mangroves, brackish water lagoons, coral reefs and sea grass ecosystems all of which are connected to the Bay of Bengal at various locations along the east coast. Hence, these coastal ecosystems have great ecological, social and economic significance within themselves and with the adjacent Bay of Bengal. The coastal habitats of South India (Figure 1) are threatened by domestic, agricultural and industrial pollution. The climate of the region is tropical and is characterized by high temperature (mean 27 to 30 °C) and medium rainfall (900 mm yr^{-1}).

2.1.1. *Coastal Lagoon (Pulicat Lake)*

Located 40 km north of Chennai city, the Pulicat Lake (Figure 1) is the second largest brackish water body flowing to the Bay of Bengal (Latitude 13°24′ and 13°43′N and longitude 80°03′ and 80°18′E). The lake is about 60 km in length and 0.2 to 17.5 km in breadth. It has a high water spread area of 460 km^2, and a low water spread area of 260 km^2. Due to deltaic deposits, the lake is extensively shallow, with an average depth of 1.5 m. The lake is separated from the Bay of Bengal by an inland spit called the Sriharikota Island. The main source of freshwater is land runoff through three small seasonal rivers that open into the lake: the Arani, Kalangi and Swarnamukhi. Water flows in these rivers only during monsoon (October–December). The Buckingham Canal, which runs parallel to the Bay of Bengal, brings in industrial and domestic wastes to the lake ecosystem and eventually to the Bay of Bengal.

The hydrology of the Pulicat Lake is influenced by local climate, the regime of the inflowing rivers, the Buckingham Canal that enters the lake, in addition to the effect of the neritic waters of the Bay of Bengal. The salinity of the surface water shows wide seasonal and spatial fluctuations. During monsoon, when freshwater inflow is greater, this lake is a positive estuary but at the peak of summer,

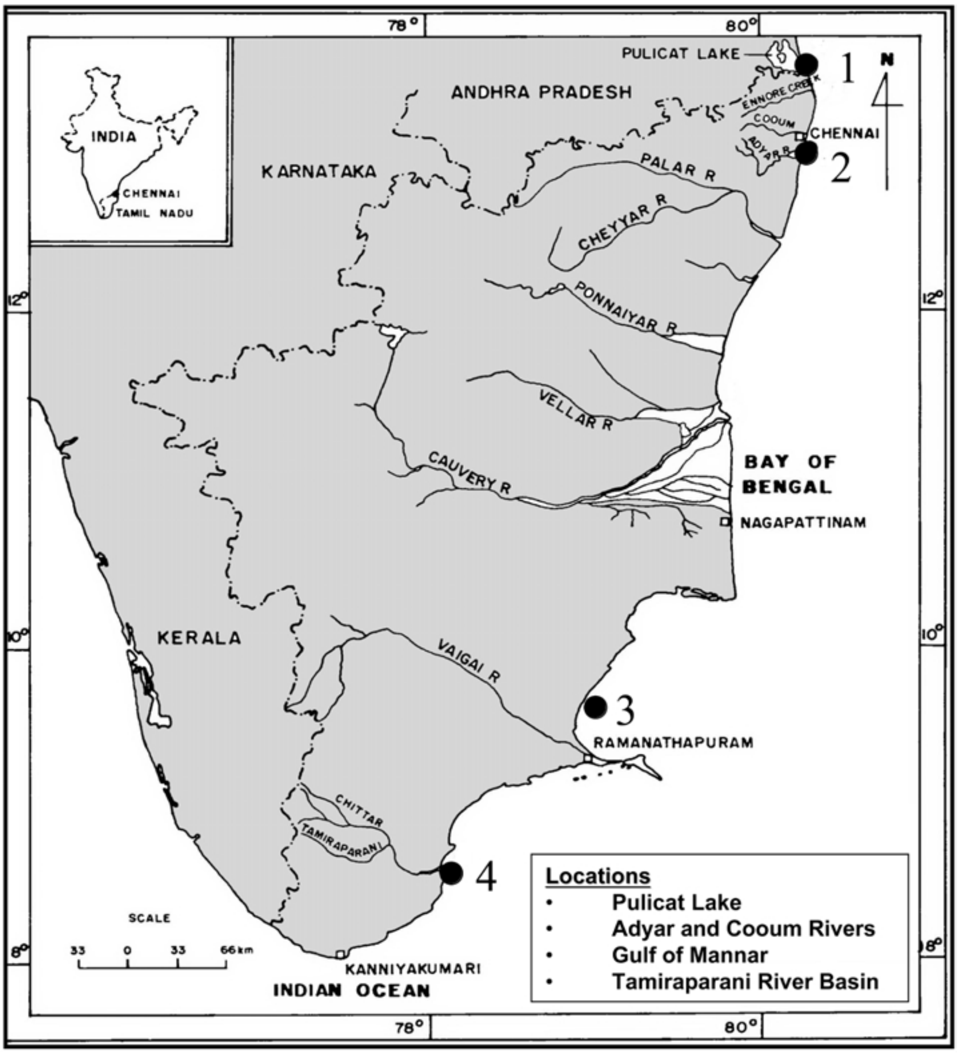

Figure 1. Major coastal ecosystems along the Tamil Nadu Coast. 1) Pulicat Lake (Brackish water lake); 2) Adyar and Cooum (Polluted Rivers); 3) Gulf of Mannar (Coral reef); and 4) Tamiraparani River (Perennial River).

the condition is reversed. Because of high rates of evaporation, low runoff and precipitation, it becomes comparable to a negative estuary.

2.1.2. *Polluted Rivers (Adyar and Cooum Rivers in Chennai)*

Chennai, the capital city of Tamil Nadu is located in the northern end of the Tamil Nadu coast (Figure 1). The concentration of industries and population (∼6 million) in the metropolitan area has led to heavy pollution of the major waterways and the adjoining coastal waters due to inadequate management of waste products.

There are three major waterways in Chennai: the Cooum and Adyar Rivers and the Buckingham Canal. The course of the Adyar River from its source to the sea is $\sim$40 km and drains a catchment area of 850 km^2. The river carries sullage, draining most of the year, resulting in severe pollution of the river while floodwaters are predominant during monsoon. The average discharge of the Adyar River is 114×10^6 m^{-3}. The mouth of the river tends to be closed by littoral drift of sand and forms lagoons. Sand bars present at the mouth of the Adyar and Cooum rivers cause stagnation and decrease the possibility of tidal flushing. This condition has also resulted in the deposition of sewage sludge and silt onto the riverbed.

2.1.3. *Perennial River/Estuary (Tamiraparani River)*

Tamiraparani River (Figure 1) is one of the perennial river basins of South India extending between latitude 8°30′ to 9°15′N and longitude 77°10′ to 78°10′E, with a drainage area of 5969 km^2 covering a distance of 150 km. Irrigation occupies 40% of the land, utilizing 90% of the water available in the basin. This basin receives an annual rainfall of 1100 mm during the southwest (June–September) and the northeast (October–December) monsoons.

2.1.4. *Coral Reefs (Gulf of Mannar)*

The coral reefs of the Gulf of Mannar are developed around a chain of 21 islands that lie along a 140 km stretch between Tuticorin and Rameshwaram (Krishnamurthy, 1987; Kumaraguru, 1997). These islands are located between latitude 8°47′ and 9°15′N and longitude 78°12′ and 79°14′E. A detailed account of each island is given in Krishnamurthy (1987) and Deshmukh and Venkataramani (1995). The islands lie on an average about 8 km from the mainland. They are a part of the Mannar barrier reef, which is about 140 km long and 25 km wide, between Pamban and Tuticorin. Different types of reef forms such as shore platform, patch, coral pinnacles and atoll type are also observed in the Gulf of Mannar. The islands have fringing coral reefs with patch reefs located on them. Narrow fringing reefs are located mostly at a distance of 50 to 100 m from the islands. On the other hand, patch reefs arise from depths of 2–9 m and extend to 1–2 km in length with width as much as 50 m. Reef flat is extensive in almost all the reefs in the Gulf of Mannar. Reef vegetation is richly distributed on these reefs. The total area occupied by the reef and its associated features is 94.3 km^2. Reef flat and reef vegetation including algae occupies 64.9 and 13.7 km^2, respectively (DOD and SAC, 1997). Visibility is affected during monsoons, coral mining and high sedimentation periods. The reefs are more luxuriant and richer than the reefs of the Palk Bay. There are about 96 species of corals belonging to 36 genera in the Gulf of Mannar. The most commonly occurring genera of corals are *Acropora, Montipora* and *Porites* (Ramaiyan *et al*., 1995).

2.2. Sample collection and laboratory analysis

Sediment cores ranging from 10 to 100 cm were collected from the coastal ecosystems mentioned above using a hand-held corer of 4.5 cm diameter. In total, 17 core samples were collected from 4 different ecosystems (Pulicat Lake: n = 8, Adyar and Cooum Rivers: n = 3, Gulf of Mannar: n = 2 and Tamiraparani River and Estuary: n = 4). The depth of the water column varied between 0.5 and 1 m. After sampling, the sediment cores were transported vertically to the laboratory and stored frozen until ready for analysis. The sediment samples were extruded from the cores and sectioned at 2 cm interval unless specified. ^{210}Pb measurements at various depths have been made. The radiochemical separation of ^{210}Pb was carried out after ashing the sediment at 400 °C. The activity of the purified ^{210}Pb was assayed in a low background β-counter, by counting the 1.17 MeV β emissions of its daughter product, ^{210}Bi. Details of the method for the chemical separation of ^{210}Pb are presented elsewhere (Subramanian and Mohanachandran, 1997). ^{226}Ra activity is obtained from the measurement of ^{222}Rn by the emanation method (Ku and Lin, 1976).

For the analysis of metals, bulk sediment samples were digested using a mixture of 0.5 mL HNO_3, 1.5 mL of HCl and 5.0 mL of HF, in Teflon® bombs, using microwave digestion technique following standard procedures (Ramesh and Anbu, 1996). The metals (Fe, Mn, Zn, Cu, Cr, Co and Ni) were determined using a Hitachi Graphite Furnace Atomic Absorption Spectrophotometer (AAS), Model Number Z 7000. Accuracy was checked using two certified sediment samples (BCSS-1 and MESS-1) obtained from the National Research Council, Canada and was found to be within 95% confidence limits of the recommended values. The overall analytical precision was ±3% for the metals analyzed.

2.3. Sedimentation model

A simple mathematical treatment was used to calculate sediment accumulation rates following Krishnaswami *et al*. (1971) and Joshi and Ku (1979). Assuming that the sedimentation rates (S in cm yr^{-1}) and the activity of excess ^{210}Pb added to the surface sediments, C_0, C (dpm g^{-1}) are constant in time, the distribution of excess ^{210}Pb is in an undisturbed sediment, governed by the relationship:

$$\ln C = \lambda/SD + \ln C_0$$

where: C_0 and C are activities of excess ^{210}Pb at the surface (C_0) and depth D (C), respectively; S is the sedimentation rate; λ is the radioactive decay constant (for ^{210}Pb the radioactive decay constant is 0.693–22.60 yr^{-1}). In graphs, $\ln C = \lambda\ SD + \ln C_0$ represents a line with slope, λ S. Thus S, the sedimentation rate was determined from the above equation.

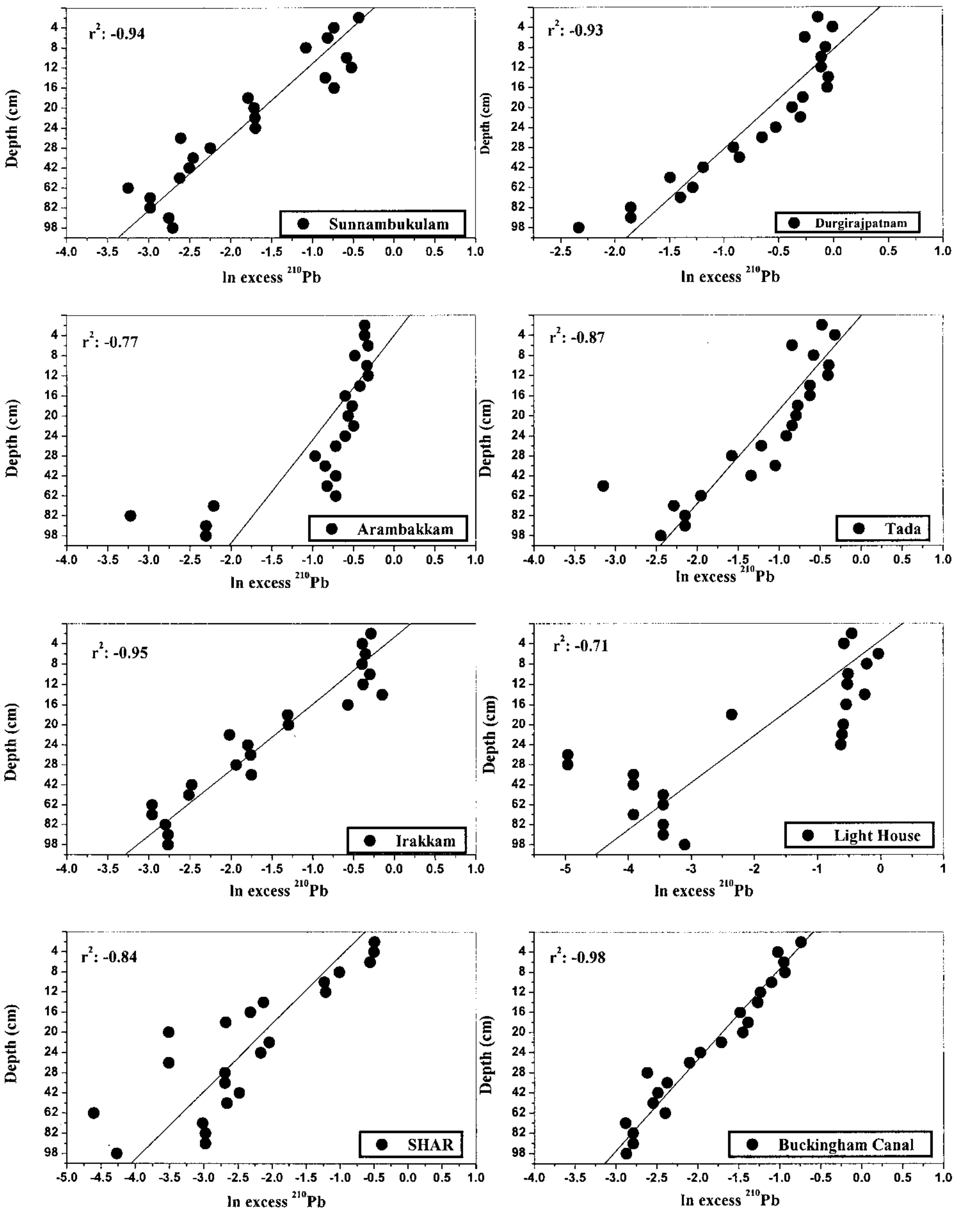

Figure 2. Measured excess ^{210}Pb (dpm g^{-1}) versus depth for core sediments from the Pulicat Lake.

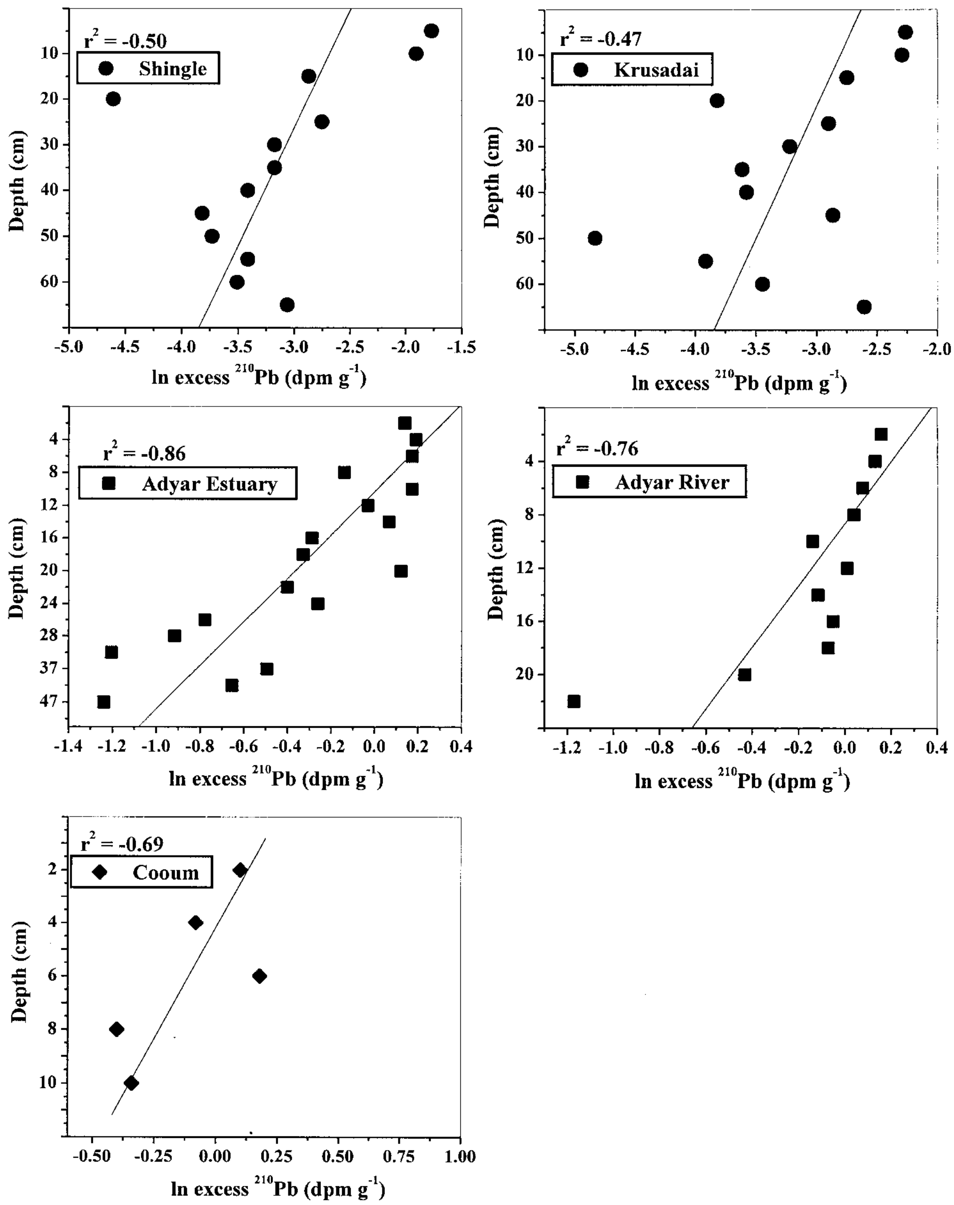

Figure 3. Measured excess ^{210}Pb (dpm g^{-1}) versus depth for core sediments from the Adyar and Cooum Rivers and Gulf of Mannar coral reef.

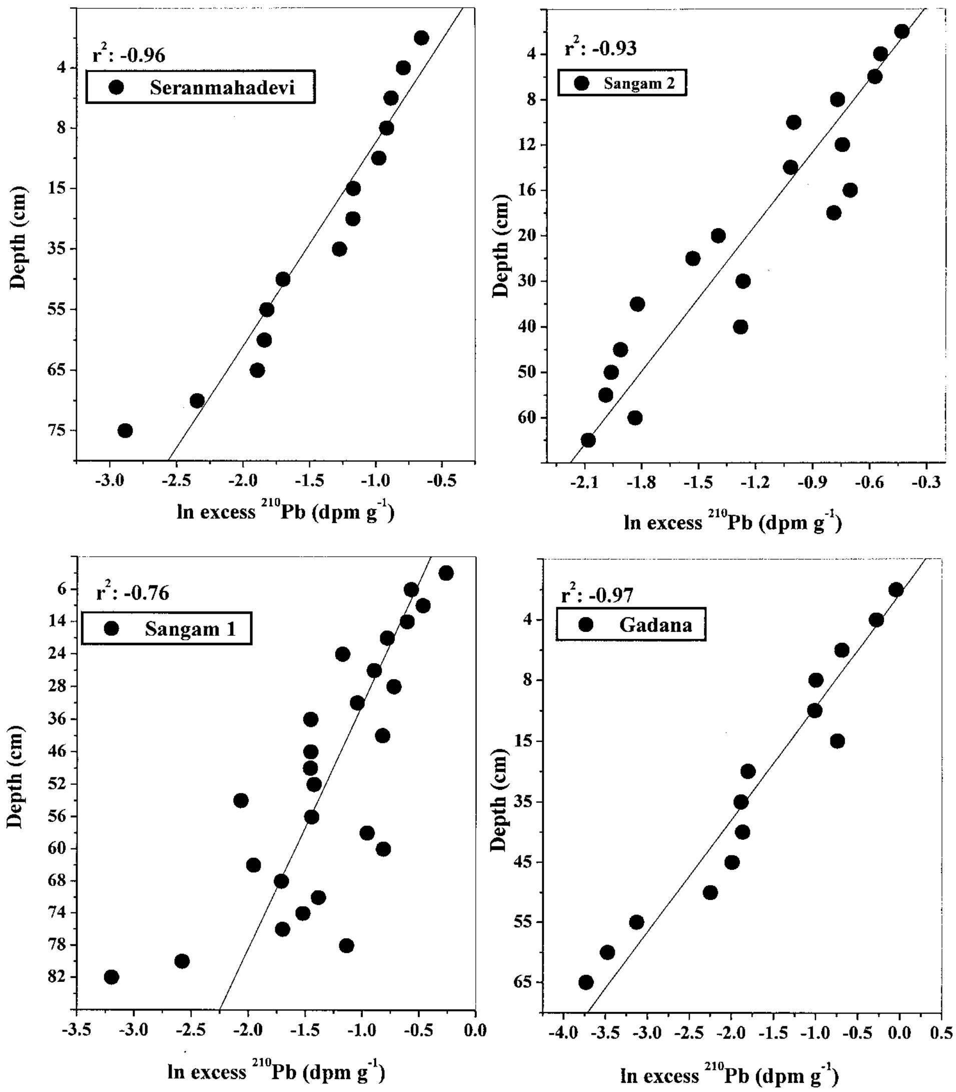

Figure 4. Measured excess ^{210}Pb (dpm g^{-1}) versus depth for core sediments from the Tamiraparani River Basin.

3. Results and Discussion

3.1. Sedimentation rates

The distribution of excess ^{210}Pb with depth for the Pulicat Lake, Gulf of Mannar, Adyar, Cooum and Tamiraparani rivers and estuarine regions are given in Figures 2 to 4 for seventeen cores. X-radiography of the cores has shown that the top layers have not been disturbed, either due to coring or due to some other natural processes such as bioturbation. Table I shows the sedimentation rates and time span estimated for the various locations in each coastal ecosystem and is discussed below.

3.1.1. *Coastal Lagoon (Pulicat Lake)*
The excess ^{210}Pb activity decreases exponentially with depth. The best-fit lines through the data points yields accumulation rates in the range between 8.29 and 15.75 mm yr^{-1}, as shown in Figure 2 and Tables I and II, respectively. The average accumulation rate for the Pulicat Lake is 12.34 mm yr^{-1}. These rates show wide variations as compared to values reported for other estuaries and for a number of freshwater and marine environments excepting Yamuna and the Gulf of Cambay. The Pulicat Lake receives discharge from the Buckingham Canal and two other rivers, the Arani and Kalangi, with sedimentation rates of 14.3, 13.7 and 15.75 mm yr^{-1}, respectively. The sedimentation rates at these discharge points are much higher as compared to the rest of the lake. Moreover, rapid sedimentation in the southern part of the lake could have also resulted from flocculation under saline conditions, in comparison to the northern part of the lake. The present study reflects an overall high rate of sedimentation, which averages 1.23 cm yr^{-1} (i.e.) approximately 1.2 m per 100 yr. An earlier work carried out by studying the fossil pollen grains preserved in sediments for a single core, reported similar results of 1 m per 100 yr (Caratini, 1994).

3.1.2. *Polluted Rivers (Adyar and Cooum)*
The sediment cores analyzed from the Adyar and Cooum rivers (Figure 3) does not show a clear exponential radioactive decay, may be due to active bioturbation prevalent in the sediments. The best-fit lines through the data points yield accumulation rates of 7.88 and 10.40 mm yr^{-1} for the Adyar River and its estuary, respectively. There is a progressive increase in the sedimentation rate towards downstream of Adyar River. The estuarine region is filled with freshly deposited sediments a large part of it is of recent origin. Intense human activities such as discharge of sewage, road and building constructions in addition to bank erosion has resulted in an accelerated rate of sedimentation. On the other hand, the rate of sedimentation in the Cooum estuary is relatively small (5.28 mm yr^{-1}) mainly due to constant dredging of the bar mouth at its confluence with the Bay of Bengal. This has enabled a connection of the Cooum River with the Bay of Bengal in recent times.

3.1.3. *Coral Reef (Gulf of Mannar)*
Coral formation in Tamil Nadu is mainly confined to the Gulf of Mannar and the Palk Bay. The reefs in the Gulf of Mannar extend from the Rameshwaram archipelago to Tuticorin over a distance of 140 km. The formation is estimated to cover an area of 100 km^2 (Wafar, 1990). The reefs are discontinuous and are mainly found around 20 small islands at a maximum depth of 6 m. The outside of the reef harbors ramose corals while the inner side has massive corals with large

TABLE I
Sedimentation rates in different coastal ecosystems of Tamil Nadu

Location/ecosystem (salinity in ‰)		Depth (cm)	Slope (r^2) (mm yr^{-1})	Time span (yr)
Pulicat Lake				
Light House	(31.30)	98	8.29 (–0.71)	118.2
Sunnambukulam	(26.77)	98	12.12 (–0.94)	80.9
Durgirajpatnam (Arani River Confluence)	(30.05)	98	13.70 (–0.93)	71.6
Arambakkam	(25.04)	98	13.13 (–0.77)	74.7
TADA (Kalangi River Discharge Point)	(27.02)	98	15.75 (–0.87)	62.2
Irakkam (Island)	(27.39)	98	10.16 (–0.95)	96.4
SHAR (Island)	(26.02)	98	11.25 (–0.84)	87.1
Buckingham Canal	(30.10)	98	14.32 (–0.98)	68.4
Cooum Estuary				
Napier Bridge	(31.00)	10	5.28 (–0.69)	18.9
Adyar River and Estuary				
Saidapet (River)	(15.00)	22	7.88 (–0.76)	27.9
Theosophical Society (Estuary)	(21.00)	47	10.40 (–0.86)	45.2
Gulf of Mannar				
Shingle Island	(33.43)	65	16.20 (–0.50)	40.1
Kurusadai Island	(31.69)	65	18.53 (–0.47)	35.1
Tamiraparani Estuary				
Gadana	(0.91)	65	6.48 (–0.97)	100.4
Serenmahadevi	(0.96)	75	13.75 (–0.96)	54.5
Sangam I	(4.80)	65	12.21 (–0.76)	53.2
Sangam II	(5.10)	82	15.51 (–0.93)	52.9

TABLE II

Mean sedimentation rates in various coastal, river and estuarine systems of India

Area/ecosystem	Mean sedimentation rate (mm yr^{-1})	Remarks	Reference
Coastal Ecosystems of Tamil Nadu			
Coastal Lagoon (Pulicat Lake)	12.34	Range for 8 cores 8.29–15.75 mm yr^{-1}	Present study
Polluted Rivers (Adyar and Cooum)	7.85	Range for 3 cores 5.28–10.40 mm yr^{-1}	Present study
Coral Reefs (Gulf of Mannar)	17.37	Range for 2 cores 16.20–18.53 mm yr^{-1}	Present study
Perennial River/Estuary (Tamiraparani)	11.99	Range for 4 cores 6.48–15.51 mm yr^{-1}	Present study
Rivers and Estuarine Systems of India			
Godavari Estuary	2.67	Range for 3 cores 0.40–4.80 mm yr^{-1}	Subramanian and Mohanachandran, 1997
Krishna River	5.50	Range for 5 cores 3.50 to 11.0 mm yr^{-1}	Ramesh *et al*., 1988
Krishna Estuary	1.39	Range for 3 cores 0.20–2.30 mm yr^{-1}	Subramanian and Mohanachandran, 1997
Cauvery Estuary	3.47	Range for 2 cores 3.20–3.80 mm yr^{-1}	Subramanian and Mohanachandran, 1997
Yamuna River (Delhi)	42.15	Range for 6 cores 5.60–82.00 mm yr^{-1}	Subramanian *et al*., 1985
Mahanadi River	7.5	Range for 5 cores 5.00–20.00 mm yr^{-1}	Chakrapani and Subramanian, 1993
Gulf of Cambay	19.00	Single core	Borole *et al*., 1982

polyps. Sedimentation on the shoreward side influences the distribution of corals. The rate of sedimentation in the Gulf of Mannar area is the highest among the coastal ecosystems of Tamil Nadu (Table I, Figure 3). At the Krusadai Island the mean sedimentation rate was 18.53 mm yr^{-1} and at the Shingle Island, the mean rate was marginally lower (16.2 mm yr^{-1}). Wafar (1990) has pointed put that the inshore waters of the Palk Bay and the Gulf of Mannar become very turbid during the northeast monsoon due to sedimentation. This causes mortality of the coral colonies on the inner side of the reef and only the species with large polyps survive.

TABLE III
Sedimentation and erosion rates in the Tamiraparani River Basin

Location	Sedimentation rate ($mm\ yr^{-1}$)	Erosion rate ($mm\ yr^{-1}$)[a]
Gadana (Upstream)	6.48	0.0036
Seranmahadevi (Midstream)	13.75	0.0180
Sangam I (Downstream)	12.21	0.0068
Sangam II (Downstream)	15.51	0.0068

[a] Erosion rates: After James, 2000.

3.1.4. *Perennial River and Estuary (Tamiraparani)*

The sediment accumulation rate in the Tamiraparani River and estuary varies from 6.48 to 15.51 mm yr^{-1} (Table I, Figure 4). When a comparison was made with the unpolluted upstream region of Tamiraparani River basin, the sedimentation rates were higher in the midstream (13.75 mm yr^{-1}) and downstream regions (12.21 to 15.51 mm yr^{-1}). The higher rate in these regions is possibly due to the effect of damming. There is also a progressive increase in sedimentation rates towards downstream (Table III). The estuarine region is filled with freshly deposited sediments, a large part of which is of recent origin. Further a sharp increase in the sedimentation rates in the estuarine region may also indicate the rapid deposition of sediments due to flocculation, occurring at the freshwater-seawater transition.

Soil erosion is a fundamental and complex natural process that is strongly modified (generally increased) by human activities such as land clearance, agriculture (plowing, irrigation, grazing), forestry, construction, surface mining and urbanization. It is an important social and economic problem and an essential factor in assessing ecosystem health and function.

Table III compares sedimentation rates and erosion rates (James, 2000) in the river basin at various locations. The spatial variation in erosion rates shown in Table III has been computed using the sediment load (tons yr^{-1}) over the catchment area of the river basin. The rate of erosion appears to be small in relation to the rate of sedimentation. The sedimentation rate varies by over an order of magnitude in all cases. The very high increase in sedimentation rate may be due to severe bank erosion and other human activities mentioned above. The Tamiraparani River is an intensely used river basin with hectic human activities such as agriculture, deforestation, grazing and dam construction at several points across the river releasing local sediments and in turn accelerating the rate of sedimentation.

The high sedimentation rate obtained for the Tamiraparani River in the estuarine region is about the same for other near-coastal regions for e.g., the Krishna delta: 11.0 mm yr^{-1} (Ramesh *et al*., 1988); Gulf of Cambay: 19.00 mm yr^{-1} (Borole *et al*., 1982). In the present study, sedimentation rates were comparable with the other Indian river systems (Table II) excepting Yamuna.

The Yamuna River near Delhi shows the maximum depositional rate of 80 mm yr^{-1}, due to solid waste discharge from the urban areas. The sedimentation rates in the Scheldt estuary, Netherlands, studied by Zwolsman *et al.* (1993) reported that the rate varied from 8.4 to 9.0 mm yr^{-1}. Similarly Ravichandran *et al.* (1995) reported sedimentation rates between 5.0 to 9.5 mm yr^{-1} for the Sabine-Neches estuary, USA. It can be concluded that the higher sedimentation rate in the estuarine region is mainly due to the settling of the sediment load from the upstream sources, high organic matter, agricultural activities and the influence of seawater.

3.2. Heavy metal distribution and deposition rates

The heavy metal contamination and accumulation rate in sediment cores have received much attention in the last two decades. Valette-Silver (1992) reviewed the available literature on the reconstruction studies and concluded that the heavy metal pollution of sediments began in early 1800s and the interest increased sharply in the 1900s, especially between 1940 and 1980. The fate of heavy metals in the aquatic environment is of extreme importance because of their impact on the ecosystem. Sediments are usually regarded as the ultimate sink for heavy metals discharged into the environment (Gibbs, 1973). In a coastal ecosystem, sediments contain significantly higher metals than the dissolved phase. For example, an average world river contains between 40 and 8.2 $\mu g\ L^{-1}$ of Fe and Mn, respectively in the dissolved phase and 48000 and 1050 $\mu g\ g^{-1}$ as particulates (Martin and Whitfield 1981). Thus, a large part of the anthropogenic discharge of heavy metals in the environment becomes part of the sediments in the coastal ecosystems, acting as efficient scavengers for the metals. Hence the present study is ideal to reconstruct the past history of metal pollution present in the coastal ecosystems. Table IV summarizes the vertical distribution of heavy metals for the core samples collected from different coastal ecosystems of Tamil Nadu. The results show that all these metals are enriched in the top few centimeters (0–10 cm) due to recent increase in anthropogenic activities and high organic matter content (3.1–9.2%, Purvaja 1995; Figures 5a and b). In the polluted rivers of Adyar and Cooum, the concentration of heavy metals were significantly higher than those observed for either the Pulicat Lake or the coral reef ecosystem. There is a consistent decrease in the metal concentration with depth. Zwolsman *et al.* (1993) have reported that the settling of particulate organic matter in suspended sediments and subsurface production by marshy plants significantly enhances the metal accumulation rate. The higher concentrations of Fe and Mn in the top 10 cm are due to diagenesis and dissolution of the oxy-hydroxides in the partly reduced sediment layer, upward migration and re-precipitation near the oxy-hydroxic interface (Shaw *et al.*, 1990).

The role of hydrous oxides of Fe and Mn in scavenging trace metals from solution is well established (Martin *et al.*, 1973; Robinson, 1981; d'Anglejan and Ramesh, 1990). This removal by 'sorption' or 'co-precipitation' is strongly pH dependant as is the formation of the host hydroxides. As these metal carriers are

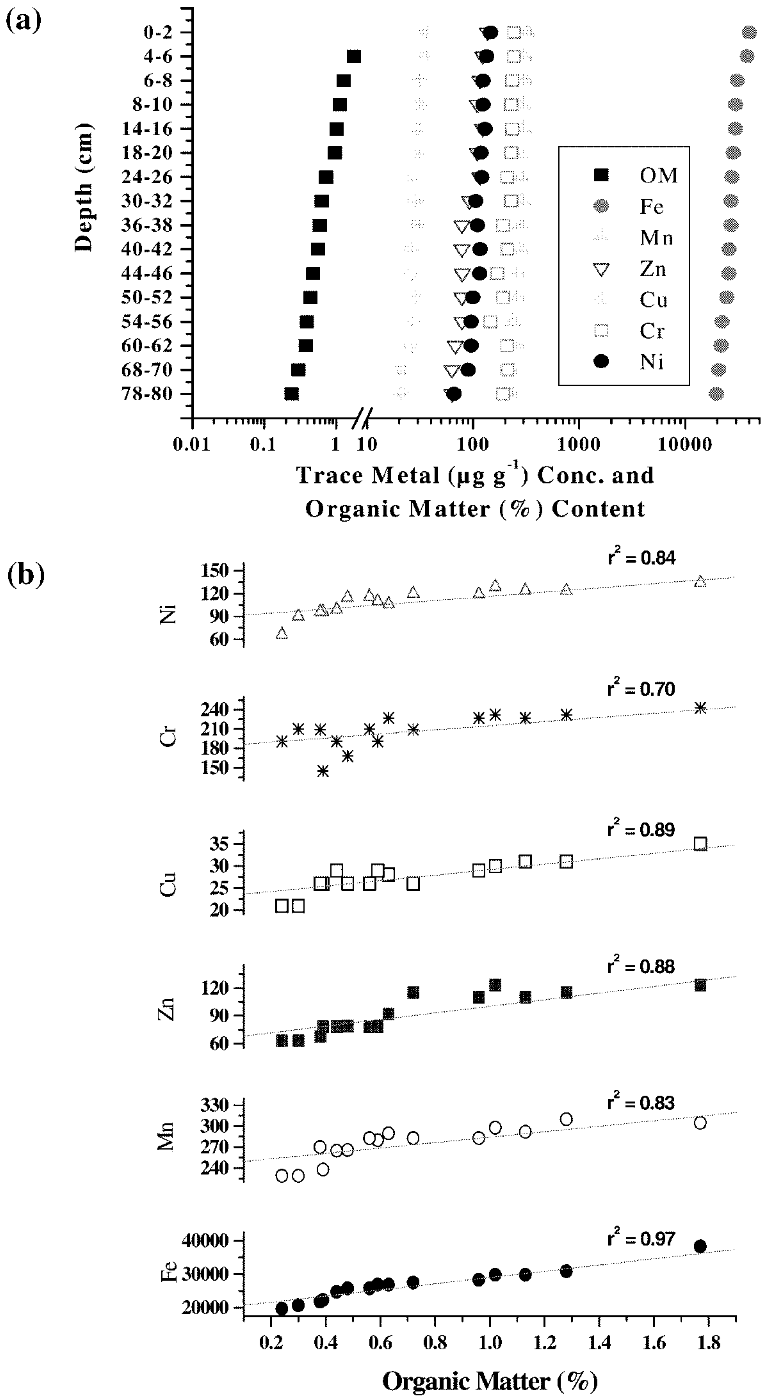

Figure 5. (a) Vertical profile in trace metal and organic matter distribution in the estuarine region (Sangam 1) of the Tamiraparani River Basin. (b) Relationship between trace metals (μg g^{-1}) and organic matter (%), their ratios in the estuarine region (Sangam 1) of the Tamiraparani River Basin.

TABLE IV

Heavy metal composition and deposition rates[a] in sediment cores from the coastal ecosystems of Tamil Nadu

Depth	Concentration (μg g^{-1})						Depositional rates (μg cm^{-2} yr^{-1})					
	Fe	Mn	Zn	Cu	Cr	Ni	Fe	Mn	Zn	Cu	Cr	Ni
Location: Pulicat Lake (Light House)												
0–2	27263	411	69	36	278	253	19889	300	50	26	203	185
6–8	25900	391	67	35	258	243	18895	285	49	26	188	177
12–14	16224	308	65	27	253	241	11836	225	47	20	185	176
18–20	15900	302	62	26	241	231	11599	220	45	19	176	169
28–30	15306	292	63	26	240	238	11166	213	46	19	175	174
38–40	15387	290	64	26	238	230	11225	212	47	19	174	168
50–52	10900	249	61	26	214	229	7952	182	45	19	156	167
70–72	12576	231	60	24	221	200	9174	169	44	18	161	146
90–92	11678	216	58	23	225	214	8519	158	42	17	164	156
96–98	10780	214	57	22	213	198	7864	156	42	16	155	144
Location: Pulicat Lake (Tada)												
0–2	20071	355	65	32	167	200	27818	492	90	44	231	277
4–6	19684	348	63	31	163	210	27282	482	87	43	226	291
8–10	18700	331	60	29	155	215	25918	459	83	40	215	298
14–16	17395	308	60	29	155	199	24109	427	83	40	215	276
26–28	18637	330	55	29	144	186	25831	457	76	40	200	258
34–36	17204	304	56	28	143	184	23845	421	78	39	198	255
48–50	14200	240	53	27	139	120	19681	333	73	37	193	166
64–66	14489	245	54	27	142	124	20082	340	75	37	197	172
80–82	13669	231	51	27	134	126	18945	320	71	37	186	175
92–94	13741	232	52	27	134	119	19045	322	72	37	186	165
Location: Pulicat Lake (Sunnambukulam)												
0–2	35205	267	57	30	212	295	37548	285	61	32	226	315
6–8	34526	262	55	29	208	289	36824	279	59	31	222	308
10–12	32691	248	54	29	197	275	34867	265	58	31	210	293
20–22	32800	249	53	28	184	274	34983	266	57	30	196	292
36–38	30176	233	28	21	197	256	32185	249	30	22	210	273
40–42	30511	232	29	22	182	253	32542	247	31	23	194	270
52–54	10200	221	30	23	133	201	10879	236	32	25	142	214
68–73	10480	229	26	21	137	197	11178	244	28	22	146	210
82-84	9819	229	27	20	140	190	10473	244	29	21	149	203
94–96	9870	220	25	19	132	190	10527	235	27	20	141	203

[a] For calculations please refer to text.

TABLE IV
(continued)

Depth	Concentration (μg g^{-1})						Depositional rates (μg cm^{-2} yr^{-1})					
	Fe	Mn	Zn	Cu	Cr	Ni	Fe	Mn	Zn	Cu	Cr	Ni
Location: Pulicat Lake (B. Canal)												
0–2	23613	403	77	35	275	333	29756	508	97	44	347	420
6–8	23157	396	75	34	270	323	29182	499	95	43	340	407
10–12	22000	367	73	33	261	310	27724	462	92	42	329	391
22–24	20465	383	73	32	260	309	25789	483	92	40	328	389
30–32	21926	380	64	30	206	289	27630	479	81	38	260	364
38–40	20240	360	62	30	202	285	25506	454	78	38	255	359
54–56	12300	341	59	28	192	271	15500	430	74	35	242	342
68–70	12551	342	59	29	192	265	15816	431	74	37	242	334
84–86	11840	318	55	30	178	256	14920	401	69	38	224	323
90–92	11903	315	54	28	177	255	15000	397	68	35	223	321
Location: Cooum Estuary (Napier Bridge)												
0–2	34600	239	48	49	118	69	16077	111	22	23	55	32
2–4	31700	230	33	49	114	65	14729	107	15	23	53	30
8–10	29500	230	29	40	109	42	13707	107	13	19	51	20
Location: Adyar River (Theosophical Society)												
0–2	31200	354	193	111	100	69	28554	324	177	102	92	63
2–4	30600	332	187	109	95	60	28005	304	171	100	87	55
10–12	28700	324	187	82	91	61	26266	297	171	75	83	56
20–22	25400	328	172	66	82	53	23246	300	157	60	75	49
45–47	24500	314	160	44	85	46	22422	287	146	40	78	42
Location: Adyar River (Saidapet)												
0–2	37900	363	255	141	146	65	26281	252	177	98	101	45
2–4	37200	319	213	126	137	61	25796	221	148	87	95	42
10–12	36900	313	180	117	128	60	25588	217	125	81	89	42
20–22	31200	266	151	96	120	55	21635	184	105	67	83	38
Location: Gulf of Mannar (Shingle Island)												
0–5	31200	378	154	49	238	NA[b]	44479	539	220	70	339	NA[b]
5–10	30600	351	135	45	235	NA	43623	500	192	64	335	NA
10–15	25900	348	127	42	225	NA	36923	496	181	60	321	NA
15–20	25600	336	126	41	217	NA	36495	479	180	58	309	NA

[b] NA = Data not available.

TABLE IV
(continued)

Depth	Concentration (μg g^{-1})						Depositional rates (μg cm^{-2} yr^{-1})					
	Fe	Mn	Zn	Cu	Cr	Ni	Fe	Mn	Zn	Cu	Cr	Ni
Location: Gulf of Mannar (Shingle Island)												
20–25	24500	335	126	41	210	NA[b]	34927	478	180	58	299	NA[b]
25–30	24800	325	126	39	208	NA	35355	463	180	56	297	NA
30–35	24200	320	98	38	206	NA	34500	456	140	54	294	NA
35–40	23500	315	98	38	200	NA	33502	449	140	54	285	NA
40–45	22100	315	92	37	192	NA	31506	449	131	53	274	NA
45–50	21700	301	89	36	197	NA	30936	429	127	51	281	NA
50–55	20200	298	85	36	198	NA	28797	425	121	51	282	NA
55–60	21000	295	83	35	196	NA	29938	421	118	50	279	NA
60–65	21300	278	74	29	198	NA	30365	396	105	41	282	NA
Location: Gulf of Mannar (Kurusadai Island)												
0–5	32000	396	142	51	245	NA	52180	646	232	83	400	NA
5–10	31600	362	125	45	245	NA	51528	590	204	73	400	NA
10–15	25700	358	137	42	236	NA	41907	584	223	68	385	NA
15–20	25800	349	125	38	235	NA	42071	569	204	62	383	NA
20–25	26200	336	120	39	211	NA	42723	548	196	64	344	NA
25–30	25800	335	125	40	210	NA	42071	546	204	65	342	NA
30–35	24500	340	89	42	212	NA	39951	554	145	68	346	NA
35–40	25900	325	95	39	198	NA	42234	530	155	64	323	NA
40–45	21500	326	98	38	196	NA	35059	532	160	62	320	NA
45–50	22500	309	96	35	200	NA	36689	504	157	57	326	NA
50–55	21200	315	98	32	205	NA	34570	514	160	52	334	NA
55–60	22700	309	85	30	201	NA	37016	504	139	49	328	NA
60–65	22400	289	71	31	197	NA	36526	471	116	51	321	NA
Location: Tamiraparani (Sangam 1)												
0–2	40200	325	136	36	243	146	54868	444	186	49	332	199
4–6	38300	305	123	35	243	134	52275	416	168	48	332	182
6–8	30900	310	115	31	232	124	42175	423	157	42	317	170
8–10	29800	292	110	31	227	124	40673	399	150	42	310	170
14–16	29800	298	123	30	232	129	40673	407	168	41	317	176
18–20	28300	283	110	29	227	119	38626	386	150	40	310	162
24–26	27500	283	115	26	209	120	37534	386	157	35	285	164
30–32	26900	290	92	28	227	106	36715	396	126	38	310	145
36–38	26900	280	78	29	191	110	36715	382	106	40	261	150
40–42	25800	283	78	26	210	116	35214	386	106	35	287	158

[b] NA = Data not available.

TABLE IV
(continued)

Depth	Concentration (μg g^{-1})						Depositional rates (μg cm^{-2} yr^{-1})					
	Fe	Mn	Zn	Cu	Cr	Ni	Fe	Mn	Zn	Cu	Cr	Ni
Location: Tamiraparani (Sangam 1)												
44–46	25800	266	79	26	168	115	35214	363	108	35	229	156
50–52	24700	265	78	29	191	99	33713	362	106	40	261	135
54–56	22300	238	78	26	145	96	30437	325	106	35	198	131
60–62	21800	270	68	26	209	96	29754	369	93	35	285	131
68–70	20700	229	63	21	210	90	28253	313	86	29	287	123
78–80	19700	229	63	21	191	66	26888	313	86	29	261	90
Location: Tamiraparani (Serenmahadevi)												
0–2	41200	531	142	47	274	151	49852	643	172	57	332	182
6–8	39300	525	135	42	262	138	47553	635	163	51	317	167
10–15	37200	468	128	39	262	134	45012	566	155	47	317	162
15–20	36800	468	124	42	257	125	44528	566	150	51	311	151
20–25	36800	453	128	35	252	129	44528	548	155	42	305	156
25–30	32800	453	124	41	248	110	39688	548	150	50	300	133
30–35	32300	407	124	42	257	114	39083	492	150	51	311	138
35–40	31900	389	128	39	239	120	38599	471	155	47	289	145
40–45	31100	424	111	32	227	129	37631	513	134	39	275	156
45–50	31900	438	98	35	207	123	38599	530	119	42	250	149
50–55	31900	424	87	31	224	102	38599	513	105	38	271	123
55–60	31100	428	98	39	227	119	37631	518	119	47	275	143
60–65	31100	389	98	32	224	99	37631	471	119	39	271	120
65–70	29700	410	64	35	207	99	35937	496	77	42	250	120
Location: Tamiraparani (Sangam 2)												
0–2	31100	389	136	48	246	209	33416	418	146	52	264	225
6–8	30900	338	123	43	242	192	33201	363	132	46	260	207
10–12	26500	327	123	41	227	186	28474	351	132	44	244	199
14–16	24800	310	111	36	208	179	26647	333	119	39	223	192
20–25	26300	327	108	38	191	173	28259	351	116	41	205	186
25–30	24200	320	123	38	227	171	26002	344	132	41	244	183
30–35	25500	323	110	32	210	179	27399	347	118	34	226	192
35–40	24900	311	108	36	197	166	26755	334	116	39	212	179
40–45	25300	305	104	32	192	152	27184	328	112	34	206	164
45–50	25400	307	97	29	208	158	27292	330	104	31	223	170
50–55	24800	298	102	32	201	165	26647	320	110	34	216	177
55–60	23700	298	98	32	198	138	25465	320	105	34	213	148

TABLE IV
(continued)

Depth	Concentration (μg g^{-1})						Depositional rates (μg cm^{-2} yr^{-1})					
	Fe	Mn	Zn	Cu	Cr	Ni	Fe	Mn	Zn	Cu	Cr	Ni
Location: Tamiraparani (Sangam 2)												
60–65	22100	292	92	32	191	142	23746	314	99	34	205	152
65–70	21200	309	78	25	210	138	22779	332	84	27	226	148
70–75	20900	298	78	29	208	95	22457	320	84	31	223	102
75–80	20700	265	63	25	227	130	22242	285	68	27	244	139
Location: Tamiraparani (Gadana)												
0–2	42000	545	152	51	275	117	23950	311	87	29	157	66
6–8	39300	532	135	39	266	107	22410	303	77	22	152	61
8–10	37800	478	132	42	265	103	21555	273	75	24	151	59
10–15	35000	472	128	36	255	96	19958	269	73	21	145	55
15–20	37800	465	129	37	251	85	21555	265	74	21	143	48
20–25	32000	435	122	40	256	88	18248	248	70	23	146	50
25–30	32100	417	125	42	245	93	18305	238	71	24	140	53
30–35	34000	448	127	38	234	99	19388	255	72	22	133	57
35–40	33000	425	125	42	232	99	18818	242	71	24	132	57
40–45	34000	428	96	34	223	95	19388	244	55	19	127	54
45–50	30300	442	96	32	225	92	17278	252	55	18	128	52
50–55	31000	432	97	31	228	79	17677	246	55	18	130	45
55–60	31000	398	94	40	208	77	17677	227	54	23	119	44
60–65	28700	378	74	30	210	76	16366	216	42	17	120	44
Location:												
Godavari Delta[c]								22	7	7	6	5
Bay of Bengal[d]								73	–	4	7	6
Deep Sea Clays[e]								500	10	20	7	20
Arabian Sea[f]								219	55	39	–	114

[c] Kalesha *et al*. (1980).
[d] Sarin *et al*. (1979).
[e] Somayajulu *et al*. (1971).
[f] Borole *et al*. (1982).

TABLE V

Relative mobility and bioavailability of trace metals (after Salomons, 1995)

Metal species and association	Mobility
Exchangeable (dissolved cations)	*High*. Changes in major cationic composition (e.g. estuarine environment) may cause a release due to ion exchange.
Metals associated with Fe-Mn oxides	*Medium*. Changes in redo conditions may cause a release but some metals precipitate if sulfide minerals is insoluble.
Metals associated with organic matter	*Medium/High*. With time, decomposition/oxidation of organic matter occurs.
Metals associated with sulfide minerals	*Strongly dependant* on environmental conditions. Under O_2-rich conditions, oxidation of sulfide minerals leads to release of metals.
Metals fixed in crystalline phase	*Low*. Only available after weathering or decomposition.

transported into the brackish environment of estuaries, they tend to flocculate and settle out of solution.

In general, decline in metal concentration with depth may be due to the recent increase in human activities since industrialization. The heavy metal data presented here (Table IV) represents the accumulation of metals over a time span of 20 to 120 yr. This clearly reflects the fact that metal accumulation has enhanced since the pre-industrial time to recent with minor fluctuations. The other factor responsible for minor variations in metal concentration within the vertical profile is attributable to the grain size and the organic matter content as observed for core collected from the Tamiraparani River basin (as an example, please see Figures 5a and b). Elevated concentrations of Cr and Ni are recorded in the Tamiraparani River and its estuary and in the Pulicat Lake, in comparison to Zn and Cu (Table IV). In the polluted rivers and in the brackish water lake environments, most Zn and Cu are scavenged by non-detrital carbonate minerals, organic matter and oxide minerals and is less mobile than Cr and Ni.

3.3. Mobilization of trace metals and association with organic matter

The aquatic sediments constitute the most important reservoir or sink of metals and other pollutants. However, due to diagenetic processes, the sediment bound metals and other pollutants remobilize and be released back to overlying waters. The relative mobility and bioavailability of trace metals analyzed in this study are shown in Table V. Metals associated with carbonate minerals in sediments constitute the carbonate fraction, which can be newly precipitated in soils. The organic fraction consists of metals bound to various forms of organic matter. As an example, the depth-wise distribution of organic matter (OM) and trace metals such as Fe, Mn, Cr, Cu, Ni and Zn in the Tamiraparani River basin has been made (Figure 5a). The organic matter content along the profile varied from 0.24 to 2.42% with the highest content in the top sediment layer. A consistent decline with depth has been observed as also the trace metals. The metal – OM ratios in the study ($r^2 > 0.70$) indicated a strong positive correlation between the two (Figure 5b).

It is believed that in recent organic carbon-rich sediments, trapped porewater can commonly from a reducing anoxic environment. During diagenesis much of the non-silicate bound fraction of potentially toxic metals as Cu, Zn, Pb and Hg can be co-precipitated with pyrite, forming insoluble sulfides and become unavailable to biota (Morse, 1994). Seasonal variations in flow rates or storms that induce an influx of oxygenated seawater can result in rapid reaction of this anoxic sediment and thereby release of significant proportions of these metals.

The Fe and Mn oxide fractions consist of metals adsorbed to Fe-Mn oxide particles or coating. In the coastal ecosystems of Tamil Nadu, the correlation between Fe and Mn (Figure 6) show a strong positive correlation (e.g. for Tamiraparani, the r^2 was > 0.86; (n = 4 cores). The correlation between Fe and other trace metals was also made for all the coastal ecosystems. It was found that in the sediment cores from the Gulf of Mannar and Tamiraparani River, Fe showed strong positive correlation ($r^2 > 0.70$ for Gulf of Mannar and $r^2 > 0.82$ for Tamiraparani) with metals such as Mn, Cu, Cr and Zn (Figure 7), while Ni showed a weak negative correlation. Thus, we can conclude that iron is a good natural sorbent and has a great effect on the above mentioned trace metals. However, at sites intensely impacted by human activities such as the Adyar and Cooum Rivers and the Pulicat Lake, Fe showed a weak positive correlation ($r^2 > 0.50$) with the other trace metals.

3.4. Metal deposition rates

In order to determine the deposition rate of elements, the rates of sedimentation are essential. In the present study, using the elemental concentration of the cores, the accumulation rates as measured by the excess ^{210}Pb method and the *in situ* density of the core material (0.88 g cm^{-3}), the detrital deposition rates of metals (Table IV) are computed for the past 20 to 120 yr using the formula provided below:

$$x\ (\mu\text{g g}^{-1}) * \text{Density of the sediment (g cm}^{-3}) * \text{Sedimentation rate (cm yr}^{-1})$$

where: x = metal concentration (For e.g. concentration of Fe at Pulicat Lake Light House = 27263 μg g^{-1}).
Density of the sediment = 0.88 g cm^{-3}.

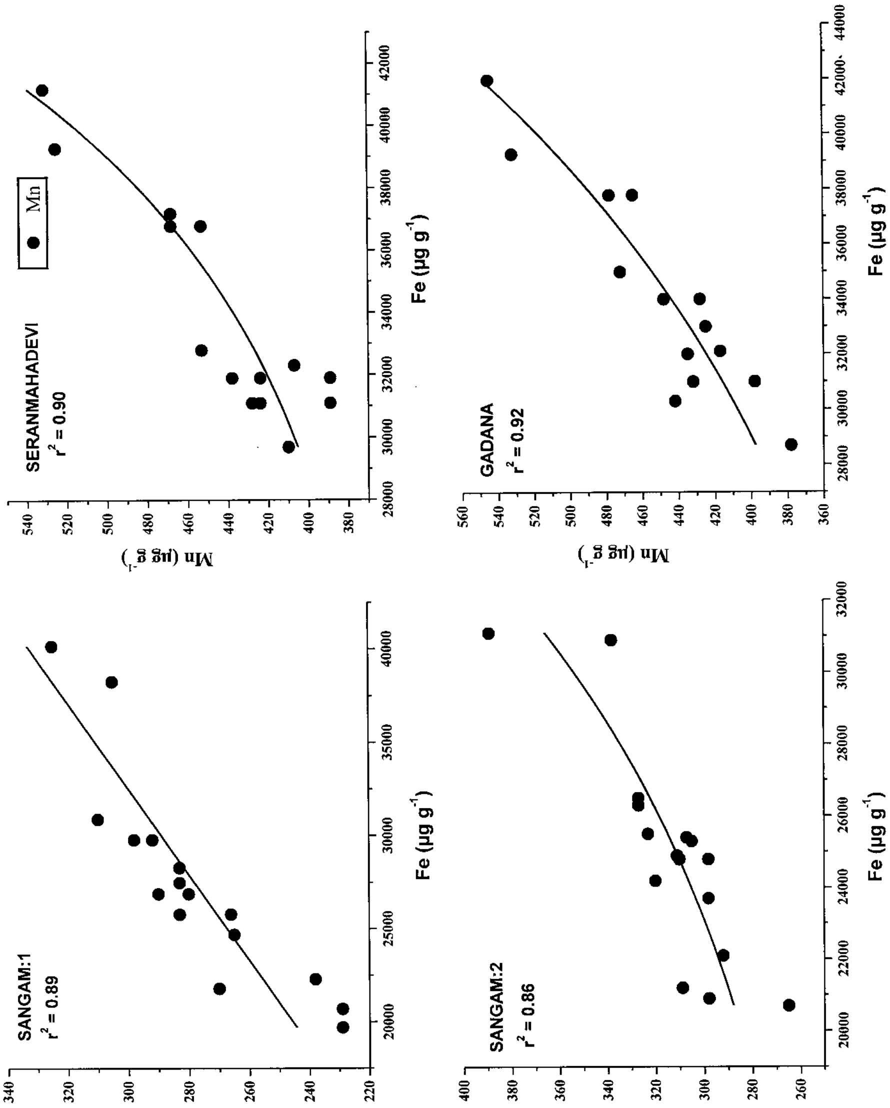

Figure 6. Fe-Mn ratios in four different cores of the Tamiraparani River Basin.

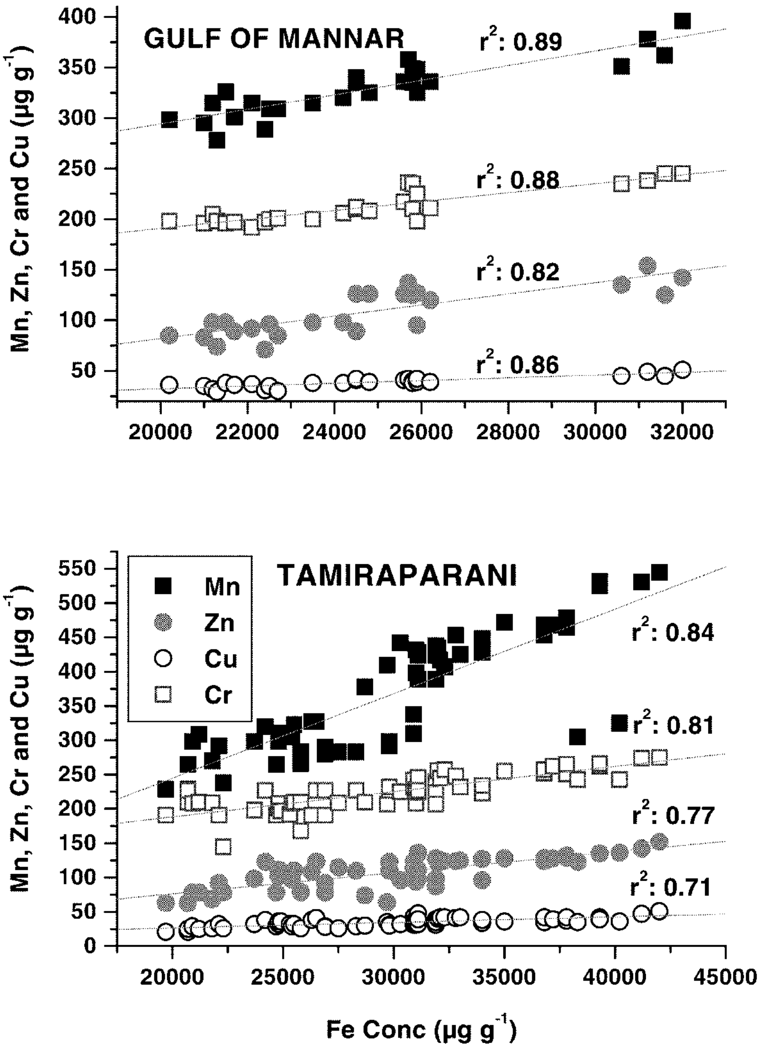

Figure 7. Correlation between Fe with other trace metals (Mn, Cu, Cr and Zn) in the Tamiraparani River Basin and Gulf of Mannar ecosystems of Tamil Nadu.

Sedimentation rate at a given location (e.g. Pulicat Lake: Light House = 0.829 cm yr^{-1}). Therefore the metal deposition rate of Fe at the Pulicat lake: Light House = 27 263 $*$ 0.88 $*$ 0.829 = 19 880 μg cm^{-2} yr^{-1} (OR) 0.19 g cm^{-2} yr^{-1}.

It is seen that the deposition rates of the measured elements in the coastal ecosystem are high compared with such rates determined for the Godavari delta, Bay of Bengal and for the deep oceans (Table IV). Borole *et al.* (1982) reported high deposition rates in the Gulf of Cambay when compared to deltaic and deep oceans. Probably such rates can only be observed in river basins and river mouths. It is also seen that for any element the variations of the deposition rate is minimal. On the other hand, Ramesh *et al*. (1988) observed large variations in deposition rates in cores collected within the Krishna river basin. Similar observations have been made in samples collected for Tamiraparani River. Such variations are expected in river basin where the nature of the suspended load varies to a large extent due to the confluence of tributaries and several other human activities. Similar studies for various coastal ecosystems and deltaic regions draining into the Bay of Bengal will help us estimate the average input of detrital material into the coastal zone and oceans.

4. Conclusion

Atmospherically delivered radionucleids such as ^{210}Pb and ^{137}Cs have been widely used over the past two decades as chronometers for estimating accumulation rates in marine/estuarine sediments. In this study, for the first time, the sediments and metal accumulation rates have been estimated for a variety of coastal ecosystems. The high rates of sedimentation in these coastal ecosystems reflect the influence of both chemical processes such as sorption (adsorption, flocculation, precipitation etc.) and human intervention. The vertical distribution of trace metals also highlight the impact of human additions to coastal ecosystems.

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