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RESEARCH

Heavy metal pollutants and their spatial distribution in surface sediments from Thondi coast, Palk Bay, South India

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Abstract

Background: The concentration of heavy metals and their spatial distribution in surface sediments collected from the Thondi coast, Palk Bay, South India were analysed in this study. The sediment grain size, pH, EC, and major elements (Fe, and Al), heavy metal concentrations (Mn, Cr, Zn, Cd, Ni, Cu, and Pb) were determined and the values for the geoaccumulation index (I_{geo}), enrichment factor (EF), potential contamination index (C_p), potential ecological risk index (RI), contamination factor (CF), modified contamination degree (mC_d), degree of contamination (C_d), and potential contamination factors (C_p) were calculated based on their background values to determine the pollution level of the study area. Multivariate analysis such as Pearson's correlation coefficient, principal component analysis/factor analysis (PCA/FA), cluster analysis, and regression analysis are a versatile method for identifying heavy metal sources and determining the relationship between pollutants in marine sediment.

Results: The pollution indices, namely EF, CF, C_d, mC_d, C_P, RI, and I_{geo}, revealed that the heavy metal contamination was due to Cd, while a moderate level of contamination was caused by Cu, Zn, Pb, and Cr. The principal component analysis and correlation matrix analysis showed a strong positive loading for Cd due to its high level of contamination in the study area. Anthropogenic inputs such as municipal wastewater, domestic sewage discharge, fishing harbour activities, and industrial and aquaculture wastes led to the increased Cd concentration in the study area. Moreover, the pollution load index revealed that the sediments were polluted by heavy metals.

Conclusion: The findings of this study revealed that the increased concentration of heavy metals in the study area increases the toxicity in the marine environment, thus affecting the ecosystem.

Keywords: Heavy metals, Toxicity, Pollution indices, Marine sediments

Background

The presence of toxic heavy metal pollutants in the aquatic ecosystem is mainly introduced through various natural and anthropogenic sources. Some of the main natural sources include the weathering processes (rocks and soils), atmospheric deposition of particles, and aeolian sediments. Anthropogenic sources, on the other hand, include sewage waste dumping, mining activities, agricultural activities, discharge of industrial wastes into water bodies, and many other human activities that discard metal pollutants into the aquatic environment [1–4]. Anthropogenic sources have a high impact on the accumulation of heavy metal pollutants in the marine environment. The heavy metals are continuously accumulated in the rivers and deposited in the marine sediment as a sink. The major issues related with the persistence of heavy metals are toxicity, bioaccumulation, and biomagnification, which lead to indelible effects on the ecosystem, human health, and other living organisms [1, 5–9]. Therefore, it is essential to assess the distribution of these

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pollutants and their level of contamination to construe the mechanism of accumulation and transportation of these pollutants into the aquatic environment as well as obtain necessary information for the supervision, maintenance, and use of coastal areas.

In recent years, the increasing level of heavy metals detected in the sediment bed has become a major concern [10–13]. Several studies have revealed that marine sediments are highly polluted due to these heavy metals [14–16]. Therefore, the evaluation of heavy metal distribution in the surface sediments is useful for determining the pollution levels in the marine ecosystem of the southeast coast of India. In previous studies, many researchers focused on the heavy metal distribution and pollution status of the marine sediments (near the shore and shelf) in various regions along the Bay of Bengal in India [17–28]. This study, however, investigates the heavy metal pollution levels of the surface sediments along the Thondi coast, Bay of Bengal, South India. Therefore, it is essential to consider the key factors such as spatial distribution, sediment quality assessment, and concentration of heavy metal pollutants in the study area.

In this study, 24 surface sediment samples were collected around the coastal area of Thondi and analysed for sediment types and chemical composition. The main objectives of this study are to (1) measure the concentration of major elements (Fe, and Al), heavy metals (Mn, Cr, Zn, Cu, Cd, Ni, and Pb) in the study area; (2) assess the level of heavy metal contamination using enrichment factors (EF), potential contamination index (C_p), geoaccumulation index (I_{geo}), potential ecological risk index (PERI), contamination factor (CF), modified contamination degree (mC_d), degree of contamination (C_d), and potential contamination factors (C_p); and (3) identify the relationship between the contaminants in the sediment and their possible sources in the study area using Pearson's correlation coefficient, principal component analysis/ factor analysis (PCA/FA), cluster analysis, and regression.

Description of the study area

Thondi lies within the latitude of 9° 43′ 26″ N and longitude of 79° 02′ 55″ E and is situated in the Palk Bay, Tamil Nadu, South India (Fig. 1a). The Palk Bay area is known for its rich marine biodiversity and resources such as seagrass, shrimps, seaweeds, lobsters, mollusks, coelenterates, holothurians, echinoderms, crabs, shellfishes, squids, and finfish. Seagrasses play a vital role in the production of commercially valuable fish in this region as it provides food and shelter for various marine organisms and is involved in the recycling of nutrients. The land use and land cover are classified as agriculture land (75%), built-up land (5%), wastelands (7%), and water bodies (13%) in the study area (Fig. 1b). This region generally receives rainfall from the north-east and south-west monsoons. The shore water has an average depth of 1–2 m and the seawater is rich in nutrients with moderately high turbidity. The wave action along the Thondi coast is minimal and the sediments are muddy. Since the area serves as a treasure of various economically important marine resources, many socioeconomic and developmental activities such as agriculture, aquaculture, and fishing are performed. Due to these economic activities, the coastal areas receive an abundance of untreated solids and liquid waste. This area is rich in valuable marine algae such as the marine brown algae (*Turbinaria conoides* and *Sargassum whitti*), red algae (*Gracilaria edulis, Hypnea musciformis*, *G. verrucosa*, *G. corticata*; *Sarconema filiforme*, *Kappaphycus alvarizii*, and *Acanthophora muscoides*), and green algae (*Ulva lactuca*, *U. reticulata*, *Caulerpa scalpelliformis*, and *Chaetomorpha linum*).

Materials and methods

Surface sediment sample collection

In total, 24 samples of surface sediments were collected from the bottom of the water at various depths of 1–2 m from eight transects around the shelf zone of the research area during January 2020. The surface sediment samples were obtained using a Van Veen grab surface sampler. Stations 1, 4, 7, and 10 were located near the estuaries, boating areas, fish market, and other areas affected by various anthropogenic activities. Several sampling sites were selected to cover the entire study area. The samples were kept in a plastic container, packed in a cooler bag at 4 °C, and transported to the research laboratory for sample processing. The texture, organic matter, and heavy metal concentrations of the surface sediment samples were analysed using standard procedures.

Texture analysis

Textural analyses were performed in the laboratory using the sieving and pipetting method. For the removal of organic matter, sediment samples were initially pretreated with H_2O_2 solution and the samples were wet-sieved in a mechanical sieve shaker using a 62 µm-sized mesh for 15 min. The samples that passed through the sieve shaker were identified as mud, while the retained sample on the sieve was identified as sand. The finer fraction of the mud was identified as silt and clay (> 0.063 mm) and determined using the pipette method. The sediment texture was classified based on the mud content classification proposed by [29, 30] and the modified classification by [31].

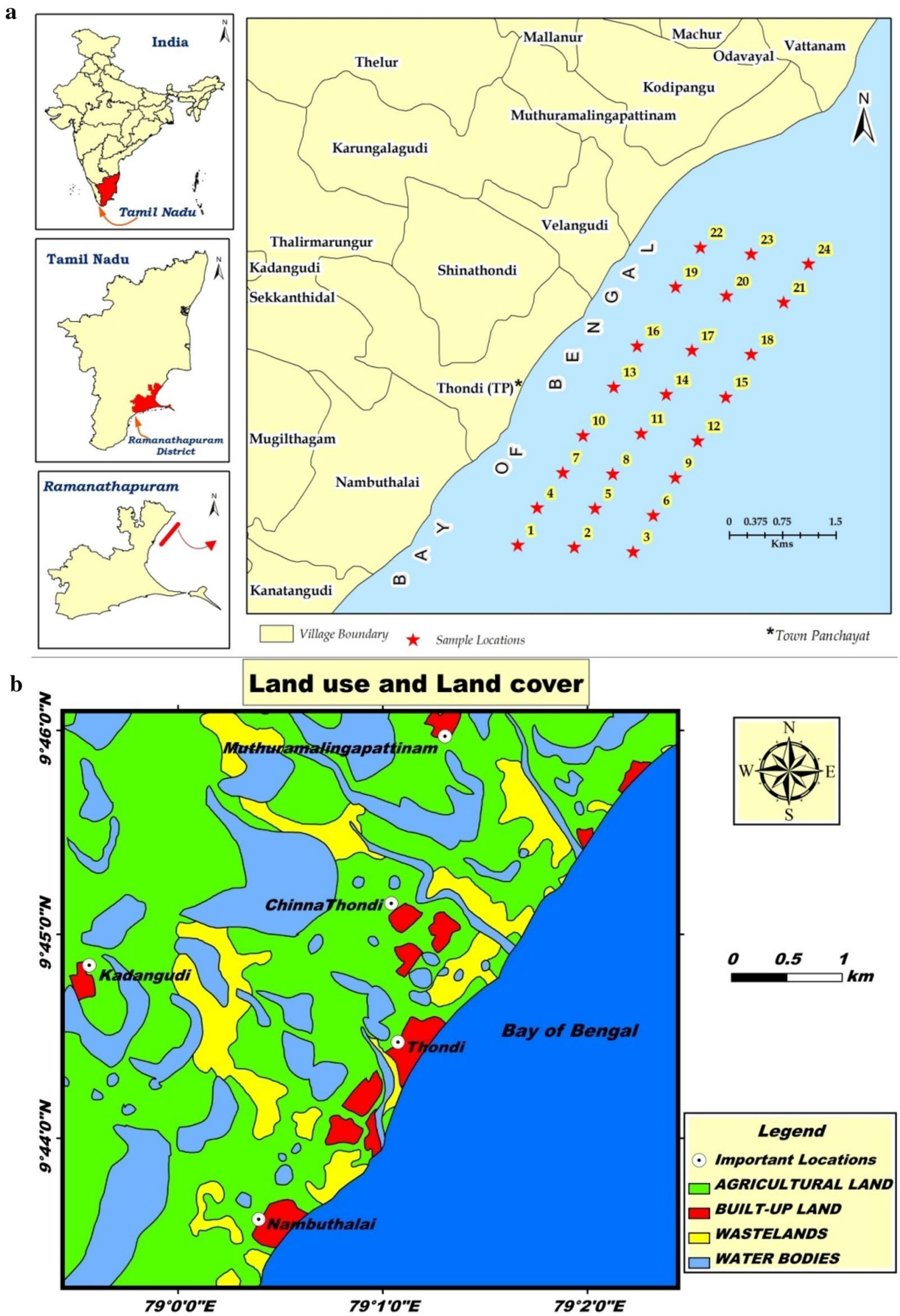

Fig. 1 **a** Location of the research area with designated sampling points. **b** Map of land use and land cover of the study area

Geochemical analysis

Approximately 1 g of surface sediment sample was treated with HNO_3, 30% H_2O_2 and HCl to determine the concentration of elements (Mn, Al, Cr, Cu, Pb, Ni, and Zn) according to the 3050B method [32]. After sample preparation, the measurement of metal concentrations was performed using ICP-MS located at the NGRI-CSIR analytical instrument facility in Hyderabad. Cd and Fe were measured separately using a flame atomic absorption spectrometer. In this study, the accuracy of the process with that of the analytical procedures was compared using reference sediment materials (MESS-1) provided by the National Research Council of Canada. By comparing the measured and certified values, the recovery values of the elements were as follows: Fe (97.93%), Al (98.42%), Mn (95.70%), Cr (97.37%), Cu (96.41%), Ni (94.78%), Cd (92.22%), Pb (100%), and Zn (97.91%). The inaccurate percentage was less than 4%.

Assessment of sediment pollution levels

The level of heavy metal contamination from both natural and anthropogenic sources in the Thondi coast, Palk Bay of Tamil Nadu, Southeast India was determined based on the complete assessment of sediment samples in the study area. Seven measurements, namely EF, CF, C_d, mC_d, RI, I_{geo}, and C_p, were used to obtain the relative pollution level of the sampling sites.

Enrichment factor

Enrichment factor (EF) is used to analyse the impact of anthropogenic sources in the sediment and the level of contamination in the study area. The geochemical normalisation of the sediment heavy metal data in relation to the content of conservative elements such as Al was used to identify the anomalous metal concentration [33–36]. The estimation of EF is based on the assessment of the trace element enrichment in the sediment [37]. It is defined based on the following formula [38]

$$\mathrm{EF} = (\mathrm{Cx}/\mathrm{Al})_{\mathrm{sample}} \Big/ (\mathrm{Cx}/\mathrm{Al})_{\mathrm{background}}, \quad (1)$$

where Cx_{Sample} and $Cx_{background}$ represent the concentration of selected metals (Fe, Mn, Cr, Cu, Ni, Cd, Pb, and Zn) in the sediment samples. $(Cx/Al)_{background}$ is the ratio of the background values of Al. The EF values for Fe (56,300%), Al (8.23%), Mn (950 $mgkg^{-1}$), Cr (100 $mgkg^{-1}$), Cu (55 $mgkg^{-1}$), Ni (75 $mgkg^{-1}$), Cd (20 $mgkg^{-1}$), Pb (12.5 $mgkg^{-1}$), and Zn (70 $mgkg^{-1}$) that were previously reported for sedimentary rocks were used as background values [39]. The results obtained were indicative of different levels of pollution. The elemental enrichment classification of the sediment is based on the following: (i) 0–1 = background concentration or no enrichment; (ii) 1–3 = minor, (iii) 3–5 = moderate, (iv) 5–10 = moderately severe, (v) 10- 25 = severe, (vi) 25–50 = very severe, and (vii) > 50 = extremely severe.

Contamination factor

Ref. [40] proposed the use of CF to assess the contamination status of the surface sediment based on the following equation:

$$\mathrm{CF} = C_{\text{metal concentration in sediment}} / C_{\text{background background value of metal}}. \quad (2)$$

The CF values according to the four classes are depicted as follows: (i) CF < 1 = low, (ii) 1 < CF < 3 = moderate, (iii) 3 < CF < 6 = considerable, and (iv) CF > 6 = very high.

Degree of contamination

The degree of contamination (C_d) represents the sum of all the CF values for all the sampling sites. It was previously proposed by [40] as shown below:

$$C\mathrm{d} = \sum_{\mathrm{i}=1}^{8} \mathrm{CF}. \quad (3)$$

The degree of contamination is depicted as follows: (i) $C_d < 6$ = low, (ii) $6 < C_d < 12$ = moderate, (iii) $12 < C_d < 24$ = considerably high, and (iv) $C_d > 24$ = high.

Modified contamination degree

Modified contamination degree is the sum of all contamination factors for the element samples to the number of elements analysed. This measure was proposed by [41] to investigate an unlimited number of heavy metals and is represented in Eq. 4:

$$\mathrm{m}C\mathrm{d} = \frac{\left(\sum_{i=1}^{i=n} \mathrm{CF}\right)}{n}, \quad (4)$$

where n is the number of analysed elements and i = i^{th} element (or pollutant) examined and contamination factor (CF). Modified contamination degree classifies the contamination level of sediment based on the following quantitative values: (i) mC_d < 1.5 = nil to very low, (ii) 1.5 ≤ mC_d < 2 = low, (iii) 2 ≤ mC_d < 4 = moderate, (iv) 4 ≤ mC_d < 8 = high, (v) 8 ≤ mC_d < 16 = very high, (vi) 16 ≤ mC_d < 32 = extremely high, and (vii) mC_d ≤ 32 = ultra-high.

Pollution load index

The pollution load index (PLI) of a specific site or a zone is assessed according to the index described by [42]. This tool is used to assess the heavy metal pollution [43] and is calculated based on the formula shown below:

$$\text{PLI for a station} = \sqrt[n]{\text{CF1} \times \text{CF2} \times \text{CF3} \ldots\ldots\ldots \text{CFn}}, \quad (5)$$

where n is the number of heavy metals, CFs, CF is $C_{metal}/C_{background}$, (C_{metal}) corresponds to the metal concentration of the sample, and $C_{background}$ is the background metal concentration:

$$\text{PLI for zone} = \sqrt[n]{\text{Station 1} \times \text{Station 2} \ldots\ldots \times \text{Station } n}. \quad (6)$$

Potential ecological risk index (RI)

Potential ecological risk index evaluates the environmental behaviour and characteristics of heavy metal contaminants in the sediments. This method was previously proposed by [40] and its primary objective is to specify the agents that cause contamination. The RI is the summation of all risk factors for the detection of heavy metal contaminants in the sediment. The RI is calculated based on the following equations:

$$C_r^i = \frac{C^i}{C_n^i}, \quad (7)$$

$$E_r^i = T_r^i \times C_r^i, \quad (8)$$

$$RI = \sum_{i=1}^{n} T_r^i \times C_r^i = \sum_{i=1}^{n} T_r^i C^i / C_n^i, \quad (9)$$

where RI is the total potential ecological risk of individual heavy metal; E_r^i is the potential ecological risk of individual heavy metal; and T_r^i= toxic response factor which represents the toxicity of a particular trace element. Hakanson proposed a standardised toxic response factor of 5, 1, 2, 5, 5, and 30 for Cu, Zn, Cr, Ni, Pb, and Cd, respectively. C^i is the measured concentration of metal n in marine sediments; and C_n^i is the standard value of metal n in the marine sediments. The conditions used to denote the risk factors and RI according to [40] are classified into nine categories of ecological risk as follows: (i) <40 = low, (ii) $40 < E_r^i < 80$ = moderate, (iii) $80 < E_r^i < 160$ = considerably high, (iv) $160 < E_r^i < 320$ = high, (v) $E_r^i > 320$ = very high, (vi) RI < 95 = low, (vii) $95 <$ RI < 190 = moderate, (viii) $190 <$ RI < 380 = considerably high, and (ix) RI > 380 = very high.

Geoaccumulation index (Igeo)

I_{geo} is used to analyse the level of pollution of trace elements and the contamination degree in marine sediments [44]. It was initially described by [46] as follows:

$$I_{geo} = \log_2[(C_n/(1.5 \times B_n)], \quad (10)$$

where C_n = the trace metals calculated (measured concentrations of the sediment samples, respectively) and B_n = background value (average value of crustal abundance) of a particular element. To decrease the possibility of variation in the background values for a specific trace element in the environment and minor anthropogenic influences, the concentration of each geochemical background value is multiplied by the factor of 1.5 [45]. The sediment classification is based on the I_{geo} value [46] as follows: (i) $I_{geo} > 5$ = extreme contamination, (ii) 4–5 = strong to extreme contamination, (iii) 3–4 = strong contamination, (iv) 2–3 = moderate to strong contamination, (v) 1–2 = moderate contamination, (vi) 0–1 = uncontaminated to moderate contamination, (vii) < 0 = uncontaminated.

Potential contamination index

The potential contamination index is calculated using the formula described below:

$$Cp = \frac{(\text{Metal})_{\text{sample Max}}}{(\text{Metal})_{\text{background}}}, \quad (11)$$

where $(\text{Metal})_{\text{sample Max}}$ is the highest concentration value of an element in the sediment, and $(\text{Metal})_{\text{Background}}$ represents the background concentration value of the element. This method was proposed by [47, 48], whereby the C_p values are classified into three levels of contamination: (i) $C_p < 1$ = low, (ii) $1 < C_p < 3$ = moderate, and (iii) $C_p > 3$ = severe.

Statistical analysis

Multivariate statistical analysis method such as Pearson's correlation coefficient, PCA, and Cluster analysis were used to determine the relationship between the contaminants in the sediment and their potential sources. The statistical software IBD-SPSS (version 20.0) was employed in this present study.

GIS analysis

The inverse distance weighted (IDW) approach using ArcGIS 10.2 software was employed for the analysis of the spatial distribution characteristics of heavy metals in the sediments.

Results

The results of the physicochemical parameters analysed during sample collection in the study area are displayed in Table 1. In total, 24 samples of surface sediment were collected from the Thondi coast of Palk Bay for analysis. The basic descriptive statistics (min, max, average, and

Table 1 Physicochemical parameters of the Thondi coastal seawater

S. no.	Physicochemical parameters	Average values
1	EC (µs)	33.9
2	Salinity (psu)	32.9
3	pH	7.85
4	TDS (ppm)	17.6
5	Atmospheric temperature (˚C)	30.6
6	Surface water temperature (˚C)	27.6
7	DO (mg/l)	5.40
8	BOD (mg/l)	0.99

SD) for the heavy metal concentrations measured at 24 locations are summarised in Table 2.

Grain size

The shelf region of the study area contains two different types of sediment, namely slightly muddy sand and muddy sand. The samples collected from stations 4, 5, 6, and 7 were slightly muddy sand and those collected from stations 1, 2, 3, 8–24 were muddy sand sediments (Fig. 2).

Enrichment factors

The EF values for the heavy metal contaminants in the sediments are listed in Fig. 3 and Table 3. The EF value of the surface sediments in the study area was 0.41—1.09 (average 0.65) (Fe), 0.30–0.89 (average 0.50) (Mn), 1.25–3.38 (average 2.00) (Cr), 0.41–1.31 (average 0.67) (Cu), 0.18—0.40 (average 0.25) (Ni), 1.52—9.97 (average 4.12) (Cd), 0.52–1.21 (average 0.76) (Pb), and 1.77–3.98 (average 2.48) (Zn). Cd displayed the highest EF value among the eight metals investigated and was classified as moderately severe enrichment with an average value of 4.12. Zn and Cr displayed a minor enrichment (average values of 2.48 and 2.00, respectively), while Ni, Mn, Fe, Cu, and Pb were classified as no enrichment (average values of 0.25, 0.50, 0.65, 0.67, and 0.76, respectively).

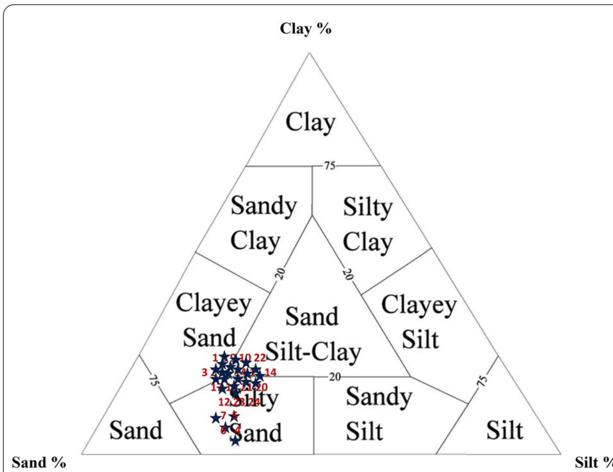

Fig. 2 Shepard classification of the sediments in the study area

Contamination factor

The contamination factor (CF) values for the heavy metals in the sediments are shown in Table 3. The average CF value for Ni (0.37), Mn (0.72), Fe (0.94), and Cu (0.99) was < 1, thus indicating that the sediment samples had a low level of contamination. The average CF value of Pb (1.13) and Cr (2.90) was 1–3, thereby indicating that the sediments were moderately contaminated. The average CF value of Zn (3.61) and Cd (5.83) was 3–6 and this indicated that the sediments had considerable contamination. In this study, the average CF values of heavy metals were ranked based on the following order: Ni < Mn < Fe < Cu < Pb < Cr < Zn < Cd. The changes in CF values at different locations are displayed in Fig. 4.

Geoaccumulation index

The calculated I_{geo} values of the heavy metals are shown in Fig. 5 and Table 3. The I_{geo} value of the surface sediments in the study area was 9.20–9.37 (average 9.29) (Fe), 5.43–5.75 (average 5.63) (Mn), 4.19–4.4 (average 4.28) (Cr), 3.06–3.49 (average 3.3) (Cu), 3–3.25 (average 3.1) (Ni), − 1.13 to − 0.49 (average − 0.8) (Cd), 1.93–2.21 (average 2.1) (Pb), and 4–4.14 (average 4.1) (Zn). The results were classified as "extremely contaminated" for Fe,

Table 2 Concentration of heavy metals in the sediments (min, max, average, SD)

Elements (mg kg^{-1})	Fe	Mn	Cr	Cu	Ni	Cd	Pb	Zn
Minimum	42,516	425	231	31	20.1	0.6	10.2	214
Maximum	62,413	896	378	84	35.6	2.5	19.5	298
Average	52,802.3	686.1	290.3	54.7	27.7	1.2	14.1	252.9
SD	6213.3	141	42.3	16.1	3.0	0.5	2.5	21.8
CV [56]	56,300	950	100	55	75	20	12.5	70

SD standard deviation, *CV* crustal value

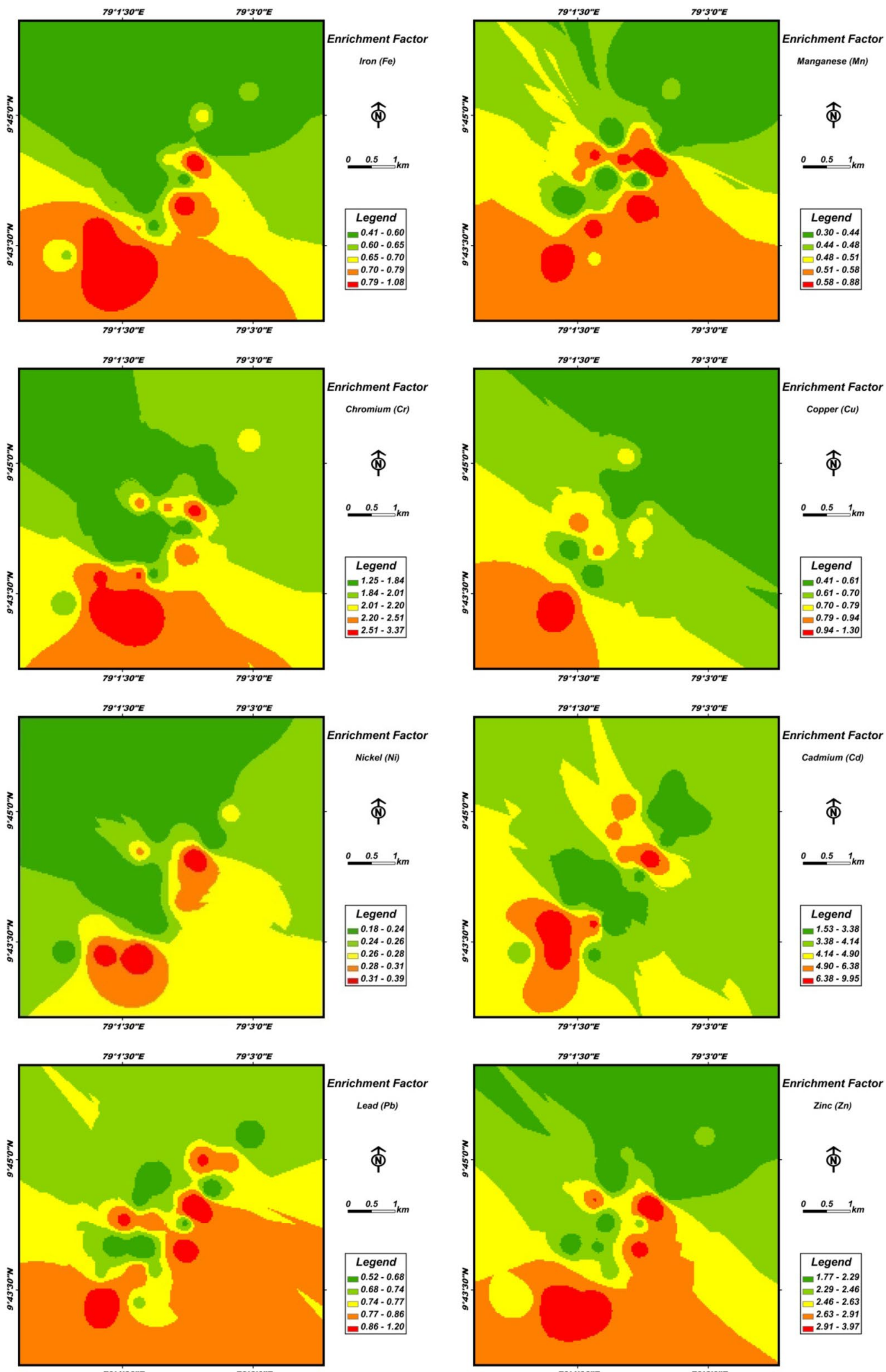

Fig. 3 Distribution maps of enrichment factor for heavy metals in the surface sediments

Table 3 EF, CF, $I_{geo}E_r^i$, and RI (min, max, and ave) values of the heavy metals contaminants in the sediments

Index		Fe	Mn	Cr	Cu	Ni	Cd	Pb	Zn
EF	Min	0.41	0.30	1.25	0.41	0.18	1.52	0.52	1.77
	Max	1.09	0.89	3.38	1.31	0.40	9.97	1.21	3.98
	Ave	0.65	0.50	2.00	0.67	0.25	4.12	0.76	2.48
CF	Min	0.76	0.45	2.31	0.56	0.27	2.75	0.82	3.06
	Max	1.11	0.94	3.78	1.53	0.48	12.25	1.56	4.26
	Ave	0.94	0.73	2.90	0.99	0.37	5.83	1.13	3.61
I_{geo}	Min	9.2	5.43	4.19	3.06	3	− 1.13	1.93	4
	Max	9.37	5.75	4.4	3.49	3.25	− 0.49	2.21	4.14
	Ave	9.29	5.63	4.28	3.28	3.14	− 0.85	2.06	4.07
E_r^i	Min	–	–	1	3	1	82.5	4	3
	Max	–	–	3	8	2.015	367.5	8	4
	Ave	–	–	2.08	5	1.81	174.88	5.58	3.71
RI	–	–	–	48	119	44.3	4197	135	87

Mn, and Cd, "strongly to extremely contaminated" for Cr and Zn, "strongly contaminated" for Ni and Cu, "moderately to strongly contaminated" for Pb in this study area. The heavy metal pollution level tends to be higher in the study area, whereby, the average I_{geo} values of metals were ranked based on the following order: Pb < Ni < Cu < Zn < Cr < Cd < Mn < Fe. These results suggest that anthropogenic sources have considerable effects on Fe, Mn, Cd, Cr, Zn, Cu, Ni, and Pb in the sediments and therefore, require more attention for the monitoring of Fe, Mn, Cd, Zn, Cr, and Cu pollution.

Potential ecological risk factor (E_r^i) and index (RI)

The results of the potential ecological risk factor, E_r^i, and RI for heavy metals in the sediments are shown in Table 3. The average E_r^i values of Cu (5), Zn (4), Cr (2), Ni (1.84), and Pb (6) was less than 40, whereby the ecological risk for each heavy metal is classified as low-potential. In contrast, there was a high ecological risk for Cd (174.88) in the sediments that were mainly due to the discharge of municipal sewage waste in the study area. The RI values of Zn (87), Cu (119), and Pb (135) indicated that there was a considerable ecological risk, followed by Ni (44.35) and Cr (48) which indicated a moderate ecological risk, and lastly, Cd (4197) which indicated a very high ecological risk.

Contamination degree (Cd), modified contamination degree (mCd), pollution load index (PLI), and potential contamination index (CI)

The calculated values of C_d, mC_d, PLI, and C_p values for the heavy metals are shown in Table 4. The C_d values within the range of $12 < C_d < 24$ (minimum-14.62, maximum-23.9, and average-17.429) indicated that there was a substantial degree of contamination at all the stations, in which the sediments in the study area had a considerable degree of contamination. The overall degree of pollution at different sampling sites and C_d data are shown in Table 4. The results indicate that the degree of contamination was nil to very low at nine stations, namely stations 3 (15.35), 6 (14.72), 7 (15.18), 10 (15.1), 11 (14.7), 12 (15.1), 18 (15.3), 20 (14.62), and 21 (14.84), and moderate at stations 1 (19.82), 2 (19.36), 4 (23.9), 5 (21.53), 8 (16.52), 9 (16.14), 13 (16.85), 14 (19.63), 15 (18.51), 16 (19.12), 17 (17.17), 19 (22.27), 22 (17.27), 23 (18.07), and 24 (17.25). The main factors affecting the moderate contamination mC_d values of stations 1, 2, 4, 5, 8, 9, 13, 14, 15, 16, 17, 19, 22, 23, and 24 were based on the CF values of Cr (3.23), Cd (4.18), and Zn (2.65) that were attributed to the anthropogenic pollution at these sites. The average value of mC_d was less than 1, thus indicating that the studied areas have been severely affected by anthropogenic contamination. The PLIs of the heavy metals are shown in Table 4 and the values ranged from 10.30 to 16.74, with an average of 11.97. These results revealed that the sediments in the study area were polluted by heavy metals. [42] reported that the PLI values were less than 1 for the heavy metals in all the sampling stations, thus indicating that the levels of the heavy metals investigated in this study were within the baseline level for all the stations. For instance, the PLI values for all the zones investigated ranged from 0.05 to 2.30. The value of Cd was higher (41) due to the effects of external sources such as industrial activities, agricultural runoff, and other anthropogenic contaminants.

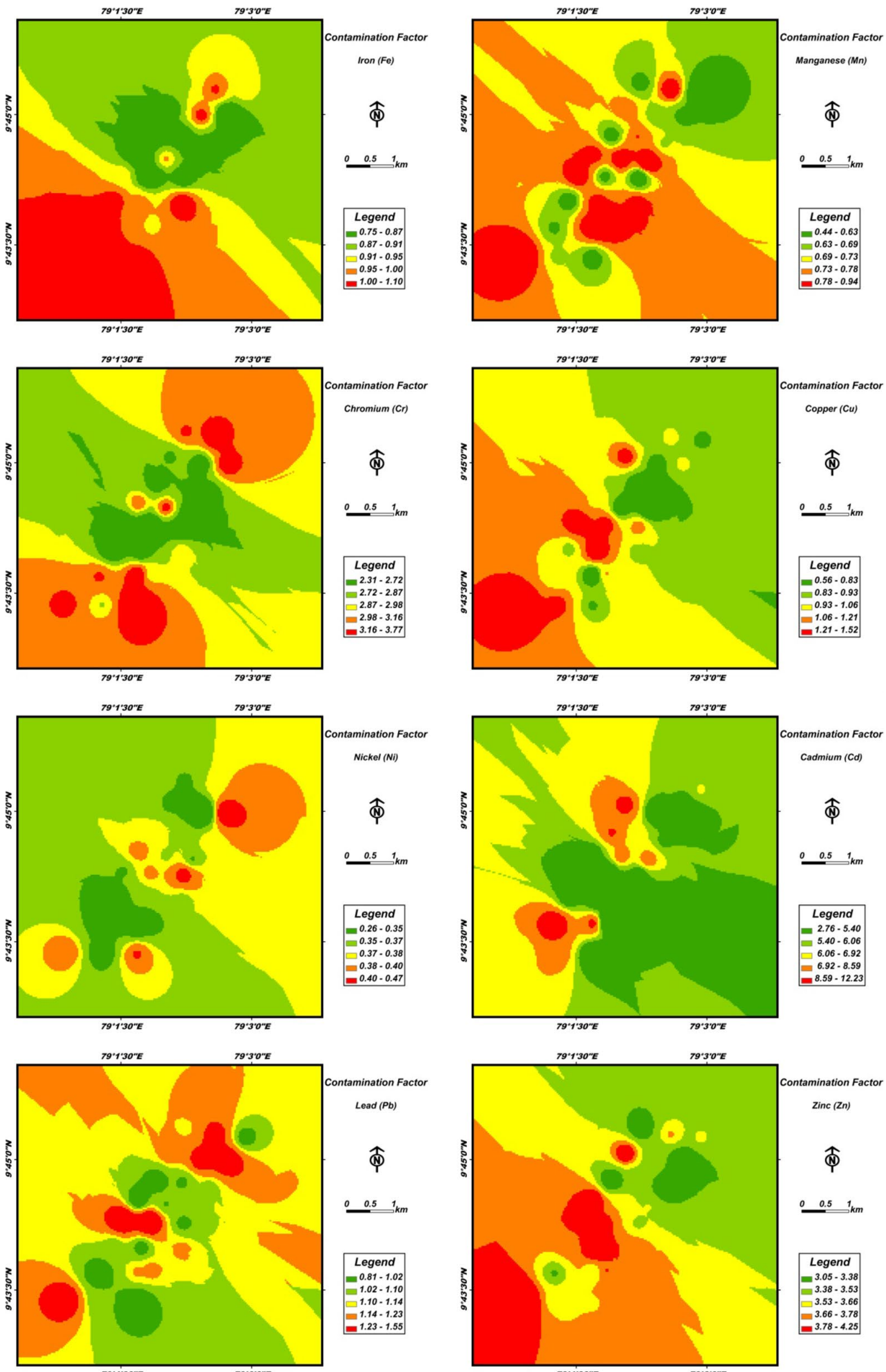

Fig. 4 Distribution maps of contamination factors for heavy metals in the surface sediments

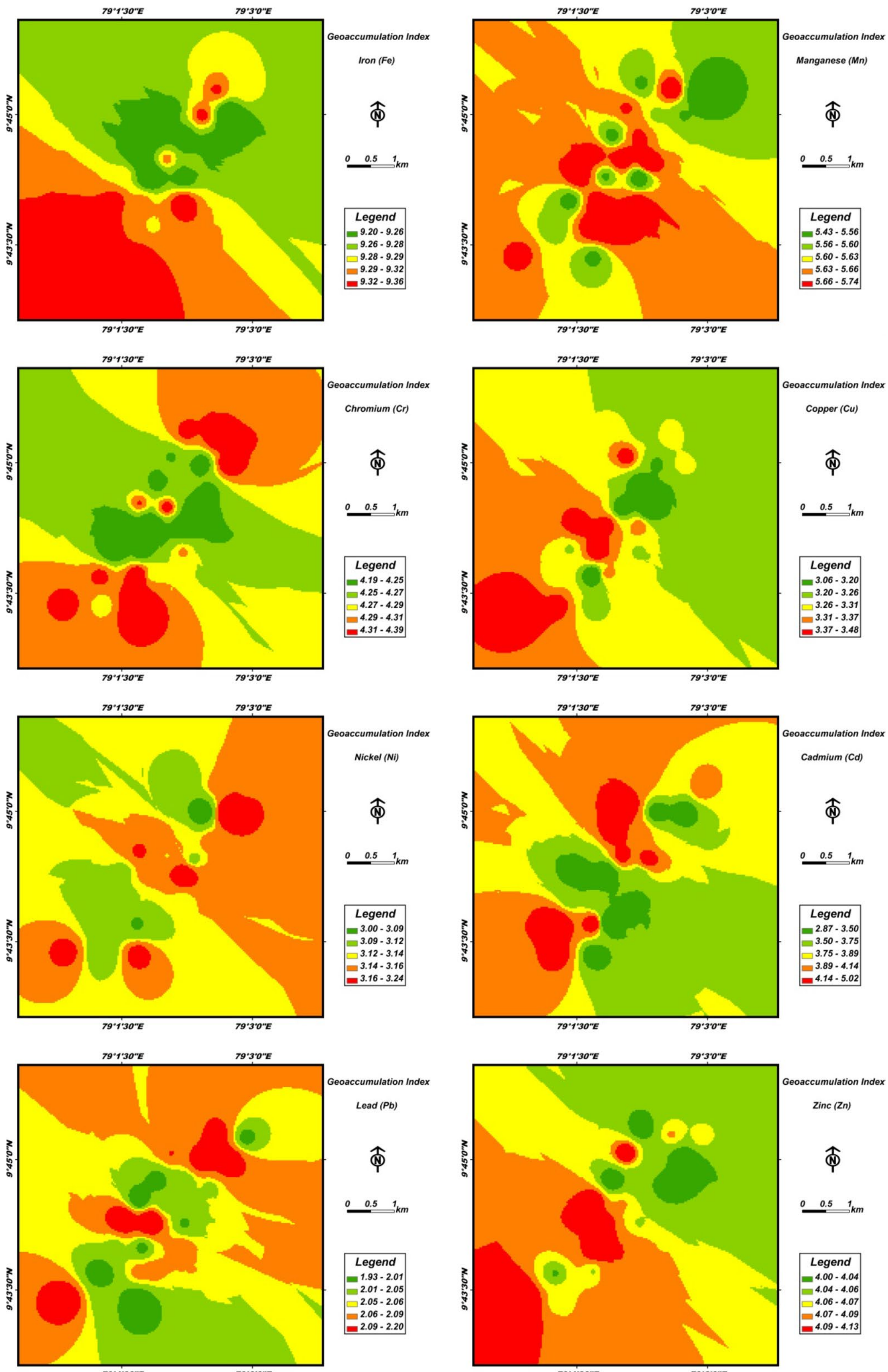

Fig. 5 Distribution maps of I_{geo} for the heavy metals in the surface sediments

Table 4 C_d, mC_d, and PLI values for heavy metals in the surface sediments

S. no.	Fe	Mn	Cr	Cu	Ni	Cd	Pb	Zn	C_d	mC_d	PLI
1	1.11	0.94	3.24	1.53	0.40	6.25	1.38	4.26	19.82	2.477	16.74
2	1.09	0.68	2.86	1.31	0.34	7.90	1.04	3.57	19.36	2.42	13.26
3	1.03	0.58	3.78	0.82	0.41	3.45	0.82	3.66	15.35	1.919	10.80
4	1.09	0.62	3.20	0.95	0.35	12.25	0.90	3.36	23.9	2.987	13.12
5	0.96	0.89	3.50	0.56	0.32	9.45	1.17	3.54	21.53	2.691	12.60
6	0.93	0.91	2.87	1.07	0.35	2.80	1.19	3.79	14.72	1.84	11.40
7	1.04	0.55	2.34	0.87	0.34	4.45	1.06	3.70	15.18	1.898	10.62
8	0.97	0.83	2.47	1.53	0.36	4.45	0.95	4.10	16.52	2.065	12.34
9	1.11	0.83	2.98	0.87	0.36	3.90	1.17	3.76	16.14	2.018	11.87
10	0.93	0.89	2.56	1.46	0.35	2.75	1.49	3.86	15.10	1.888	12.15
11	0.76	0.59	2.31	1.36	0.39	2.80	1.56	3.83	14.70	1.837	11.02
12	0.82	0.5	2.41	1.16	0.43	4.35	0.98	3.50	15.10	1.887	10.74
13	0.82	0.83	3.14	1.06	0.39	4.90	0.88	3.97	16.85	2.106	11.91
14	0.96	0.89	3.28	0.62	0.37	7.80	1.02	3.66	19.63	2.454	12.30
15	0.87	0.94	2.47	0.64	0.35	7.95	1.08	3.546	18.51	2.314	11.73
16	0.85	0.57	2.59	0.91	0.38	8.90	0.93	3.06	19.12	2.39	11.31
17	0.82	0.79	2.71	0.66	0.37	6.30	1.00	3.44	17.17	2.146	11.03
18	0.85	0.69	2.58	0.79	0.38	5.10	1.06	3.20	15.30	1.913	10.77
19	0.84	0.78	2.69	1.36	0.35	9.90	1.22	4.00	22.27	2.784	14.40
20	1.04	0.66	2.49	0.76	0.27	3.25	1.35	3.44	14.62	1.827	10.30
21	0.76	0.60	3.41	0.96	0.47	3.15	1.40	3.06	14.84	1.855	11.10
22	0.91	0.59	3.20	0.86	0.35	6.15	1.14	3.17	17.27	2.159	11.40
23	1.02	0.89	3.47	0.96	0.38	5.55	1.30	3.69	18.07	2.258	13.51
24	0.92	0.45	3.12	0.82	0.39	6.15	0.96	3.56	17.25	2.156	10.93

Table 5 Correlation coefficient (*R*) values ($p < 0.05$) between different metals and sand (%), mud (%), and OM in the marine sediments

Correlations ($p < 0.05$)										
.001										Mud
.648	.645									OM
.226	.239	.977								Fe
.963	.967	.344	.310							Mn
.673	.678	.969	.182	.428						Cr
.748	.739	.420	.911	.499	.180					Cu
.507	.508	.832	.024	.276	.167	.414				Ni
.693	.688	.669	.455	.864	.392	.321	.263			Cd
.996	.995	.577	.596	.197	.476	.085	.844	.063		Pb
.962	.964	.774	.181	.006	.777	.002	.653	.412	.381	Zn
Sand	Mud	OM	Fe	Mn	Cr	Cu	Ni	Cd	Pb	

Pearson's correlation matrix

Pearson's correlation analysis defines the relationship between the heavy metals and their major contributors in the environment [49–51]. Pearson's correlation coefficients for sand, mud, OM, Fe, Mn, Cr, Cu, Ni, Cd, Pb, and Zn components are shown in Table 5. Sediment sand, mud, and OM displayed a strong positive correlation with all the elements investigated. For instance, Fe was strongly correlated with Cu ($r^2 = 0.911$), Cd ($r^2 = 0.455$), and Pb ($r^2 = 0.596$) and weakly correlated with Cr, Ni, Mn, and Zn. On the other hand, Mn was strongly correlated with Cr ($r^2 = 0.428$), Cu ($r^2 = 0.499$), and Cd ($r^2 = 0.864$) and weakly correlated with Pb, Ni and Zn. Additionally, Cr was strongly correlated with Pb ($r^2 = 0.476$), and Zn ($r^2 = 0.777$) and weakly correlated with Cu ($r^2 = 0.180$), Cd ($r^2 = 0.392$), and Ni ($r^2 = 0.167$), while Ni was strongly correlated with Pb ($r^2 = 0.844$), and Zn ($r^2 = 0.653$) and weakly correlated with Cd ($r^2 = 0.263$).

Table 6 The result of PCA/FA for entire data set

	PC1	**PC2**	**PC3**
Component matrix[a]			
Eigen value	2.463	2.023	1.270
% of Variance	2.034	22.596	49.965
Cumulative %	1.321	14.682	64.646
Loading of variances			
Fe	−0.178	0.771[b]	0.017
Mn	0.329	0.680[b]	0.208
Cr	−0.371	0.211	0.811[b]
Cu	0.270	0.126	0.676[b]
Ni	0.123	−0.590	0.721[b]
Cd	0.568[b]	0.293	−0.085
Pb	0.206	0.612[b]	−0.066
Zn	0.603[b]	0.203	0.174

[a] Extraction method: principal component analysis

[b] Loading values of being equal to or greater than 0.5

Principal component analysis/factor analysis, Q-mode cluster

Multivariate analysis is commonly used to distinguish factors such the natural and anthropogenic contributions of the elements according to the various levels of relationship [17, 18, 50–52]. In this study, PCA/FA was employed to ascertain possible relationships of the variables and their input sources among the pollutants. The Kaiser–Meyer–Olkin (KMO) and Bartlett's values obtained in the study were 0.624 and 72 (df = 24, $p < 0.001$), respectively, thereby indicating that PCA/FA could be used for the reduction of dimensions. FA performed on the PCs and three VFs shows eigenvalue > 1 that explains the quality of the sediments [53, 54].

The PCA/FA plots for various parameters investigated in this study were obtained using the rotated matrix analysis and varimax normalisation was used to calculate the variables. Therefore, the 8 variables from the surface sediments of the study area were summarised by three PCA/FA, with the cumulative percentage of 1.321%, 14.682%, and 64.646%, respectively. These three components accounted for 2.034%, 22.596%, and 49.965% of the variances as listed in Table 6. The resulting dendrogram of Q-mode hierarchical cluster analysis represents the grouping of samples according to the heavy metal. The dendrogram shows two different groups (i.e.) Cluster 1 (9, 10, 11, 12, 13, 14, 15, 16, 17, 18, 19, 20, 21, 22, 23 and 24), Cluster 2 (1, 2, 3, 4, 5, 6, 7, and 8) (Fig. 6).

Discussion

The spatial distribution of heavy metals in marine sediments is of great significance in elucidating the pollution summary of aquatic environments [55–57]. Besides that, those distributions of marine sediments is influenced by

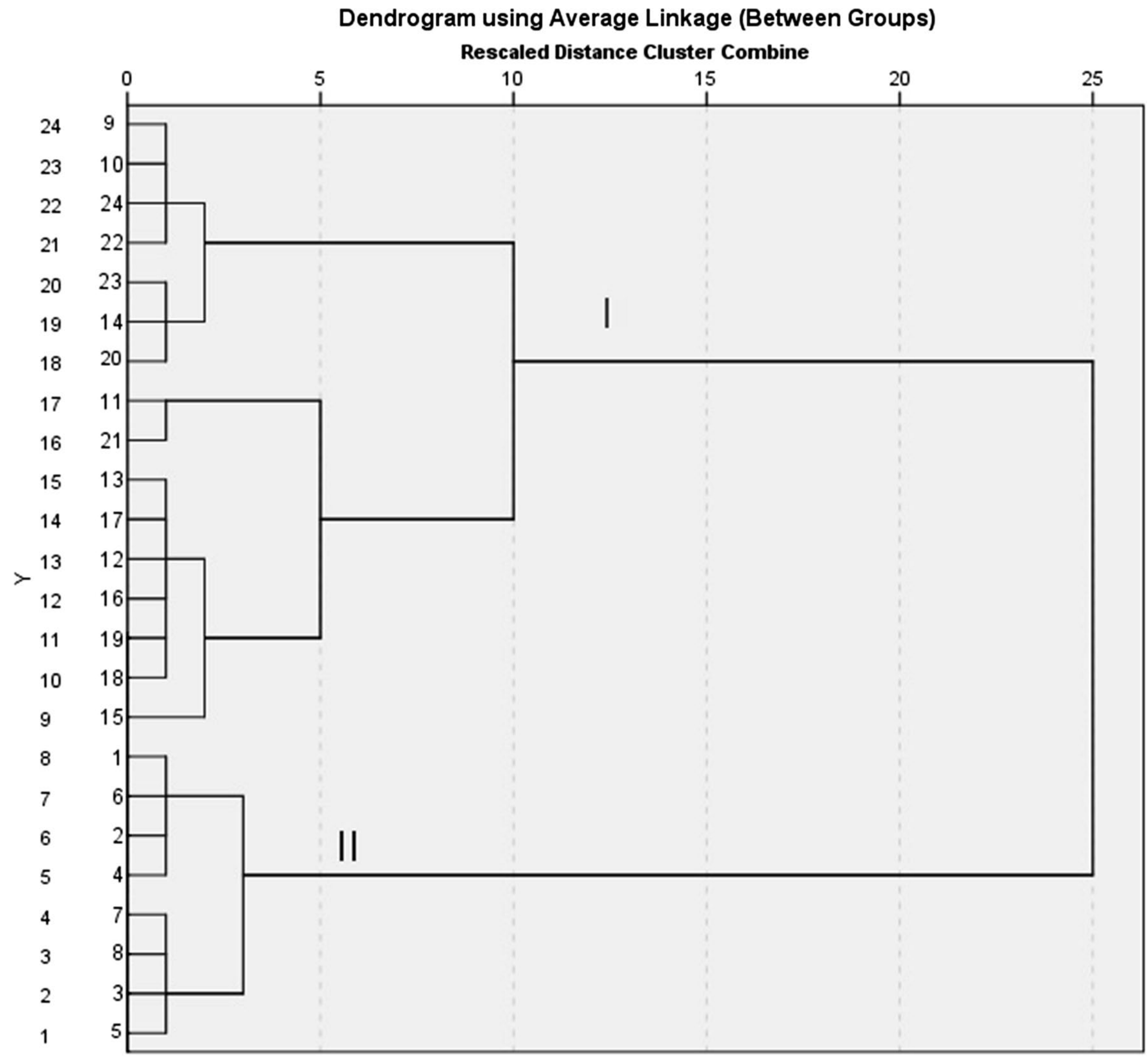

Fig. 6 Dendrogram representing sampling sites with surface sediments of heavy metals in the study area

natural and anthropogenic factors such as weathering of parent rock, industrial wastewater, transportation, agriculture and environment (Morillo et al., 2004; El Nemr et al., 2006; Luo et al., 2006) The link between natural and anthropogenic influences on heavy metals in marine sediments is critical. Sediment analysis is crucial in determining the pollution status of the marine environment. The spatial distribution of the total metal concentration of Fe > Mn > Cr > Zn > Cu > Ni > Pb > Al > Cd is illustrated in Fig. 7. In general, the concentration of heavy metals in the sediments decreased in the following sequence: Fe > Mn > Cr > Zn > Cu > Ni > Pb > Al > Cd. The concentration of metals in the sediment were as follows: 42,516–62,413 mg kg^{-1} (Fe), 425–896 mg kg^{-1} (Mn), 231–378 mg kg^{-1} (Cr), 214–298 mg kg^{-1} (Zn), 31–84 mg kg^{-1} (Cu), 20.1–35.6 mg kg^{-1} (Ni), 10.2–19.5 mg kg^{-1} (Pb), 7.32–15.4 mg kg^{-1} (Al), and 0.6–2.5 mg kg^{-1} (Cd).

The classification suggests that the muddy sand sediments observed in many stations were due to the poor wave action and shallow regions in the area [58]. The high value of Fe (62,413 mg kg^{-1}) was observed due to the convergence of ephemeral streams and the presence of rich mangrove ecosystems along the southern part of the study area at station 1 [59]. The red inorganic pigment used for painting boats is based on iron (III) oxide (Fe_2O_3). Iron oxide leads to the co-precipitation of heavy metals which subsequently, increases the metal concentration in the sediment [60]. It is worth noting that the concentration of Fe in the east coast of India was higher than that of the study area due to chemical weathering. However, in this study, the maximum

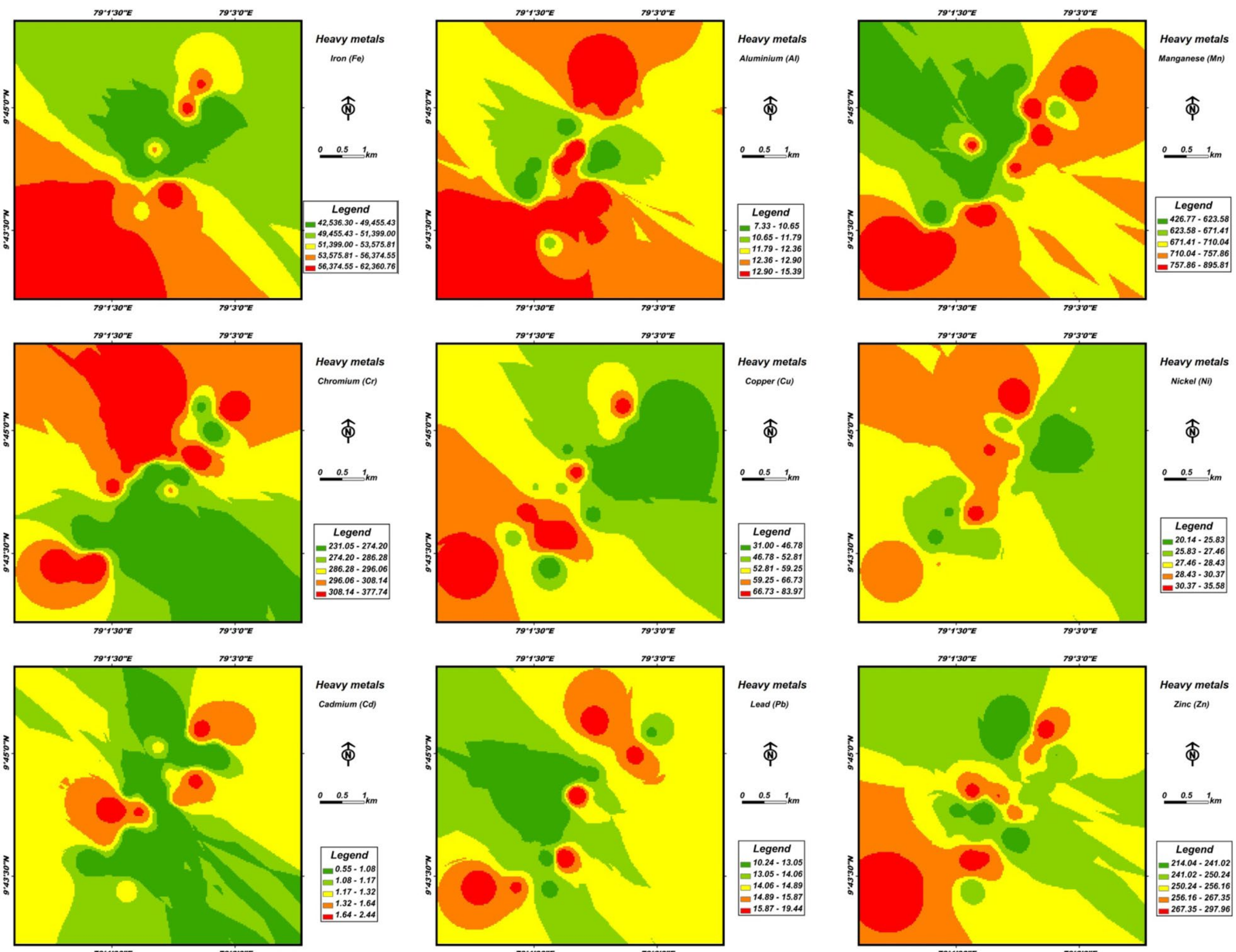

Fig. 7 Distribution maps of heavy metals concentrations in the surface sediments of the Thondi coast

concentration of Fe was greater than the average crustal values [61]. The highest value of Mn (896 mg kg^{-1}) was detected at station 1 due to the presence of mangrove vegetation along the coast. Overall, the concentrations of Fe and Mn indicate that both these elements are predominantly regulated by the riverine input and existence of the mangrove vegetation [20]. The main source of Mn is anthropogenic inputs such as industrial effluents and emissions [62]. The residual, Fe/Mn oxide fractions are the dominant geochemical phases for heavy metals in these metal-enriched sediments (Fig. 8). For instance, the concentration of Mn in the Pitchavaram Mangrove region, east coast of India, was higher than that of the study area [63]. The concentration of Ni ranged from 20.1 to 35.6 mg kg^{-1} in the study area and it was mainly derived from the wind-blown dust, vegetation, and weathering of rocks and soils [17]. The concentration of Ni in the southeast coast of India, however, was higher than that of the study area and the average crustal values [61]. The concentration of Cr ranged from 231 to 378 mg kg^{-1} and the average concentration of Cr in the sediment was 290.3 mg kg^{-1}. This value was higher than the average crustal value, thus indicating the input of Cr to the study area. Both natural and anthropogenic sources were responsible for the accumulation of Cr in the sediment. The values of Cd ranged from 0.6 to 2.5 mg kg^{-1} and the mean Cd concentration was higher (1.2 mg kg^{-1}) than the average crustal value, thus indicating that the input of Cd was likely from both natural and anthropogenic sources, especially from the municipal sewage wastes nearby the study area. Nevertheless, it was previously reported that the concentration of Cd in the sediments was the main indicator of anthropogenic activity [61, 64]. Similarly due to anthropogenic inputs the concentration of Cd is 2.9 ppm recorded in marine sediments

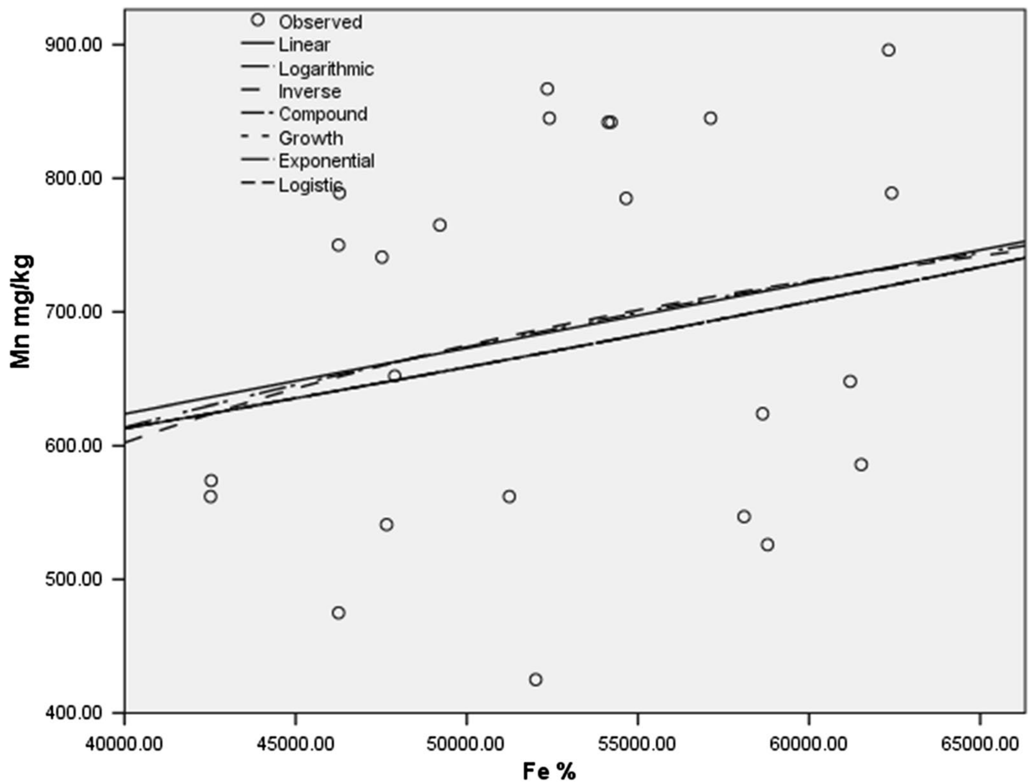

Fig. 8 Scatter plots of the linear regression models ($N = 24$): Fe/Mn ($r^2 = 0.043$)

in the Gulf of Mannar and Palk Bay, southeast coast of India [65]. Municipal wastewater, for instance, has a higher concentration of Cd, particularly from the fishing harbour activities, domestic sewage, oil, fish, and industrial and aquaculture wastes in the Ramnad District in Tamil Nadu. The concentration of Zn was observed to be higher at station 1 (298 mg kg^{-1}) and lower at station 17 (214 mg kg^{-1}). The anticorrosive paint used on the boat mainly consists of $ZnSO_4$ [66, 67]. Ocean currents and transportation activities erode this paint and increase the concentration of Zn in the shelf sediments. Additionally, emissions and effluents from the industries represent the main sources of zinc [62].

Cu was present in all the sediments and the values ranged from 31 to 84 mg kg^{-1}, with an average concentration of 54.7 mg kg^{-1}. The confluence point of the river was found to be rich in Cu, thus indicating that the presence of trace elements in the marine environment was mainly due to the riverine runoff and drainage of untreated industrial wastes into the river. This inference was obtained from the low concentration of heavy metals and crustal average values detected in the nearby sampling areas. For instance, e-wastes such as waste from electroplating and printed circuit boards increased the levels of Zn, Pb, Cu, Cd, and Hg in the sediments collected from Hong Kong coastal areas [68]. Likewise, on the coast of Bangladesh, heavy metal contamination was mainly due to industrial pollutants [69]. The untreated effluents and solid wastes from many commercial and small-scale industries such fertilisers, sugar, paint, tobacco, jute, plastic, refinery, textiles, paper, and shipwreaking set up along the coastline and riverbank areas contributed to metal pollution [70–72]. Heavy metals such as Ni, Pb, Zn, Cu, and Cr, derived from both natural and anthropogenic sources, are thought to be related to sedimentary phases such as organic matter and carbonate [73–75]. The heavy metal concentration in this study was compared with other coastal regions in India and other countries (Table 7). The heavy metal concentrations of Cu, Cr, Zn, Cd, and Pb in this study surpassed the crustal average and mean values of heavy metals in other coastal areas.

According to the classification [46], the majority of heavy metals showed minor to extremely severe enrichment in the sediments or displayed no enrichment to moderate enrichment. Based on the EF values, 65.63% of the samples belongs to no enrichment, 29.68% of the samples fall under minor enrichment followed by 6.25% of the samples categorised under moderately and 3.64%

Table 7 Comparison of heavy metal concentration and crustal average values obtained in this study with other coastal regions in India and around the world

Study area	Fe	Mn	Cu	Pb	Cd	Ni	Cr	Zn
Present study	52,802	686.1	290.3	54.7	1.2	27.7	14.1	252.9
Average crustal [39]	56,300	950	55	12.5	0.2	75	100	70
Coromandel Coast, Bay of Bengal [17]	7,144	–	76.45	49.629	19.8	27.984	109.45	78.76
Southeast coast of India [92]	2,780	543	6.7	19.7	–	50.7	191.3	58.7
Van Island, Gulf of Mannar, India [93]	31,219	163	57.81	348.4	–	110.04	108.73	52.5
Koswari Island, Gulf of Mannar [18]	30,988	147	54.2	496.7	–	92.7	67.5	16.9
Off Chennai, India [52]	15,032	–	113.2	325	–	51.2	200.1	123.9
Cuddalore, SE coast of India [21]	10,982	291	40	33.9	–	39.2	127	37.7
Palk Strait, southeast coast of India [94]	55,680	661	69	19	0.4	27	302	244
Parangipettai coast, India [95]	11,804	45	30.6	30.12	–	25.2	77.8	44.7
Arabian Gulf coast [96]	4,100	120	18	12	–	6	130	46
Tuticorin Coast, India [97]	28,717	330	52	42	–	30	15	247
South Port Klang, Malaysia [98]	-	219.1	24.89	96.02	1.46	13.9	60.19	72.2
East Coast of India [23]	85,800	–	140.14	–	–	81.76	133.22	–
Tuticorin Coast [24]	28,717	–	52	42	0.2	75	15	247
Bay of Bengal, off Ennore [60]	27,200	–	506.2	32.36	6.58	38.61	194.8	126.8
Southwest coast of Spain [99]	35,300	569	336	197	2.5	50	92	649
Pitchavaram Mangrove region [63]	32,482	941	43.4	11.2	6.6	62	141.2	93

of the samples categorised under moderately severe in the study area. In the stations of 2, 4, 5, 14, 15, 16, and 19 EF values for Cd showed moderately severe enrichment, thereby indicating that Cd was mainly derived from anthropogenic inputs. The higher concentration of Cd was mainly due to sewage discharge, mining agriculture, and industrial activities [17, 76–79]. Moreover, Cd is highly toxic to animals and plants and it has no proven essential biological function [80]. The overall enrichment of the elements in the marine sediments shows Ni > Mn > Fe > Cu > Pb > Cr > Zn > Cd. EF shows that the metal pollution occurred both from the natural and anthropogenic sources [81]. Both natural and anthropogenic sources are possible for the Cd contamination of samples around Thondi coast, Palk Bay, South India. Since all of the sites reported EF values greater than 5, the analysis concluded that anthropogenic inputs have resulted in substantial metal enrichment.

The ranking of RI for the heavy metals in the sediments was Ni < Cr < Zn < Cu < Pb < Cd. As previously mentioned, the major potential ecological risk of heavy metals in the surface sediment was from Cd and this observation was mainly due to the effects of anthropogenic activities such as the use of phosphate fertilisers and swine manure in irrigation [82].

Based on the values of I_{geo}, the sediments of Thondi coast are "extremely contaminated" with Fe, Mn, and Cd, "strongly to extremely contaminated" for Cr and Zn, "strongly contaminated" for Ni and Cu, "moderately to strongly contaminated" for Pb possibly due to the anthropogenic inputs such as discharge of sewage, oil pollution from ships, use of paints for fishing and tourist boats. The high contents of Fe, Mn, and Cd, in the study area result from various anthropogenic activities including dredge, land filling, localised oil pollution, using antifouling and anticorrosive paints from fishing and tourist boats, and sewage discharging from various sources within the study area. In similar that the recent bottom sediments of Mabahiss Bay, North Hurghada, Red Sea, Egypt [83].

The report by [84] indicated that the heavy metal, Cd, originates from agricultural soil contamination, municipal sewage waste, mining effluent, and sludges as well as from the erosion of phosporites, sulfide ores, hydrothermal mineralised rocks, and black shale deposits. In contrast, the values for Fe, Cr, Mn, Cu, Pb, Ni, and Zn were lower, thus indicating that there was no pollution based on the comparison with the worldwide sediment values. The difference in the indices, however, was due to the difference in the sensitivity of these indices in determining the pollutants in sediments [85]. The PLI values obtained for the sediments revealed that the sediments have been polluted by heavy metals. The values of both mC_d and PLI suggest the effect of anthropogenic sources on the levels of heavy metal pollution in the sediments [40, 41, 86, 87]. The potential contamination index (C_p) values of Fe, Mn, Ni, and Pb suggest a low level of contamination, while Cu and Zn indicated moderate contamination. In contrast, Cr and Cd showed a severe contamination level due to various sources such as domestic sewage, oil and fish, industrial, aquaculture waste, fishing harbour activities, and aquaculture waste in the study area.

The metals Cd, Cr, Ni, Cu, and Pb were significantly correlated as they were related to anthropogenic sewage and wastewater. The level of Fe indicated that the trace metal elements were acquired from their source [88], in which the same hydrogeochemical process redistributes these trace elements into the sediments [89]. Lastly, the significant and positive correlation observed for Mn with Cr, Ni, and Cu components substantiate their presence in the sediment [82–90]. It should be noted that Cr, Ni, Cu, and Pb are extensively recognised to have anthropogenic activities, whereby Cu, Pb, and Ni are commonly derived from anthropogenic sewage and wastewater, while Cr is generally associated with industrial activities in the area [91]. The first cluster of the sampling sites 9, 10, 11, 12, 13, 14, 15, 16, 17, 18, 19, 20, 21, 22, 23 and 24 with significant contamination the background sites. Cluster 2 includes three sub-clusters of the sampling sites 1, 2, 3, 4, 5, 6, 7, and 8 with moderate enrichment of heavy metals.

Principal component analysis/factor analysis

The PCA/FA, which corresponded to three eigenvalues were greater than 1. The three PCA/FA accounted for 74.60% of the total variance in the data and displayed the different factors (sources) influencing sediment pollution in the study region. Table 6 shows that PC1 accounted for 2.034% of the total variance, and Cd and Zn contributed to the strong positive loadings. The results for EF (moderately severe enrichment), CF (considerable contamination), C_p (severe contamination level), I_{geo} (extreme contamination) and RI (very high ecological risk) suggest that the presence of Cd (0.568) was due to high anthropogenic inputs (81). The CF (considerable contamination for Zn), C_p (moderate contamination for Zn), and RI (considerable ecological risk for Zn) values suggest that Zn (0.603) is influenced by anthropogenic sources. Therefore, it is assumed that PC1 primarily represents the contribution of pollutants by anthropogenic sources. The high positive loadings of Cd (0.568), and Zn (0.603) in the PC1 indicate that these metals may have a similar distribution and common anthropogenic sources in the study area. Similarly, from the PCA results the presence of Cd, and Zn pollutants signifies the anthropogenic contamination in the Indian Sunderban [100]. In PC2 (22.596% of the total variance), Fe, Mn, and Pb contributed to the strong positive loadings. The results for EF (no enrichment for Pb, low level of contamination for Fe, and Mn), I_{geo} (extremely contaminated for Fe, Mn and moderately to strongly contaminated for Pb), and RI (considerable ecological risk for Pb) suggest that levels of Fe, Mn, and Pb are influenced by low levels of pollutants. Therefore, it can be assumed that PC2 primarily represents contaminants of low anthropogenic sources. For PC3 (49.965% of the total variance), a strong positive loading was observed for Cr, Cu, and Ni. The results for EF (minor enrichment for Cr, no enrichment for Cu and Ni), CF (moderately contaminated for Cr, low level of contamination for Cu and Ni), I_{geo} (strongly to extremely contaminated" for Cr, strongly contaminated" for Ni and Cu), and RI (considerable ecological risk for Cu, moderate ecological risk for Ni and Cr) suggest that levels of Cr, Cu, and Ni are influenced by low levels of pollutants. Therefore, it can be assumed that PC3 primarily represents contaminants of low anthropogenic sources. From the PCA results, it can be concluded that factors 1 and 3 are from anthropogenic sources such as domestic sewage, waste water discharge, construction activities and urban runoff.

Conclusion

The evaluation of heavy metals in the surface sediments along the Thondi coast in Palk Bay was undertaken, whereby sand, mud, organic matter, and heavy metals such as Fe, Al, Mn, Cr, Cu, Ni, Cd, Pb, and Zn were analysed. The mean concentration of heavy metals was shown to decrease in the following order: Fe > Mn > Cr > Zn > Cu > Ni > Pb > As > Cd. The sedimentary texture observed in most of the stations was muddy sand due to the shallow depth and poor wave action in the study area. The pollution indices such as EF, CF, mC_d, CI, E_r^i, I_{geo}, and RI indicated that Cd was responsible for the high contamination level in the study area except for I_{geo}. The PCA results also confirmed that Cd had a high contamination level, as indicated by the strong positive loadings. The main source of Cd was due to anthropogenic inputs such as municipal wastewater, domestic sewage discharge, fishing harbour activities, and industrial and aquaculture wastes. Based on the results obtained for EF, CF, m$C_{d,}$ CI, E_r^i, and RI, it was found that the presence of heavy metals such as Cu, Zn, Pb, and Cr led to moderate contamination in the study area. The PLI results suggest that the sediments in the study area were polluted by heavy metals. The findings of this study revealed that the study area frequently receives heavy metal contaminants from different sources and if the concentration of these heavy metals continues to increase, the toxicity will also increase, thus affecting the entire food chain within the marine ecosystem. Therefore, to protect the marine ecosystem, illegal discharges into the marine environment should be properly monitored and effluents from the industries, municipal, and domestic areas should be pretreated before its discharge into the coastal areas.

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Authors' contributions
Karthikeyan Perumal: conceptualisation, supervision, investigation, methodology, writing—original draft, writing—review and editing. Joseph Antony: data curation, formal analysis; Subagunasekar M: software; data analysis; investigation.

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Declarations

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