Chapter 22 Assessment of Metal Pollution in Ulhas Estuary, Mumbai, India

Lina L. Fernandes and G.N. Nayak

Abstract In order to understand historical trend of pollution level by heavy metals in Mumbai – a commercial and industrial centre, a sediment core from the intertidal region of Ulhas estuary was analyzed for various chemical parameters such as organic matter, sediment components and selected metals. Analysis of these data through Pearson's correlation coefficient displayed inter-elemental associations. Factor and cluster analyses showed that finer sediment components and organic matter along with Fe-Mn oxides played a significant role in accumulation and distribution of metals. Various pollution indices such as Enrichment Factor (EF), Pollution Load Index (PLI) and Geo-accumulation index (Igeo) were computed which showed that the concentrations of Fe and Ni in the sediments were slightly higher than or equal to their background values. On the other hand, the concentrations of Mn, Cu, Pb, Co, Cr and Zn in the sediments were found to be significantly higher than their background values. PLI showed increasing trend of metal pollution over the years. Based on Igeo values, all the elements were grouped in the unpolluted class except for Cu and Pb which were found to be moderately polluted, especially in the upper portion of the core. Elevated levels of Cu and Pb may be attributed to the increasing use of antifouling paints and vehicle exhausts, respectively.

22.1 Introduction

With the onset of industrialization and urbanization, the input of heavy metals into estuarine or coastal environments through river runoff and land based point sources have increased drastically and, as a result, such coastal water bodies often act as reservoirs of river-borne and marine-derived pollutants (Duinker [1989\)](#page-13-0). Tidal flats of estuarine regions are considered as important heavy metal sinks

L.L. Fernandes \cdot G.N. Nayak (\boxtimes)

Department of Marine Sciences, Goa University, Goa 403206, India e-mail: gnnayak@unigoa.ac.in

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(Birch [2000\)](#page-13-0) and as such, the occurrence of elevated levels of trace metals especially in the sediments can be good indicators of man-induced pollution. Sediments of these regions not only provide an indication of the environmental quality of the aquatic system but also provide important information in identifying the contamination sources, extent, and history and indicate the degree of pollution (von Gunten et al. [1997;](#page-14-0) Anu et al. [2009\)](#page-13-0). Therefore, study of trace metal distribution in marine sediments is commonly undertaken in order to understand geochemical and environmental processes with a view to assess the environmental quality and pollution levels. High levels of heavy metals can often be attributed to anthropogenic influences, rather than natural enrichment of the sediment by geological weathering (Davies et al. [1991;](#page-13-0) Morton and Blackmore [2001](#page-13-0)). In addition, the accumulation of heavy metals in sediments can be a secondary source of water pollution, once environmental conditions change (Chen et al. [1996](#page-13-0); Cheung et al. [2003\)](#page-13-0). Hence, the assessment of heavy metal contamination in the sediments is an indispensable tool to assess the risk of an aquatic environment.

Numerous studies have been conducted on trace metal distribution in estuarine sediments throughout the World. The objectives of the present work are to illustrate the distribution and levels of sediment contamination by heavy metals [Iron (Fe), manganese (Mn), copper (Cu), lead (Pb), cobalt (Co), nickel (Ni), zinc (Zn) and chromium (Cr)] in intertidal region of Ulhas estuary to assess the pollution level of the region. These metals were chosen because of their abundance and toxic effects in the environment of highly industrialized and urbanized areas.

22.2 Study Area

The study area lies between latitudes 18° 45′ N to 19° 00′ N and longitudes 72° 45′ E to 73° 20' E (Fig. [22.1](#page-2-0)). The Ulhas River originates from Sahyadri Hills, near Khandala, and passes through Ulhasnagar and Kalyan areas and finally meets the Arabian Sea at Vasai. The estuary is lined with vast mudflats and extensive mangrove areas. An outlet of the river touches the head of the Thane creek, south of Mumbra. Dredging activities near Mumbra re-suspend the finer sediments and disturb the normal distribution of particulate and dissolved materials along its course. The region receives an annual mean rainfall of 2,600–2,800 mm which is essentially concentrated during the monsoon season (June to September). The estuary is strongly influenced by tides, with spring tidal range varying from about 4.5 m at the mouth to 2.8 m at the upstream end. The River has barrages constructed in the upstream region which permit only limited river discharge into the estuary during the early dry season. The estuarine flushing time has been estimated to vary between 73 and 211 tidal cycles during the dry season. The estuary receives effluents from multiple industries situated in the Kalyan-Ulhasnagar belt and domestic sewage from the surrounding localities.

Fig. 22.1 Map of the study area showing the sampling location

22.3 Materials and Methods

A sediment core (70 cm length) was collected at low tide, from the intertidal region of the Ulhas estuary, by driving a handheld PVC tube into the sediment. In the laboratory, the core was sub-sampled at 2 cm intervals and stored at 4° C till further analyses. For geochemical analysis, the sediment samples were dried in the oven at 60° C. Analysis of the sediment components was carried out using standard sieve and pipette technique after destruction of organic matter with H_2O_2 (Folk [1974\)](#page-13-0). For chemical analysis, the dried samples were ground into fine powder using mortar and pestle. Total organic carbon (TOC) was determined employing a rapid titration method (in chromic acid environment) adopted from Gaudette et al. ([1974\)](#page-13-0), while Total Phosphorus (TP) and Total Nitrogen (TN) were analyzed using Grasshoff [\(1999](#page-13-0)) method wherein the sediment samples were autoclaved with persulphate, boric acid and sodium hydroxide mixture and then analyzed for TN, as a azo dye complex, while TP was measured using ammonium molybdate method with ascorbic acid reduction. For the metal analysis, the sediment samples were digested in an open digestion system with Hydrofluoric acid: Nitric acid: Per-chloric acid mixture (Jarvis and Jarvis [1985](#page-13-0)). Flame Atomic Absorption Spectrophotometer (Varian AA 240 FS) was used for determination of nine selected metals (Fe, Mn, Cu, Pb, Co, Ni, Zn, Cr and Al) with an air/acetylene flame for all of the above elements except for Al, for which nitrous oxide/acetylene flame was employed at specified wavelengths. Each sediment sample for metal determination was analyzed in triplicate and the relative standard deviation was $< 5 \%$. In addition to blanks and replicates, a certified reference material (BCSS- Marine Sediment Reference Material for Trace Metals and Other constituents) from National Research Council Canada was included in the analytical procedure, to evaluate the effectiveness of the digestion procedure. The percent recoveries of Fe, Mn, Cu, Co, Ni, Cr, Pb, Al and Zn were 84.8 %, 94.2 %, 100.9 %, 74.1 %, 83.9 %, 75.6 %, 92.8 %, 77.4 % and 85.5 %, respectively.

22.3.1 Statistical Analysis

Pearson's correlation coefficient, cluster and factor analyses were employed, using Statistica 6.0 software, to understand the relationships between the different variables. Cluster analysis was used to group objects of similar kind while factor analysis made the components more interpretable.

22.3.2 Pollution Indices

Various methods for quantifying the degree of metal enrichment in sediments have been employed. The degree of contamination in the sediments was determined with the help of three parameters – Enrichment Factor (EF), Pollution Load Index (PLI) and Geo-accumulation Index (Igeo). EF can be used to differentiate between the metals originating from anthropogenic and natural processes. The EF method normalizes the measured heavy metal content with respect to a sample reference such as Fe or Al (Mendiola et al. [2008\)](#page-13-0). The values of EFs are obtained using the following equation,

$$
EF = \left[(\text{metal}/\text{Al}) \right]_{\text{sediment}} / \left[(\text{metal}/\text{Al}) \right]_{\text{shale}} \tag{22.1}
$$

In this study, normalizing relative to Al has been used to compensate for variations in both grain size and composition, since it represents the quantity of aluminosilicates, which is the predominant carrier phase for adsorbed elements in the coastal sediments (Feng et al. [2004\)](#page-13-0). The background value taken was that of average shale (Turekian and Wedepohl [1961](#page-14-0)). Five contamination categories recognized on the basis of the enrichment factor (Sutherland [2000](#page-14-0)) are: $EF < 2$ deficiency to minimal enrichment, $EF \leq 2-5$ moderate enrichment, $EF \leq 5-20$ significant enrichment, $EF \le 20-40$ very high enrichment, $EF > 40$ extremely high enrichment. As the EF values increase, the contributions of the anthropogenic origins are also found to increase (Sutherland [2000](#page-14-0)).

Pollution intensity	Sediment accumulation	Igeo class	
Very strongly polluted	>5	6	
Strongly to very strongly polluted	$4 - 5$		
Strongly polluted	$3 - 4$		
Moderately to strongly polluted	$2 - 3$	3	
Moderately polluted	$1 - 2$		
Unpolluted to moderately polluted	$0 - 1$		
Practically unpolluted	< 0		

Table 22.1 Geoaccumulation index proposed by Muller ([1979\)](#page-13-0)

22.3.2.1 Contamination Factor

The level of sediment contamination by a metal is often expressed in terms of a contamination factor calculated as

Contamination Factor (CF) = Metal content in the sediment/ Background value of metal. (22.2) of metal. (22.2)

According to Hakanson ([1980\)](#page-13-0) classification, $CF < 1$ refers to low contamination, $1 \leq C$ F \leq 3 means moderate contamination, $3 \leq C$ F \leq 6 indicates considerable contamination and $CF > 6$ indicates very high contamination.

22.3.2.2 Pollution Load Index (PLI)

PLI has been evaluated following the method proposed by Tomilson et al. ([1980\)](#page-14-0). This parameter is expressed as:

$$
PLI = (CF1 \times CF2 \times CF3 \times \dots \times CFn)^{1/n}
$$
 (22.3)

where "n" is the number of metals (eight in the present study) and CF is the contamination factor.

22.3.2.3 Index of Geoaccumulation (Igeo)

A common criterion to evaluate the heavy metal pollution in sediments is the Geoaccumulation index, which was originally defined by Muller [\(1979](#page-13-0)) to determine metal contamination in sediments, by comparing current concentrations with pre-industrial levels and can be calculated by the following equation (Muller [1979\)](#page-13-0),

$$
Igeo = Log_2 (Cn/1.5Bn)
$$
 (22.4)

where, Cn is the measured concentration of a heavy metal in sediment, Bn is the geochemical background value in average shale of element "n" and 1.5 is the background matrix correction due to terrigenous effects. The Igeo scale consists of seven grades (0–6) ranging from unpolluted to highly polluted (Table 22.1).

Fig. 22.2 Down-core variations of organic matter (TOC, TP, TN) and sediment components (sand, silt, clay) with vertical lines of average values

22.4 Results and Discussion

TOC ranges from 1.09 % to 2.57 % while TP and TN varied from 0.51 to 0.97 mg/g and 0.13 to 1.66 mg/g, respectively. Among the sediment components, sand ranged from 2.01 % to 13.65 %, silt from 23.15 % to 64.60 % and clay from 25.20 % to 70 %. The depth-wise plots of sediments components and metals are presented in the Figs. 22.2 and [22.3.](#page-6-0) Except for few fluctuations near the surface, from bottom to top, increasing trends of concentrations of TOC, TP and TN, can be observed from the Fig. 22.2. Sand exhibits an increasing trend from the bottom to the surface while silt and clay showed corresponding opposite trends. The variable admixture of sand, silt and clay fractions in the core reflected variable rates of erosion and deposition in the study area. In the case of elemental distribution, ranges of the metals were 4.09–5.49 % for Fe, 769–1090 ppm for Mn, 117–331 ppm for Cu, 26–67 ppm for Pb, 41–60 ppm for Co, 52–86 ppm for Ni, 123–223 ppm for Zn, 90–231 ppm for Cr and $5.51-9.95\%$ for Al. The depth profiles of Cu, Pb, Co, Ni and Zn (Fig. [22.3](#page-6-0)) showed a gradual increase from the bottom to the surface while Fe and Al showed more of a consistent trend. Mn showed an increase from the bottom to 20 cm depth of the core followed by a decrease till the surface. Cr displayed a fluctuating trend from the bottom to the surface. The order of the abundance of average

Fig. 22.3 Down-core variations of selected metals with vertical lines of average values

concentrations of metals in the core sediments were as follows; Al $>$ Fe $>$ Mn $>$ $Cr > Zn > Cu > Ni > Co > Pb$.

Al and Fe were found to be the main components of the sediment and their contents remained almost constant all through the studied profile. Increasing trends observed for most of the remaining elements especially, in the upper few centimetres of the core, can be related to high contents of the TOC. Baptista Neto et al. [\(2000\)](#page-13-0) and Marchand et al. ([2006](#page-13-0)) have shown that the heavy metal distributions in marine sediments are related to the strong association of metals and organic carbon in the form of organometallic complexes. Peak values were observed at 56 cm depth in the case of most of the studied elements, while high values were observed for Mn and Zn between the depths of 28 to 18 cm. It is well known that early diagenetic processes such as metal chelation by organic matter, scavenging by Fe and Mn oxides and redox reactions often produce decreasing or increasing metal concentrations along sediment profiles (Spencer et al. [2003\)](#page-14-0). Therefore, the observations seen in the core might be due to such processes.

22.4.1 Pearson's Correlation Coefficient

The role of organic matter and sediment grain size in relation to the accumulation of heavy metals in sediment has often been emphasised (Davies et al. [1991\)](#page-13-0). Pearson's correlation coefficient matrix of the sediment components, organic matter and selected metals is presented in the Table [22.2.](#page-7-0) From the table it follows that

the TOC showed significant association with TN, silt, Ni and Zn while TP was associated well with Mn, Pb, Ni and Zn. TN exhibited good association with Cu, Pb, Ni and Zn which indicated bonding with the organic matter. Taking into account of its high specific surface area, organic matter (OM) can form complexes with heavy metals and consequently influence their distribution (Loomb [2001\)](#page-13-0). Flocculation of organic matter leads to more adsorption surfaces, thus resulting in an increase of adsorption rates. Clay showed negative correlation with all the metals studied, except for Al, while silt showed good positive correlation with most of the metals. Fine sediment fraction can contain organic matter and Fe and Mn oxide surface coatings that favor adsorption processes and play an important role in controlling trace elements concentrations in sediment (Li et al. [2000;](#page-13-0) de Falco et al. [2003;](#page-13-0) Ip et al. [2007\)](#page-13-0).

22.4.2 Inter-element Correlation Coefficients

Significant associations are observed among all the metals studied, indicating their identical behaviour during incorporation in the estuarine sediments. Since Al and Fe were observed to be the major components of sediments, the higher values of correlation coefficient of Fe, Mn, Co and Ni with Al, suggested the simultaneous accumulation of these metals in the sediment matrix. Fe and Mn oxides/hydroxides have a high affinity with most trace metals and Fe often correlates with concentrations of other metals in the aquatic environment (Zabetoglou et al. [2002\)](#page-14-0). The observed correlation values suggested that Al (might be in aluminosilicate phase), Mn and Fe (in oxide phases) and organic matter have had a major role in the accumulation of trace metals in the sediments.

22.4.3 Multivariate Analysis

In order to understand the paths and the processes through which metal deposition occurred, factor and cluster analyses were employed. Cluster analysis represented by three main clusters is shown in the Fig. [22.4](#page-9-0). The first cluster comprises of Al and clay. The second cluster consists of Co, Fe, Cr, sand, Mn and TP suggesting the role of Fe, Mn and TP as dominant metal carriers. The third cluster is made up of two sub-clusters namely of Zn, Pb, Cu, TN and of silt, Ni, TOC indicating organic matter as the dominant metal carrier.

Factor analysis with varimax rotation accounted for four factors with a total variance of 78.35 % (Table [22.3;](#page-9-0) Variables with significant loadings are highlighted). Factor 1 explains 40.85 % of variance and showed significant positive loadings of TN with Mn, Pb and Zn, while factor 2 with 15.22 % variance exhibited significant loadings of Cu with Co and also with Fe and Al but to a lesser extent. Factor 3 with 12.04 % variance displayed significant associations of TOC and silt on Ni. Factor 4 with 10.23 % variance displayed significant loading of TP and good loadings of Fe, Zn and Cr. The first, third and fourth factors were governed by

Fig. 22.4 Hierarchical cluster analysis

	Factor 1	Factor 2	Factor 3	Factor 4	
Variables					
Eigen value	6.54		1.93	1.64	
% Total variance	40.85	15.22	12.04	10.23	
TOC	0.41	0.05	0.77	-0.01	
TP	0.20	0.07	0.06	0.93	
TN	0.87	-0.07	0.31	0.00	
Sand	0.01	-0.04	0.60	0.19	
Silt	0.18	0.18	0.78	0.06	
Clay	-0.16	-0.15	-0.92	-0.12	
Cu	-0.17	0.87	0.25	0.06	
Fe	0.20	0.60	0.21	0.68	
Mn	0.73	0.05	0.32	0.11	
Pb	0.95	0.09	-0.05	0.16	
Co	0.40	0.82	0.09	0.19	
Ni	0.62	0.34	0.54	0.21	
Zn	0.74	0.28	0.23	0.50	
Cr	-0.42	0.07	0.16	0.51	
Al	-0.13	0.60	-0.37	-0.59	

Table 22.3 Factor analysis matrix after varimax rotation

organic matter and might have originated from the same source, while the second factor was dominated by Fe. The similarity in element behaviour as seen from the factor and cluster analyses revealed that the metal variability was controlled by similar sources.

22.4.4 Pollution Indices

The EF values, calculated to evaluate the abundance of metals in the sediments, ranged from 0.84 to 1.26 for Fe, 1.16–2.09 for Mn, 2.46–9.07 for Cu, 1.27–3.89

Fig. 22.5 Down-core distribution of EF for selected metals (Fe, Mn, Cu, Pb, Co, Ni, Zn and Cr)

for Pb, 1.95–3.29 for Co, 0.72–1.55 for Ni, 1.17–2.86 for Zn and from 1.13 to 3.15 for Cr. The sediments were found to be enriched with metals in the following order: Fe \lt Ni \lt Mn \lt Zn \lt Cr \lt Pb \lt Co \lt Cu. The enrichment factor for Cu, Pb, Co, Ni, Zn and Cr were observed to be increasing from the bottom towards top (Fig. 22.5). The higher values of EF indicated an increase in metal deposition in recent years, probably reflecting the input of contaminants. These contaminants might be transported from the upstream industrial outfall, either in solution or particulate form, through tributaries which got subsequently deposited in the estuarine sediments. The low EF values for Fe and Ni indicated that these elements fall in the deficiency to minimal enrichment category and hence the enrichment of the sediment by these metals must be by natural processes. Further, from the Fig. 22.5, EF values of Fe and Ni were found to be close to unity and hence support the predominant terrigenous origin and devoid of the anthropogenic activities. The EF values for the remaining elements fall in the moderate enrichment category.

Table [22.4](#page-11-0) shows the Contamination Factor (CF) of each element and also the Pollution Load Index (PLI). Based on the CF calculation, maximum value for Cu and minimum for Fe was observed. Based on CF classification (Hakanson [1980\)](#page-13-0), Fe showed low contamination; Mn, Pb, Co, Ni, Zn and Cr showed moderate contamination while Cu exhibited considerable contamination. The PLI values, in general, increased from the bottom to the surface, with minor fluctuations. At a depth of 60 cm, PLI showed the lowest value (1.52) whereas highest value (2.07) was observed at a depth of 4 cm. Lower values of PLI (< 2) throughout the core implied no appreciable inputs from the anthropogenic sources. The Fig. [22.6](#page-12-0) shows

Depth(cm)	$CF-Fe$	CF-Mn	$CF-Cu$	$CF-Pb$	$CF-Co$	CF-Ni	$CF-Zn$	$CF-Cr$	PLI
$\overline{2}$	0.87	1.43	6.24	2.68	2.26	1.07	1.97	2.17	1.96
$\overline{4}$	0.96	1.49	4.40	3.38	2.76	1.20	2.10	2.29	2.07
6	0.95	1.44	3.56	3.15	2.74	1.22	2.02	1.93	1.94
8	0.96	1.53	3.71	3.04	2.64	1.20	2.07	1.83	1.94
10	0.95	1.49	3.57	2.99	2.61	1.22	2.02	1.09	1.79
12	0.96	1.56	3.64	2.93	2.67	1.27	2.01	1.30	1.86
14	0.95	1.50	3.46	2.65	2.72	1.22	1.97	2.51	1.95
16	0.99	1.65	3.34	2.58	2.66	1.15	2.12	2.36	1.96
18	0.98	1.72	3.37	2.84	2.62	1.18	2.42	2.35	2.02
$20\,$	0.95	1.75	3.17	2.64	2.51	1.12	2.24	2.14	1.92
22	0.98	1.72	3.25	2.69	2.62	1.19	2.21	2.32	1.98
24	0.94	1.73	3.15	2.20	2.59	1.12	2.10	2.51	1.90
26	0.95	1.69	3.14	2.85	2.78	1.18	1.95	2.26	1.95
28	0.98	1.65	3.25	2.23	2.79	1.12	1.83	2.18	1.86
30	0.96	1.48	2.79	1.48	2.21	1.10	1.63	2.10	1.63
32	0.96	1.49	2.89	1.64	2.30	1.06	1.57	2.02	1.64
34	0.96	1.55	2.83	1.50	2.34	1.07	1.56	1.92	1.62
36	0.95	1.41	3.06	2.13	2.36	1.10	1.91	1.64	1.70
38	0.97	1.48	2.89	2.14	2.38	1.11	1.68	1.70	1.69
40	0.97	1.51	2.82	1.83	2.47	1.09	1.46	1.24	1.57
42	0.98	1.45	2.92	1.45	2.29	1.04	1.56	2.07	1.61
44	0.97	1.47	2.86	1.40	2.38	0.96	1.50	2.47	1.62
46	1.00	1.46	2.91	1.38	2.42	1.01	1.53	2.78	1.67
48	0.95	1.44	2.80	1.31	2.32	0.95	1.47	2.40	1.58
50	0.95	1.46	2.84	1.55	2.42	0.96	1.54	2.63	1.66
52	0.94	1.39	2.87	1.44	2.45	0.99	1.43	2.55	1.62
54	0.92	1.39	2.73	1.56	2.28	0.93	1.30	2.17	1.54
56	1.16	1.82	3.71	1.58	3.18	1.18	1.84	2.29	1.93
58	0.95	1.50	2.86	1.68	2.53	0.76	1.70	2.34	1.64
60	0.93	1.28	2.77	1.70	2.16	0.93	1.30	1.92	1.52
62	0.91	1.32	2.69	1.89	2.41	0.90	1.46	2.07	1.59
64	0.90	1.31	2.61	2.18	2.39	0.87	1.43	2.03	1.59
66	0.89	1.36	2.66	2.24	2.43	0.91	1.48	1.57	1.57
68	0.93	1.49	2.71	2.33	2.39	0.94	1.57	1.96	1.67
70	0.94	1.41	2.96	2.31	2.59	0.98	1.71	1.99	1.73

Table 22.4 Depth-wise values of Contamination Factor (CF) and Pollution Load Index (PLI)

down-core variations of Geo-accumulation Index (Igeo) for the studied elements. From the figure, it is seen that all the elements are grouped in the unpolluted class except for Cu and Pb, which are found to be moderately polluted, especially in the upper portion of the core. Higher population and establishments in the estuarine area might have resulted in increased human activities accounting for the higher metal values. The use of Cu as anti-fouling agent on fishing trawlers and other commercial boats that are being operated in the study area might be one of the reasons for the increase in Cu concentration. Sources of Pb in sediment are the usage of leaded gasoline in boats and also emissions from automobiles, that consume leaded petrol (Ong [2006\)](#page-14-0). Igeo values calculated according to Muller's scale (Table [22.1](#page-4-0))

Fig. 22.6 Down-core distribution of Igeo for selected metals (Fe, Mn, Cu, Pb, Co, Ni, Zn and Cr)

indicated the level of pollution to range from no pollution to moderate pollution. The increased EF, PLI and Igeo values in the upper portion of the core can be attributed principally to the anthropogenic activities such as vehicular and industrial emissions, inflows of metal works and commercial establishments in the upstream region. Also industrial effluents from paint factories, sewage treatment plants and inflows from the tributaries might have contributed significantly.

22.5 Conclusions

The present study carried out to understand levels of metal pollution in the Ulhas estuary indicated that

- The coastal and estuarine regions acted as reservoirs of metal pollutants, principally drawn from the upstream regions of the Ulhas estuary that is bordered by urban, industrial and fishing establishments.
- Correlation, cluster and to some extent factor analyses of the studied variables suggested that the organic matter and Fe-Mn oxides played a major role in the distribution and abundance of trace elements in sediments.
- Based on the different indices of pollution, the area under investigation is unpolluted with respect to Fe, Mn, Co, Ni, Cr and Zn and moderately polluted by Cu and Pb.
- The metal concentrations and the pollution indices showed an upward increase in the depth-wise profile studied, meaning, the pollutants entered the coastal and estuarine region more recently.

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