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A study of Coastal Sediments of Sindh Pakistan Using Geochemical approach for evaluation of heavy metal pollution

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ABSTRACT: Twenty one locations at Coastal area of Sindh, Pakistan were selected in order to study the availability of heavy metals and their concentrations in the surficial sediments (<60µm). These soil samples were analyzed for Fe (Iron), Mn (Magnese), Cr (Chromium), Pb (Lead), Zn (Zinc), Cu (Copper), Co (Cobalt) and As (Arsenic) to examine metal concentrations in sediments. The concentration of metals Fe, Mn, Cr, Pb, Zn, Cu, Co and As were determined by atomic absorption spectrophotometer. Assessment of anthropogenic pollution in sediments, Enrichment Factor (EF) and Index of Geoaccumulation (Igeo) are calculated. The concentration of cadmium is indicating very high contamination in all the sampling sites. The observations suggested high EF values of the coastal sediments are polluted by Mn and Zn acts as a sink for heavy metals contributed from a multitude of anthropogenic sources in the study area possibly displays the effluent discharge. EFs data and geoaccumulation index values (Igeo) indicated that these trace metals predominately originate from anthropogenic sources. Considering the calculated EF and I_{geo} values, the increased I_{geo} values in south and south eastern part of the study areas are attributed principally to anthropogenic activities.
Key words: Heavy metals, anthropogenic pollution, Enrichment Factor, Geoaccumulation

INTRODUCTION

Soil is not only a medium for plants to grow or a pool to dispose of undesirable materials, but also a transmitter of many pollutants to surface, ground and coastal water. So the accumulated pollutants in surface soils ultimately transported to different environmental components of coastal aquifers. Soil analysis offers advantages over water analysis for the control and detection of metal pollution in estuaries (Forstner and Wittman, 1983), although metal concentrations may also fluctuate over time (Araujo *et al.,* 1988) and it is observed that in the study area rate of change is well below that of the water (Boyden *et al.,* 1979).

Trace elements found in soils/sediments is immobilized in water and thus could be involved in absorption, co-precipitation, and complex formation (Okafor and Opuene, 2007; Mohiuddin *et al.,* 2010). Sometimes they are co-adsorbed with other elements as oxides, hydroxides of Fe, Mn, or may occur in particulate form (Awofolu *et al*., 2005; Mwiganga and Kansiime, 2005). Their concentrations in stream and coastal sediment compartments can be used to reveal the history and intensity of local and regional pollution (Nyangababo *et al*., 2005a). Sentongo (1998); Matagi (1998) and Kansiime *et al.,* (1995).

In the study area, heavy metals enter into aquatic ecosystems mainly from anthropogenic sources, such as industrial wastewater discharges, sewage wastewater and fossil fuel combustion (Linnik and Zubenko, 2000). It has been observed worldwide that the impact of anthropogenic perturbation is most strongly felt by estuarine and coastal environments adjacent to the study areas (Nouri *et al.,* 2008)

In view of collection of soil samples for trace metals in the study area has been done to serve a basis for the planning of control strategies to achieve better environmental quality, and will as a key for an effective management of soil quality; similar extensive investigations of coastal soils have been carried out recently in many countries (Weiss *et al.,* 1994; Pouyat *et al.,* 1994). These soil samples were analyzed for Fe, Mn, Cr, Pb, Zn, Cu, Co and As to examine metal concentrations in sediment. Therefore, the study will attempt to evaluate the extent of heavy metal contamination from the surface to the bottom sediments and the degree to which heavy metals are influenced. The interrelationships among elements, and Enrichment Factor (EF), geoaccumulation index values (Igeo) is calculated to differentiate the origin of metals between anthropogenic and geogenic sources.

MATERIAL AND METHODS

A total twenty one samples of coastal sediments from same number of sites were collected during 2011-2012 (Figure 1) (Table 1). The estimation of the total metal concentration, from sediments was determined according to Oregioni and Aston (1984). Sediment samples were taken at a depth of 0-15 cm which was quickly packed in air tight plastic bottles. Sub-samples of the material were oven dried at 500C for 48 hours and ground using mortal and pestle. Then the samples were sieved by <63 μm sieving net and 2 gm of sub-sediment sample were digested by using acids mixtures (HNO3 +, HCLO4 and HF) respectively, to obtain the total concentration of the metals in the sediments as been recommended by Balcerzak (2002). Precautions were taken to avoid contamination during drying, grinding and sieving. Cd, Co, Ni, and Pb concentrations were determined with Atomic Absorption Spectrophotometer.
According to Ergin *et al.,* (1991), the metal EF is calculated by using this formula:

$$EF = \frac{(Fe/M)_{sample}}{(Fe/M)_{Background}}$$

Where Fe is the content of Fe in sample and M is the content of the metal in the sample.
And the geoaccumulation index (Igeo) introduced by Muller (1979) and calculated by using this formula:
$Igeo = \log_2 (Cn / 1.5 \times Bn)$
Where,
Cn - measured concentration of heavy metal in the sediment,
Bn - geochemical background value in average shale of element 'n'.
1.5 is the background matrix correction in factor due to lithogenic effects

Table No. 1: Sampling Sites during the Present Study

Sample No.	X	Y	Name of Locations
1	68°47'4.26"E	24°21'0.15"N	Qazi Muhammad, 1 Runn of Kutch
2	67°11'5.43"E	24°55'2.46"N	Qazi Muhammad, 2 Runn of Kutch
3	67°27'9.47"E	24° 8'20.63"N	Main keti Bandar (East)
4	67°27'9.59"E	24°8'0.43"N	Keti bandar 1
5	67°27'6.97"E	24°7'.78"N	Keti bandar 2
6	67°34'8.22"E	24°7'7.58"N	Port Qasim JT
7	67°33'9.03"E	24°78'7.37"N	Steel mill, Bin Qasim
8	67°3'9.91"E	24°78'7.63"N	Mazar Russian Beach, Back side of Steel Mill, Bin Qasim
9	67°14'0.41"E	24°78'4.84"N	Ibraheem Hyderi ,Korangi
10	67°02'7.72"E	24°80'3.22"N	Sea View, Hyper Star Building, Clifton
11	67°0'4.47"E	24°78'9.71"N	Sea View, Floating Ship, Clifton
12	66°58'11.15"E	24°48'13.99"N	Main Manora
13	66°58'17.29"E	24°47'48.93"N	Manora beach 1
14	66°56'41.81"E	24°48'56.98"N	Manora beach 2
15	66°98'3.42"E	24°8'3.36"N	Native Jeti Bridge, Left Side, Kemari
16	66°55'23.30"E	24°49'57.74"N	Main Sandspit
17	66°52'26.79"E	24°51'22.07"N	Main Hawks bay
18	66°50'54.48"E	24°51'40.73"N	Hawaks Bay 1
19	66°89'7.76"E	24°84'5.01"N	Hawaks Bay 2
20	66°77'5.79"E	24°84'5.51"N	Paradise Point 1
21	66°80'9.69"E	24°84'5.73"N	Paradise Point 2

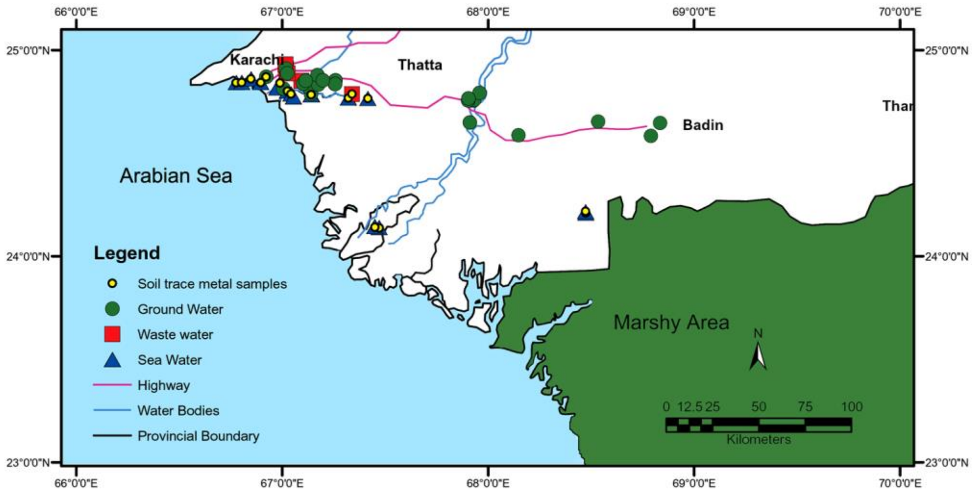


Figure 1: Map of the study area.

RESULT AND DISCUSSION

The primary purpose of sediment quality guidelines (SQGs) are to protect aquatic biota from the harmful and toxic effects related with sediment bound contaminants and is a useful tool for evaluating potential for contaminants within sediment to induce biological effects. The source of pollution is determined through the normalization of geoaccumulation values to the reference element. The degree of pollution in sediments can be assessed by determining the enrichment factor and indices such as Geo-accumulation index.

Enrichment Factor

The enrichment factor (EF) is a convenient measure of geochemical trends and is used for making comparisons between areas. Because of the natural origin of metal elements, the gross concentrations of metal elements don't show the anthropogenic contribution specifically. The assessment of heavy metals from anthropogenic contribution must be made clear. The natural occurrence of heavy metals complicates assessments of potentially contaminated estuarine sediments; measurable quantities of metals do not automatically infer anthropogenic enrichment in the estuary.

The enrichment factor is a good tool to differentiate the metal source between anthropogenic and naturally occurring (Morillo et al. 2004). EF is calculated to determine if levels of metals in sediments of coastal area of the province and its surrounding marine environment are of anthropogenic origins (e.g., contamination) shown in Table 2.

Differentiating the elements originating from human activities and those from natural weathering is an essential part of geochemical studies. One such technique largely applied is 'normalization' where metal concentrations were normalized to a textural or compositional characteristic of sediments. To identify anomalous metal concentration, geochemical normalization of the heavy metals data to a conservative element, such as Al, Fe, and Si was employed. Several authors have successfully used iron to normalize heavy metals contaminants (Schiff and Weisberg, 1999). Similarly in various geochemical studies, normalizing metals relative to Al or Fe is widely used to compensate for both the granulometric and mineralogical variability of metal concentrations in sediment (Chapman and Wang 2001). Statistical methods can also be used to determine the reference element as metal concentration normalizer (Liu et al. 2003). Selvam P.A et al 2011, found from the linear relationship strength of metals with Al and Fe confirms the applicability of Fe as the most appropriate normalizer element in their study area. Several authors have used Fe to normalize heavy metal contaminant in estuarine sediments (Schiff and Weisberg 1999; Neto et al. 2000). In this study, iron is also used as a conservative tracer to differentiate natural from anthropogenic components.

EF values were interpreted as suggested by Birch (2003) where $EF < 1$ indicates no enrichment, < 3 is minor; 3-5 is moderate; 5-10 is moderately severe; 10-25 is severe; 25-50 is very severe; and > 50 is extremely severe. A value of $0.5 \leq EF \leq 1.5$ suggests that traces of metal may be due to crustal materials or natural weathering processes. Samples having EFc value greater than 5 are considered to be contaminated with that particular element.

From table no. 2 (EF) represents the EFc values of all the trace metals measured in coastal sediments. In the industrial and agricultural waste area all the sampling sites have EFc values are $\leq$ 2 shows minor enrichment except at site 1 and 2 for Mn & Zn. In the same way at industrial and municipal waste fishing harbor sampling site EFc values are also $\leq$ 2 showing minor enrichment at all the sampling sites except for Mn and Zn. Similarly EFs values at beach were also $\leq$ 2 shows minor enrichment except for Mn. EFc values < 5 are considered significant. Areas with EFc values <1 should be viewed with caution as they imply preferential release of these metals, making them bioavailable. If the value of EFc >1.5, the trace metal is delivered from others sources suggesting environmental contamination by that particular trace element. It is suggested that high EFc values indicate an anthropogenic source of trace metals, mainly from activities such as industrialization, urbanization, deposition of industrial values and others. Since, the bioavailability and toxicity of any trace metals in sediments depend on the chemical form and concentration of the metal, it can be inferred that trace metals in sediment samples with high EFc values, along with higher labile fractions in sediments are potential sources for mobility and bioavailability in the aquatic ecosystems.

EFs close to unity point indicate crustal origin whiles those greater than 10 are considered to be non-crustal source (Nolting et al., 1999). In the study area minimum EFs obtained for the elements (Cr, Cu, Fe, Pb, Co, Zn and As) are less than unity in most of the samples implying that these elements are depleted in some of the phases relative to crustal abundance in the study area. However, it is evident that all elements with an EFs value greater than unity reveal sediment contamination, for example the higher EFs values for Mn and Zn. An overall higher EF values for Mn particularly and Zn generally in all the three functional area consider both upper and lower littoral zones suggest the presence of contaminated sediments derived from multifarious sources like domestic sewage, power-plant operation, major storm events or dumping of sediments dredging along the international shipping zones as endorsed by Batley and Brockbank (1990).

Table No. 2: Enrichment factor values of trace metals for coastal sediments

Sample No.	Name of Locations	Cr	Cu	Fe	Mn	Pb	Co	Zn	As
1	Qazi Muhammad, 1Runn of Kutch	0	0	1	10	1	0	6	1
2	Qazi Muhammad, 2 Runn of Kutch	0	0	1	7	1	0	5	1
3	Main keti Bandar (East)	1	1	1	2	1	1	1	1
4	Keti bandar 1	1	1	1	2	1	1	1	1
5	Keti bandar 2	1	0	1	2	0	1	1	1
6	Port Qasim JT	1	1	1	1	1	1	1	1
7	Steel mill, Bin Qasim	1	1	1	1	1	1	1	1
8	Mazar Russian Beach, Back side of Steel Mill, Bin Qasim	1	1	1	1	1	1	1	1
9	Ibraheem Hyderi Korangi	1	1	1	3	1	1	1	1
10	Sea View, Hyper Star Building, Clifton	1	1	1	20	1	1	3	1
11	Sea View, Floating Ship, Clifton	1	1	1	21	1	1	3	1
12	Main Manora	2	1	1	15	1	1	25	1
13	Manora beach 1	2	1	1	13	1	1	20	1
14	Manora beach 2	1	1	1	13	1	1	22	1
15	Native Jeti Bridge, Left Side, Kemari	1	1	1	14	1	1	22	1
16	Main Sandspit	1	1	1	22	1	1	3	1
17	Main Hawks bay	1	1	1	12	1	1	2	1
18	Hawaks Bay 1	1	1	1	13	1	1	2	1
19	Hawaks Bay 2	1	1	1	13	1	1	2	1
20	Paradise Point 1	1	1	1	10	1	1	2	1
21	Paradise Point 2	1	1	1	11	1	1	2	1

Following the interpretation of (Birch 2003) shows no enrichment with respect to Cr, Cu, Fe Pb, Co and As in the study area, although high EFs value of Mn and Zn shows severe enrichment. EFs (<1) in most of these metals in the sediment indicate its origin predominantly from lithogenous material and suggest the absence of contamination by these metals in the study region. Almost at all sites, Mn and Zn indicate severe enrichment particularly in south and south west sampling stations suggesting the anthropogenic input of these metals. The observations suggest that the coastal sediment is polluted by Mn and Zn acts as a sink for heavy metals contributed from a multitude of anthropogenic sources. High EF values of Mn and Zn in the study area possibly displays the effluent discharge of nearby chemical industries (fertilizers, heavy metal processing, pesticides, insecticides, petrol refining, chemical and allied industries) and urban activities through Indus river. In the south and south east estuary industrial effluents, flow pattern of Indus river and synchronous tides lead to high deposition of

Mn in sites (9-10) and Zn in points (1, 2, 12-15) whereas agricultural and domestic activities influence major portion in the south eastern part of the estuary makes less pollution compared with the south and south west.

Geoaccumulation Index
The geoaccumulation index (Igeo) introduced by Muller (1979) for metal concentrations in the
< 2 lm fraction and developed for the global standard shale values. This is used here to assess metal pollution in sediments of the coastal area of the province.

However, several researchers (Subramanian and Mohanachandran, 1990; Barreiro, 1991; Sahu and Bhosale, 1991) have used the previous expression using regional backgrounds and on the less than 63±65 lm sediment fraction. Muller (1981) proposed seven grades or classes of the geo accumulation index. Different geo accumulation index classes along with the associated sediment quality are given in table 3.

Table no. 3: Geoaccumulation index classes to assess sediment quality

0-0	< 0	Uncontaminated
0-1	1	Uncontaminated to moderately contaminated
1-2	2	Moderately contaminated
2-3	3	Moderately to highly contaminated
3-4	4	Highly contaminated
4-5	5	Highly to very highly contaminated
5-6	6	Very highly contaminated

Appendix 1: Geoaccumulation index Igeo values were calculated for different metals as introduced by Muller (1969) is as follows:

S. No.	**Cr**	**Cu**	**Fe**	**Mn**	**Pb**	**Co**	**Zn**	**As**
A1	0.0	0.0	-0.6	0.2	0.7	0.0	0.8	-9.4
A2	0.0	0.0	-0.2	0.1	1.4	0.0	0.9	-10.4
A3	-1.8	-0.6	2.0	-0.3	-0.6	-0.5	-0.2	1.8
A4	-0.1	-0.3	1.5	-0.6	-0.6	-0.6	-0.6	2.2
A5	-0.6	0.0	1.6	-0.5	0.00	-0.6	-0.3	1.7
A6	5.0	1.2	6.4	2.6	3.6	0.1	3.0	0.6
A7	4.7	0.9	6.3	2.2	3.6	0.3	2.6	0.3
A8	4.5	0.6	6.0	2.4	3.2	-0.1	2.9	-0.6
B09	5.1	1.4	4.8	3.7	3.3	0.4	2.5	-5.1
B10	-0.6	-1.2	1.4	3.3	1.9	0.8	1.6	-5.3
B11	-0.2	-0.9	1.3	3.3	2.1	1.0	1.5	-4.6
B12	4.6	0.8	0.8	2.3	2.6	1.0	4.4	-4.5
B13	4.7	1.1	1.0	2.3	2.9	1.1	4.3	-4.8
B14	4.6	0.9	0.9	2.2	2.7	1.0	4.4	-6.1
B15	3.9	1.0	1.0	2.3	3.1	1.5	4.5	-4.3
C16	0.4	-0.4	-0.6	-0.2	-0.6	-0.4	-0.4	-2.9
C17	0.0	0.5	0.2	-0.4	-0.4	-0.3	-0.5	-1.3
C18	-0.6	-0.4	0.2	-0.2	-0.3	-0.2	-0.2	-3.3
C19	0.2	-0.5	0.1	-0.3	-0.2	-0.2	-0.3	-3.9
C20	-0.3	-0.3	0.5	-0.3	-0.4	-0.6	-0.4	3.8
C21	-0.6	-0.6	0.7	-0.1	-0.5	-0.5	-0.6	3.0

The I_{geo} class 0 indicates the absence of contamination while the I_{geo} class 6 represents the upper limit of the contamination. The highest class 6 (very strong contamination) reflects 100-fold enrichment of the metals relative to their background values.

I_{geo} calculation is carried out for each functional area separately for the coastal sediments of the study area. The calculated values for coastal sediments and its surrounding marine environment are given in **appendix 1** and their model representations are in Fig. 1 (a, b & c).

The values for Cr and Fe in the coastal sediments acquired from industrial and agricultural waste area exhibited class I_{geo} 4-5 and I_{geo} 6-7 and hence they are very highly contaminated with these metals. From Fig.1 (a) it can be interpreted that sampling stations from the industrial area of the

Port Qasim may face a severe trace metal contamination problem with respect to Cr and Fe, while other trace metals such as Pb, Mn, Zn are moderately contaminated, and As and Cu has not shown any significant impression of pollution i.e., uncontaminated.

The functional area pertaining to industrial and municipal waste (fishing harbor) (Fig. 1b) are somewhat showing similar picture as of industrial and agricultural waste area. Here Cr and Zn exhibited in I_{geo} class 4-5 highly polluted, Mn and Pb exhibited in I_{geo} class 2-3 moderately

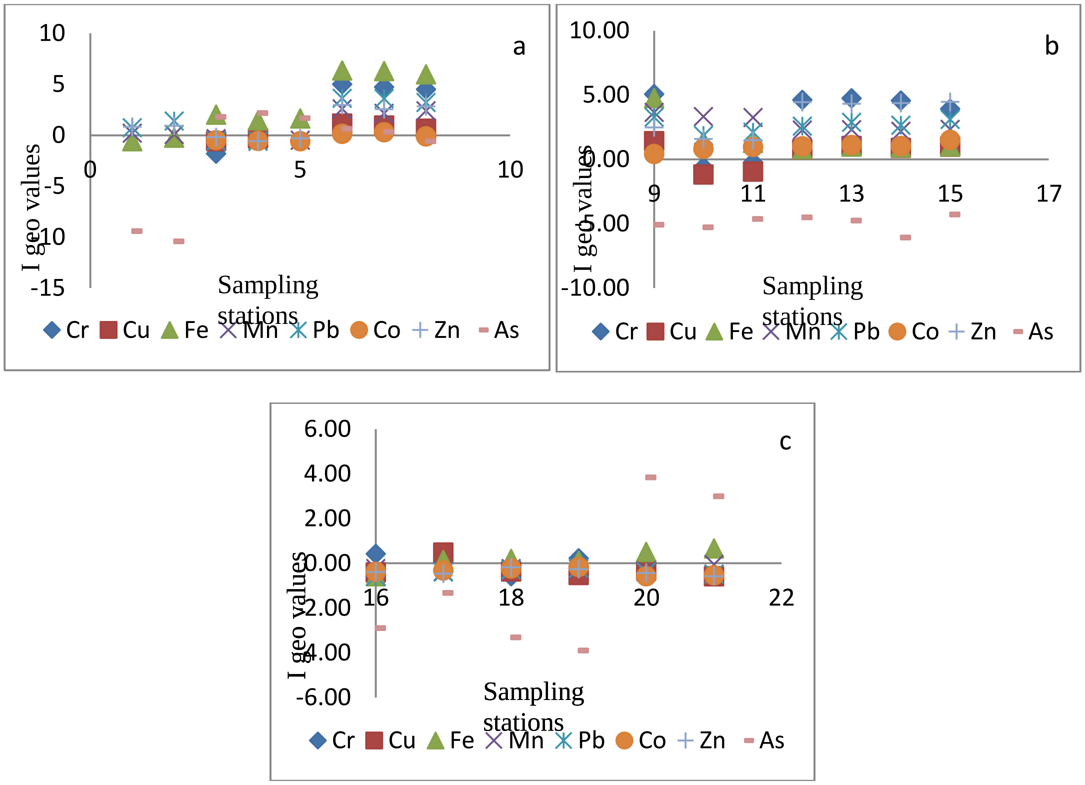


Figure 1: *Igeo* values of different sampling sites at coastal belt of Sindh.
a. Industrial and agriculture wastes surface sediments
b. Industrial and municipal wastes (at fishing harbor) surface sediments
c. Beach / creek surface sediments

Polluted sediments that could be attributed to terrigenous sources, Cu and Co exhibited in I_{geo} class 0-1unpolluted, while the Arsenic has not show any marked contamination in this functional area. While the sediments collected from the beach / creek (Fig.1c) does not show any significant impact of contamination with any trace metal studied.

The I_{geo} values for Arsenic fall in class '0' in most of the samples collected, indicating background concentration in all the sites at different functional areas are not polluted with this metal. Due to varying quality of sediment and local contamination I_{geo} values of Fe, Cr, Zn, Pb and Mn fall into five classes 6,5,4,3 and 2 indicating moderate to very high contamination which is attributed to the sludge, sewage plant, wastewater discharge and industrial effluent.

Figure 2 shows the range and average values of *Igeo* values for each metal, using local reference material in the form of box and whisker plot. Based on average values of *Igeo*, the ranking of intensity of heavy metal pollution of the functional area of industrial and agriculture wastes surface sediments are as follows: Fe > Cr > Pb > Zn > Mn > Cu > Co > As. Similarly the *Igeo*, ranking of intensity of heavy metal pollution of the functional area of industrial and municipal wastes (at fishing harbor) surface sediments are as under: Cr > Fe > Zn > Pb > Mn > Co > Cu >As. In the same way *Igeo*, ranking of intensity of heavy metal pollution of the functional area beach/creek surface sediments are mentioned below: As > Fe > Cu > Cr > Mn > Co > Pb > Zn

From figure 3, functional area belongs to industrial and agriculture waste can be displays sample percentages in Müller classes for Cr, Cu, Fe, Mn, Pb, Co, Zn and As. Co, Cr, Cu Pb, Zn and As concentrations fall mainly in classes '0'. For Cr mainly fall in class '0' (62.5% of total samples) and the remainder 37.5% in class 6. Cu mainly fall in class '0' (62.5%) and the remainder 37.5% in class 1. Fe fall in class '0' (25%) and the remainder 37.5% in class 6 and 7 each. For Mn mainly fall in class 1 (62.5%) and the remainder 37.5% in class 4. Pb samples mainly fall in class

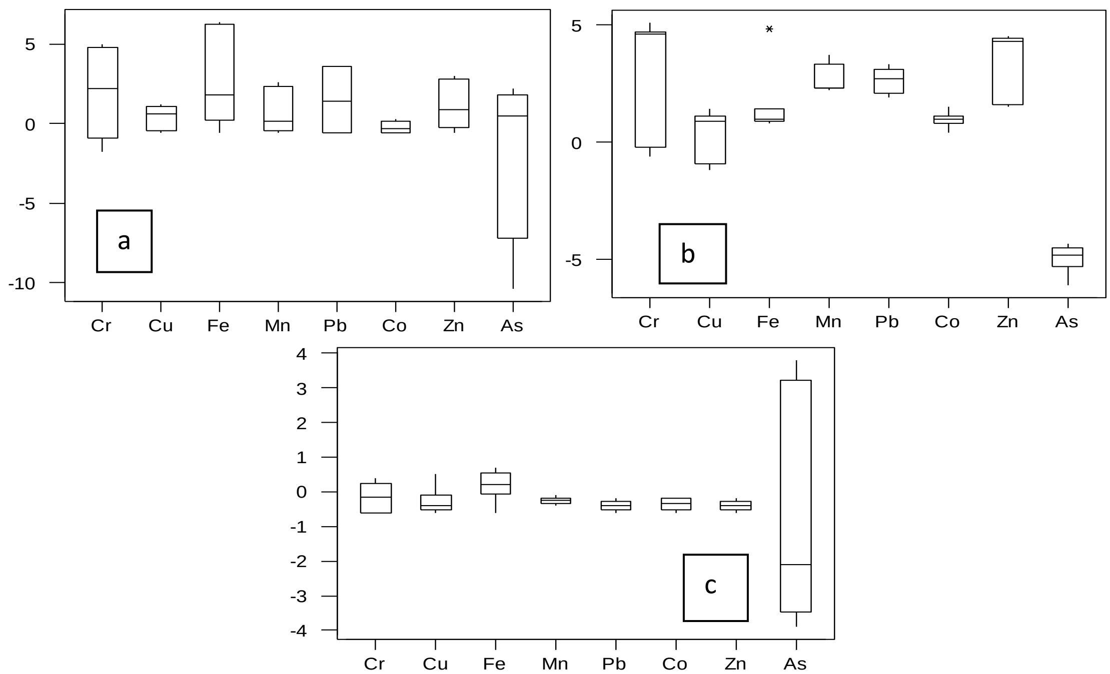
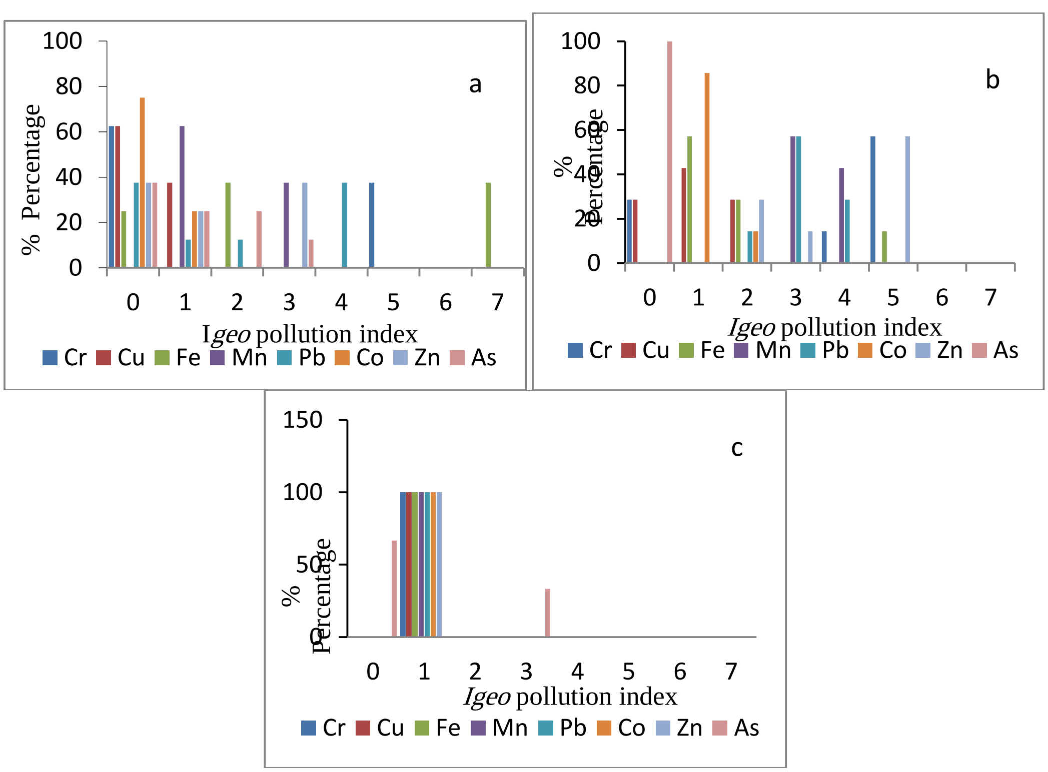


Fig. 2. Box-and-whisker plots of the geoaccumulation index
(a) Industrial and agriculture wastes surface sediments.
(b) Industrial and municipal wastes (at fishing harbor) surface sediments.
(c) Beach / creek surface sediments



a. Beach / creek surface sediments

Figure 3: Percentage of I*geo* pollution index of different sampling sites.

a. Industrial and agriculture wastes surface sediments
b. Industrial and municipal wastes (at fishing harbor) surface sediments
c. Industrial and municipal wastes (at fishing harbor) surface sediments

Similarly functional area of industrial and municipal waste (fishing harbor) displays that As concentrations fall mainly in classes ‘0’. For Cr mainly fall in class 5 (57.1% of the total sample) and the remainder 28.6% and 14.3% are in class ‘0’ & 4. Cu fall in class ‘0’ (28.6%) and the remainder 42.9% and 28.6 % are in class 1 & 2. Fe mainly fall in class 1 (57.1%) and the remainder 28.6% and 14.3% fall in class 2 & 5. Mn included 57.1% and 42.9% in class 3 and 4. Pb mainly falls in class 3 (57.1%) while the remainder 14.3% and 28.6% fall in class 2 and 4. Co fall 85.7 % of total samples in class 1 and 14.3% are in class 2. For Zn, 28.6% of the samples fall in class 2, 14.3% in class 3 and 57.1% in class 5, and As 100% fall in class ‘0’.

These results indicate that the surface sediments of this functional area can be categorized as follows: unpolluted with Cr, Cu, and As (average *Igeo*<0), unpolluted to moderately polluted with Cu, Fe and Co (0< average *Igeo*<1), moderately polluted with Cu, Fe, Pb, Co and Zn (1< average *Igeo* < 2), moderately to highly polluted with Mn, Pb and Zn (2< average *Igeo*<3), strongly polluted with Cr, Mn and Pb (3< average *Igeo*<4), and strongly to extremely polluted with Cr, Fe and Zn (4< average *Igeo*<5). In the same way surface sediment collected from the functional area of beaches and creeks along the coastal belt of the province demonstrates that 66.6% and 33.3% of the samples fall in class ‘0’ and 3. While the rest of the trace metal (100% of total samples) fall in class 1 i.e. unpolluted to moderately polluted (0< average *Igeo*<1).

Fig. 3 reflects that, independent of the background used in three different functional area such as industrial and agricultural waste area, industrial and municipal waste (fishing harbor) and beach / creek area having 40%, 100% and 70% of the samples fall in Class 0 (background concentrations) especially for the metals As, i.e., it leads us to conclude that in the study area this metal has not shown any significant impact. While the contamination status in sediments with respect to trace metal depending on mobility and solubility of these metals that can be displaced from discharge places.

Considering the calculated EF and I_{geo} values, the increased I_{geo} values in south and south eastern part of the study area are attributed principally to anthropogenic activities. In general terms, the south and south eastern part of the study areas are most affected by pollution, while south western part are unaffected.

CONCLUSION

Enrichment factor has showed no enrichment with respect to Cr, Cu, Fe Pb, Co and As in the study area, although high EFs value of Mn and Zn indicated severe enrichment. High EF values of Mn and Zn in the study area possibly displays the effluent discharge of nearby chemical industries (fertilizers, heavy metal processing, pesticides, insecticides, petrol refining, chemical and allied industries) and urban activities through Indus river. Geo accumulation index reflects that, independent of the background used in three different functional area such as industrial and agricultural waste area, industrial and municipal waste (fishing harbor) and beach / creek area having 40%, 100% and 70% of the samples fall in Class 0 (background concentrations) especially for the metals As, i.e., it leads us to conclude that in the study area this metal has not shown any significant impact. While the contamination status in sediments with respect to trace metal depending on mobility and solubility of these metals that can be displaced from discharge places. EFs data, geoaccumulation index values (Igeo) concluded that these trace metals predominately originate from anthropogenic sources.

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