Oceanologia 64 (2022) 276—286

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ORIGINAL RESEARCH ARTICLE

Ecological assessment of heavy metals accumulation in sediments and leaves of *Avicennia marina* along the Diu coast of the northeast Arabian Sea

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Received 6 May 2021; accepted 3 December 2021
Available online 17 December 2021

KEYWORDS
Ecological assessment;
Heavy metals;
Sediments;
Avicennia marina;
Diu coast

Abstract The Coastal region of Diu is the natural habitat dominated by *Avicennia marina* mangrove species at the southeast coast of Saurashtra in Gujarat state of India. However, Diu being a famous industrial and tourism place survival of these mangrove species is threatened due to anthropogenic activities. In present studies, sediment and leaf samples of *A. marina* were collected from the Diu coast to evaluate the ecological threat of heavy metals accumulation in the marine habitat. There was remarkable presence of heavy metals such as copper, nickel, cadmium, chromium and lead in sediments and leaf samples of *A. marina*. The values of Biological concentration factors (BCFs) of heavy metals in leaf samples were high for cadmium, chromium and lead which suggest chelation of these heavy metals with biomolecules. The geo-accumulation index suggested that Site-4 and Site-5 were heavily contaminated with copper and nickel. The ecological risk index suggested that there is no significant effect of heavy metals on growth of plants in the mangrove ecosystem. Principal component analysis revealed that the samples collected from the natural habitats (Site-4 and Site-5) near the fishing and industrial areas were the main sources of heavy metal contamination. Hence, it was concluded that the concentration of heavy metals in the studied ecosystem had limited impact on growth of plants at Site-1, Site-2 and Site-3. However, growth of plants at Site-4 and Site-5 were threatened due to the toxic effect of copper and nickel present in its sediments.

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Peer review under the responsibility of the Institute of Oceanology of the Polish Academy of Sciences.

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https://doi.org/10.1016/j.oceano.2021.12.002

1. Introduction

The coastal region of the Diu with vast tidal flats is one of the richest zones for mangroves along the west coast of India. It is the natural habitat of *Avicennia marina* in the Gulf of Khambhat. Conservation of this natural habitat of *A. marina* is important as it is the only island dominated by these plants. *A. marina* species are distributed sparsely in these regions in the form of patches and survival of these mangrove patches is significantly threatened due to anthropogenic human activities in subtidal and intertidal zone of coastal region. As it is a famous tourism place and industrial zone of Gujarat, it is continuously influenced by urbanization activities. This strong anthropogenic forcing has a highly negative impact on subtidal and intertidal habitats within the region. Major industries present in the Diu region are polyester, cotton yarn, plasticizers, paper petroleum byproducts, pharmaceuticals, plastics, electrical conductors and marble tiles. These industries are known to induce high concentrations of heavy metals in water and sediments of the marine ecosystem through pollutants from urban runoff and industrial waste disposal (Bodin et al., 2013; Marchand et al., 2006).

Various studies have depicted that mangroves have ability to survive in changing environmental conditions such as alternating floods, low oxygen level in sediments, changing osmotic potential, organic, inorganic or metal pollution (Buajan and Pumijumnong, 2010; Li et al., 2016). Even, they have ability to accumulate heavy metals in aerial roots and translocate them to other organs of the plant (Fernández-Cadena et al., 2014; Li et al., 2016; Usman et al., 2013). The concentrations of heavy metals were found high in aerial roots of mangrove species such as *Avicennia, Rhizophora* and *Kandelia* as compared to shoots (Chiu et al., 1995; MacFarlane and Burchett, 2002; Peters et al., 1997; Tam and Wong, 2000; Thomas and Fernandez, 1997). Heavy metals are hazardous to mangrove plants if its concentration increases above 5 gm/cm^3. They threaten survival of mangroves by affecting on their metabolic processes which in turn reduces growth and reproduction in plants (Wright and Welbourn, 2002). These in turn reduces density and diversity of plant species in the ecosystem (Järup, 2003; Yan et al., 2017). Therefore, the ecological assessment and subsequent protection measures framework for preserving the only natural habitat of *A. marina* are a necessity needed promptly.

2. Material and methods

2.1. Geographical locations of the study area

Present research was conducted on five locations where *A. marina* inhabits the coastal region of Diu. These five locations were selected for this research on the basis of the geographical dominance of *A. marina* species, the anthropogenic activities, aggregation of fishing community and sewage pollution in the area. The sites were located at the following geographical locations: 1) Site-1 (Near Goghala bridge: GPS 20°43′46.66″N 70°59′17.52″E); 2) Site-2 (Opposite Jethibai Bus Station: GPS 20°43′7.31″N 70°58′52.16″E); 3) Site-3 (Airport road, Diu: GPS 20°42′51.90″N 70°57′28.26″E); 4) Site-4 (Behind Goa Industrial Development Corporation area: GPS 20°42′57.40″N 70°56′59.87″E); 5) Site-5 (Taad village bridge: GPS 20°44′7.35″N 70°55′48.07″E) (Figure 1).

2.2. Sediments samples collection and heavy metals analysis

Heavy metals analysis from sediments and leaves samples was done by following the method prescribed by Enders and Lehmann (2012) with some modifications. Sediments and leaves samples were collected from five different places (Site-1 to Site-5) in plastic collection vessels. Sediments and leaf samples were dried in an oven at 70°C and then grinded using mechanical grinder. The powder of samples was passed through 0.3 mm size sieves. Further dry ash oxidation method was used to digest the powder samples of soil and leaves. The well grinded dry powder of each sample was filled in cruscible of muffle furnish. In Muffle furnesh ash of each sample powder was prepared by incubating the cruscible for 5 hrs. at 550°C. Then after cooling ash was dissolve in 5 ml of 25% HCl. The mixture was filtered using acid wash filter paper and final volume of 50 ml was prepared using distilled water. Concentration of all microelements (Mg, S, B, Fe, Zn, Cu, Mn, Ni) and heavy metals (As, Cd, Cr, Pd, Hg, Se) were determined from this digested sample solution using ICP-MS (ICAP Q Thermo Fisher Scientific, Waltham, MA, USA). Samples were analyzed in triplicate using the following operation conditions of instrument: Power=1550W, cool gas flow=14.1 L/min, nebulizer gas flow = 0.94 L/min, auxillary gas flow = 0.79 L/min, dwell time =0.01 s, perostatic pump speed = 40 rpm, total time for each sample measurement= 3 min.

2.2.1. Comparison of heavy metal estimation data with sediment quality guidelines (SQGs)

The data obtained in present studies was compared with sediment quality guidelines (SQGs) as described by Bakan and Özkoc (2007), Luo et al. (2010) and MacDonald et al. (2000). These guidelines consists of the data on probable effect level (PEL), threshold effect level (TEL), lowest effect level (LEL), effect range low (ERL), toxic effect threshold (TET), effect range median (ERM), and severe effect level (SEL) of heavy metals in coastal regions.

2.2.2. Determination of the biological concentration factor (BCF) in leaves samples

The biological concentration factor (BCF) of leaves was calculated to evaluate the accumulation level of heavy metals in plant tissues from sediments. BCF values for leaves were expressed as original data averages for the heavy metals such as Cu, Ni, Cd, Cr, and Pb. The BCF values were calculated based on the equation described by Cui et al. (2007) and Yoon et al. (2006):

$$BCF_{leaf}=C_{leaf}/C_{sediment}$$

where C_{leaf} are the heavy metal concentrations in the leaf and $C_{sediment}$ is the heavy metal concentration in the sediment.

Figure 1 Topography of study area which shows the five natural habitats of *Avicennia marina* at Diu coast.

2.2.3. Determination of geo-accumulation index (I_{geo})

Geo-accumulation index (I_{geo}) of heavy metals was determined to assess the changes of concentration of heavy metals by associating it with the heavy metal concentrations in aquatic sediments with the geochemical background. It was calculated by following the equation described by Müller (1969)

$$I_{geo} = \log_2 C_n/(1.5)B_n$$

where, C_n is the concentration of metal measured in mangrove sediments, B_n is the geochemical background value of the earth's crust (Taylor and McLennan, 1985).

1.5 is a constant applied to account for the potential variability in the reference value due to the influence of lithogenic processes.

I_{geo} values can be categorized in the following manner: uncontaminated sediment when $I_{geo} = 0$; uncontaminated to moderately contaminated sediment when $0 < I_{geo} < 1$; moderately contaminated sediment when $1 < I_{geo} < 2$; moderately to heavily contaminated sediment when $2 < I_{geo} < 3$; heavily contaminated sediment when $3 < I_{geo} < 4$; heavily to extremely contaminated sediment when $4 < I_{geo} < 5$; and extremely contaminated sediment when $5 < I_{geo}$.

2.2.4. Determination of the probable ecological risk coefficient (E^i_r)

The probable ecological risk coefficient (E^i_r) was determined by following the formula given by Hakanson (1980)

$$E^i_r = T^i_r * C^i_r = T^i_r * C^i_r / C^i_n$$

where, T^i_r values for measured heavy metals are Cr = 2, Cu = 5, Ni = 5, Pb = 5 and Cd = 30; C^i_r is the contamination factor, C^i_s is the concentration of heavy metals in the sediment, C^i_n − a background value for heavy metals and T^i_r is the metal toxic response factor.

E^i_r values can be categorized in the following manner:

$E^i_r < 40$ then low risk, $40 < E^i_r < 80$ then moderate risk, $80 < E^i_r < 160$ then considerable risk, $160 < E^i_r < 320$ then high-risk and $E^i_r > 320$ then very high risk

2.2.5. Statistical analysis

Multivariate cluster analysis was performed to construct a dendrogram based on the similarity matrix data using the paired group (UPGMA) method with arithmetic averages and Euclidean similarity index. All the measured parameters were also subjected to Principle component analysis to determine significant relationship of one component with another. Comparisons and similarity groupings of all measured parameters were done by using two-way ANOVA to determine significant variation between means. All the analysis was performed using Past: Palaeontological Statistics software package.

3. Results

3.1. Sediments quality assessment at natural habitats of *Avicennia marina*

From present studies, it was observed that the mean concentration of heavy metals in sediments varied in range from 1.34 to 2.64 μg/g for copper, 0.028 to 2.093 μg/g for nickel, 0.023 to 0.09 μg/g for cadmium, 0.012 to 0.099 μg/g for chromium, and 0.024 to 0.098 μg/g for lead. The con-

Table 1 Variation in mean concentration of heavy metals accumulation in leaves samples due to season changes.

ANOVA *Source of Variation*	*SS*	*df*	*MS*	*F*	*P-value*	*F crit*
Sites	0.000105	4	2.63E-05	6.701746	**0.000176**	3.674045
Seasons	0.032951	14	0.002354	600.3941	**1.24E-55**	2.417951
Error	0.00022	56	3.92E-06			
Total	0.033276	74				

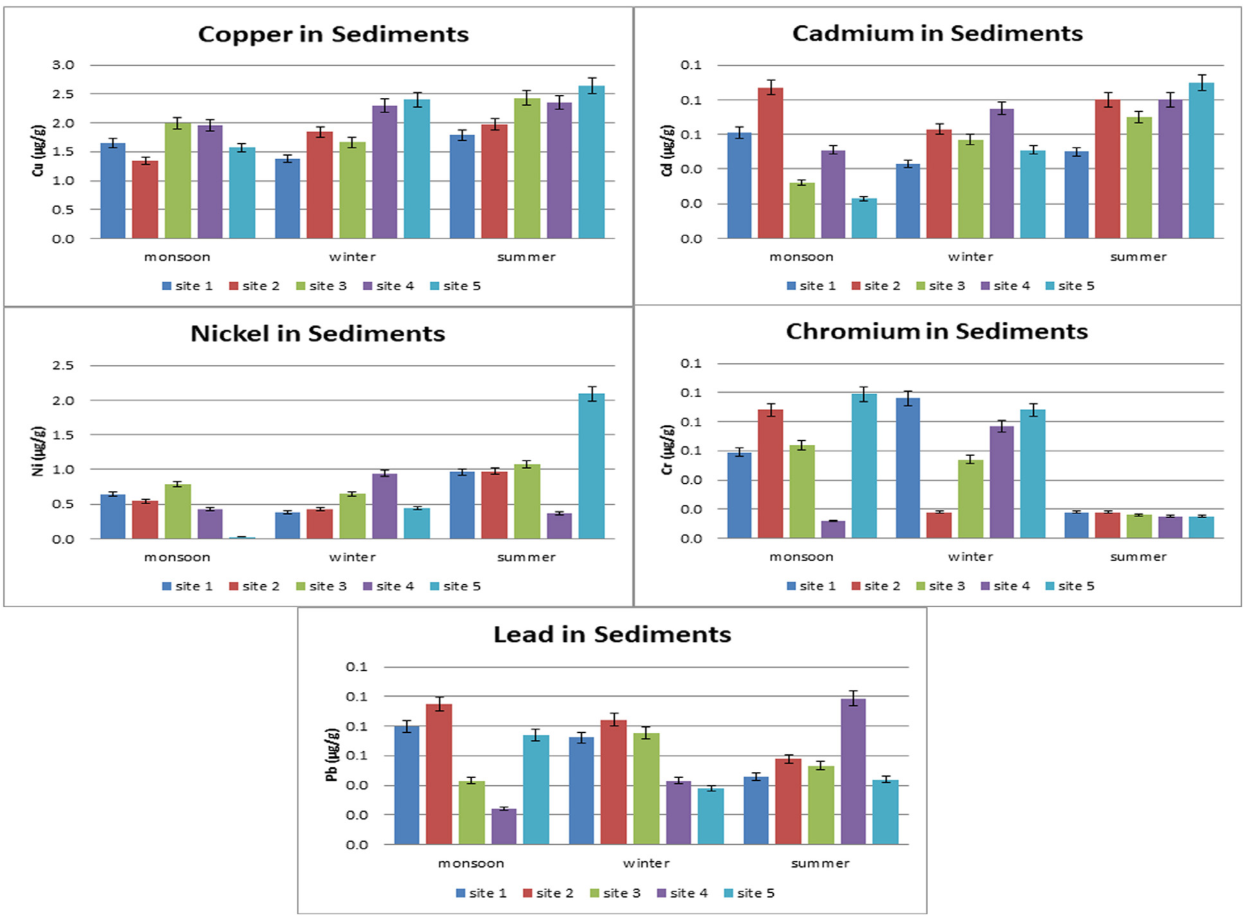


Figure 2 Changes in the mean concentration of heavy metals in sediments at all the five natural habitats of *Avicennia marina* during different seasons.

centrations of heavy metals in sediments were in following order Cu>Ni>Cd>Cr>Pd. There was remarkable significant variation ($p<0.01$) of mean heavy metals concentration at all the five natural habitats of *A. marina* at Diu coast (Table 1). There was also significant ($p<0.01$) influence of seasonal changes on concentration of heavy metals in sediments of all the five natural habitats of *A. marina* at Diu coast. The concentration of copper was high during summer season as compared to winter and monsoon season at all the five habitats. The copper concentration remained high at Site-4 and Site-5 as compared to other sites (Figure 2). The concentration of nickel was high during summer season as compared to monsoon and winter season at all the five habitats (Figure 2). The concentration of cadmium was high during summer and winter season as compared to monsoon. The cadmium concentration remained high at Site-4 and Site-5 as compared to other sites (Figure 2). The concentration of chromium was high during monsoon and winter season as compared to summer. The chromium concentration remained high at site-1 and Site-5 as compared to other sites (Figure 2). The concentration of lead was high during winter and monsoon season as compared to summer. The concentration of lead remained high at Site-2 and Site-4 as compared to other sites (Figure 2).

Table 2 Variation in the mean concentration of heavy metals in sediments at all the five natural habitats due to seasonal changes.

ANOVA

Source of Variation	*SS*	*df*	*MS*	*F*	*P-value*	*F crit*
Sites	0.255424	4	0.063856	1.053742	**0.387995**	3.674045
Seasons	42.59691	14	3.042636	50.20916	**1.74E-26**	2.417951
Error	3.393557	56	0.060599			
Total	46.24589	74				

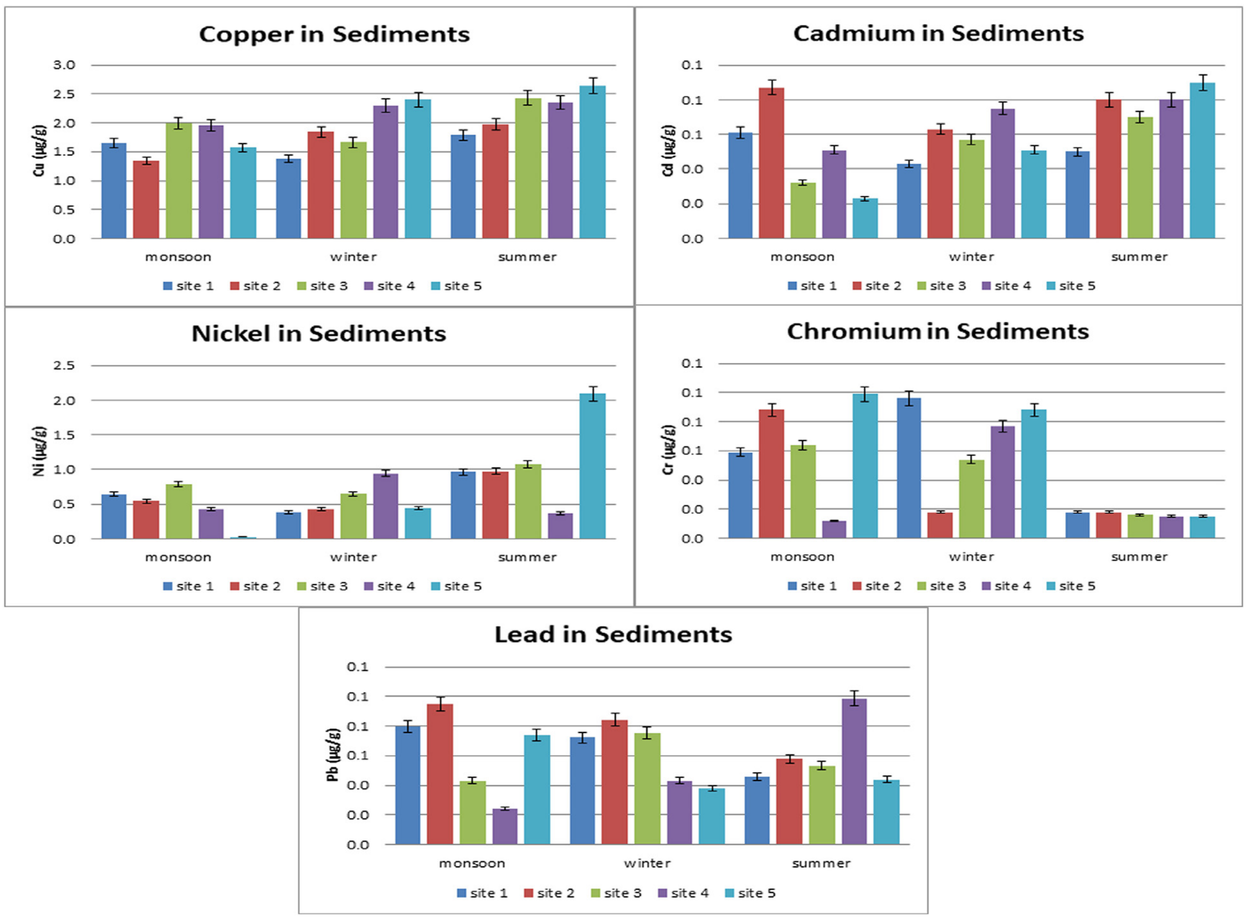


Figure 3 Changes in accumulation of heavy metals in leaves of *Avicennia marina* during different seasons.

3.2. Accumulation of heavy metals in leaves of *Avicennia marina*

From present studies, it was observed that the mean concentration of heavy metals in leaves varied in range from 0.049 to 0.067 µg/g for copper, 0.009 to 0.028 µg/g for nickle, 0.003 to 0.009 µg/g for cadmium, 0.021 to 0.052 µg/g for chromium and 0.001 to 0.0039 µg/g for lead. The concentrations of heavy metals in leaves were in following order Cu>Cr>Pd>Ni>Cd. There was no significant ($p>0.01$) difference in the accumulation of heavy metals in leaves of *A. marina* at all the five natural habitats. However, there was remarkable high influence of seasonal change on accumulation of heavy metals in leaves (Table 2). There was slightly higher accumulation of copper in leaves of *A. marina* during monsoon as compared to summer and winter (Figure 3). There was high accumulation of nickle in leaves of *A. marina* during summer season as compared to monsoon and winter (Figure 3). There was high accumulation of cadmium during winter and summer season as compared to monsoon (Figure 3). There was high accumulation of chromium during summer season as compared to winter and monsoon (Figure 3). There was high accumulation of lead during summer season as compared to monsoon and winter (Figure 3).

3.3. Biological concentration factors

The mean values of biological concentration factors obtained from leaves samples were in range 0.023—0.044 µg/g for copper, 0.01—0.39 µg/g nickel, 0.033—0.26 µg/g for cadmium, 0.0029—0.36 µg/g for chromium and 0.015—

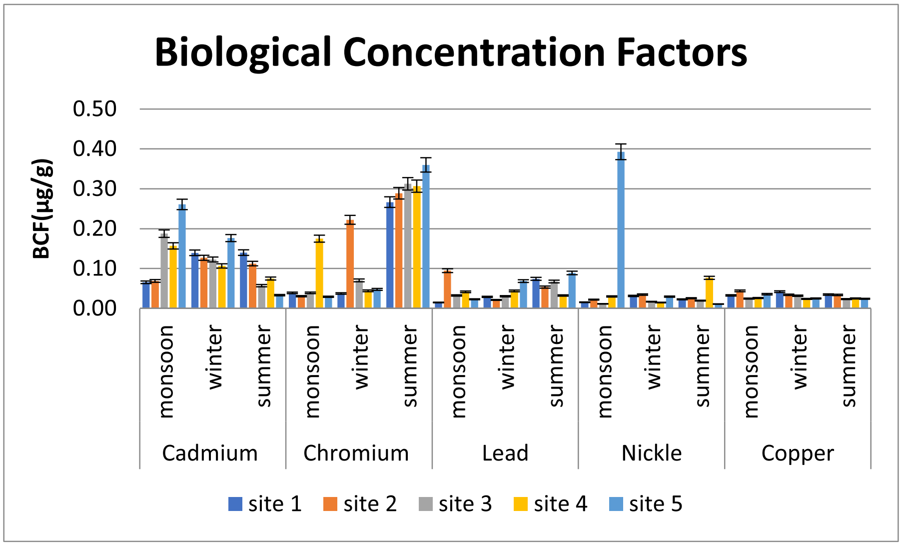
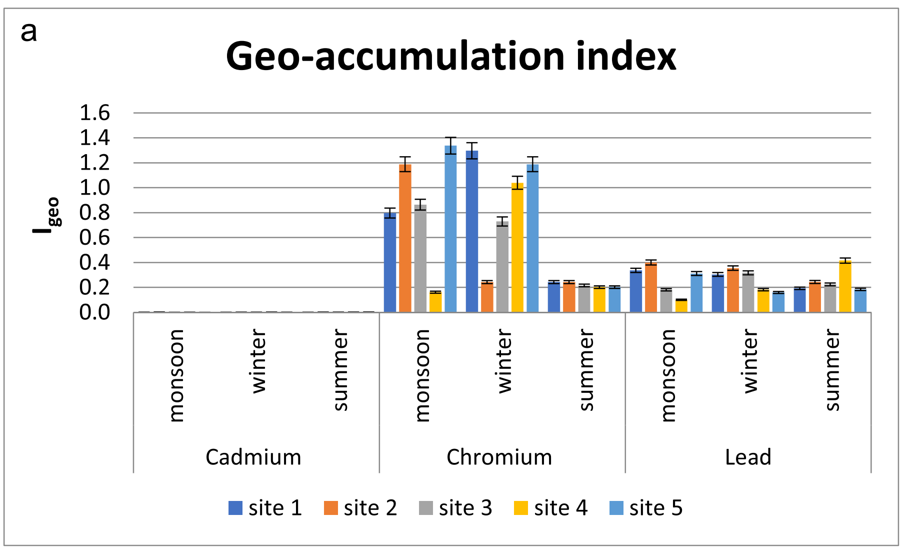


Figure 4 Biological concentration factors of heavy metals in leaves of *Avicennia marina* during different seasons.



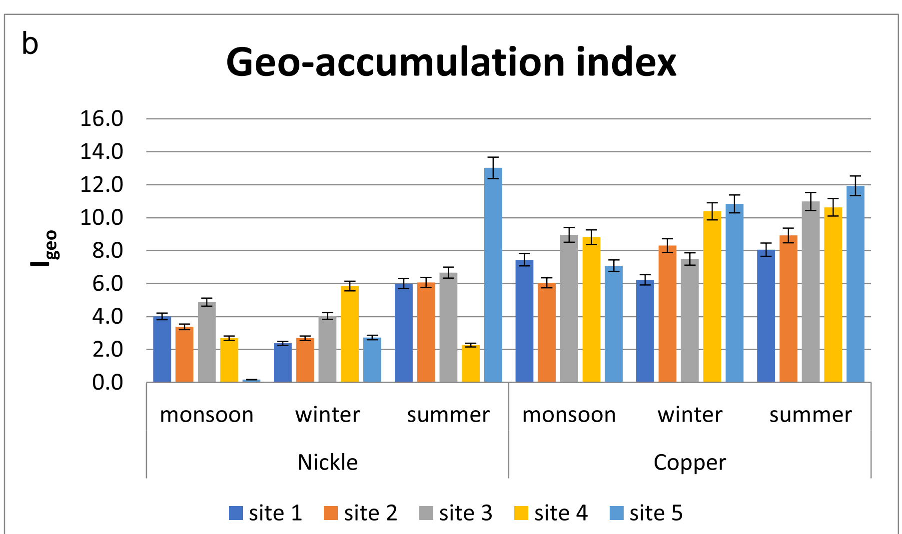


Figure 5 Geo-accumulation index of heavy metals (Cd, Cr, Pb) in sediments of *Avicennia marina* during different seasons (a). Geo-accumulation index of heavy metals (Ni, Cu) in sediments of *A. marina* during different seasons (b).

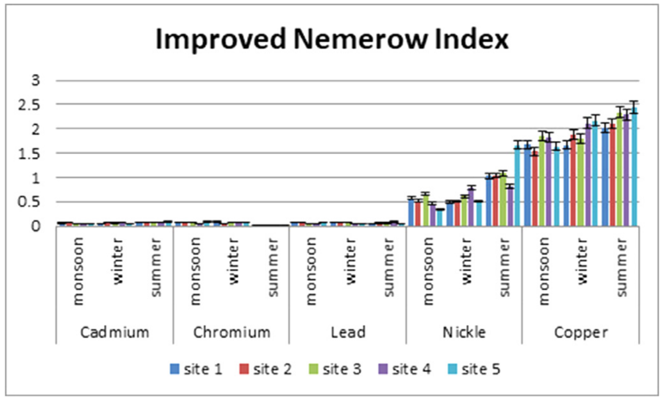


Figure 6 Improved Nemerow index of heavy metals in sediments of *Avicennia marina* during different seasons.

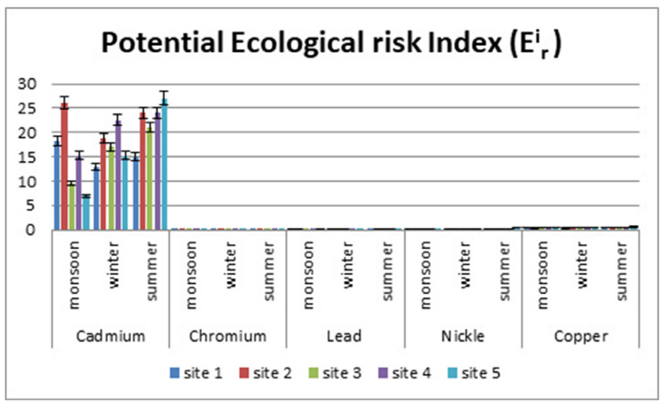


Figure 7 Potential Ecological Risk index (E^i_r) of heavy metals in sediments during different seasons.

0.094 μg/g for lead. The highest biological concentration factors among all the five habitats during three seasons were obtained for cadmium and chromium in leaves samples (Figure 4).

3.4. Geo-accumulation index and ecological risk coefficient

The geo-accumulation index (I_{geo}) suggested that cadmium and lead demonstrated uncontaminated sediments with I_{geo} greater than equal to 0 at all the five habitats during all seasons. The chromiun demonstrated moderately contaminated sediments with I_{geo} value greater than 1 at Site-2 and Site-5 during monsoon season and at Site-4 and Site-5 during winter season. However, during summer season chromium showed uncontaminated sediments with I_{geo} value greater than 0. The copper and nickel demonstrated moderately to heavily contaminated sediments with I_{geo} value greater than 2 in all the five habitats during all seasons (Figures 5a and 5b). The improved Nemerow index (Figure 6) and potential ecological risk index showed the same pattern as that of the geo-accumulation index. However, the copper, nickel, cadmium, chromium, and lead demonstrated uncontaminated sediments with E^i_r values less than 40 (Figure 7).

3.5. Multivariate cluster analysis

Multivariate cluster analysis was used to detect the similarity between the five habitats during three seasons. Cluster analysis grouped the five habitats in to two groups on the basis of its mean heavy metal concentrations in sediments during three seasons. Group A included Site-1, Site-2 and Site-3 and Group B included Site-4 and Site-5. Group B corresponds to high concentration of Cu, Ni, Cr, Pd and Cd during summer and winter season as compared to group A which suggests high pollution in group B as compared to group A. Similar clusters were also formed by non-metric multidimentional scaling (MDS) (Figure 8) which supported the results of multivariate Cluster analysis. From the Prin-

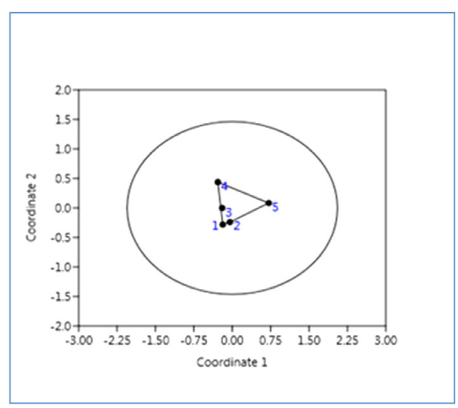


Figure 8 Non-metric multidimensional scaling of five natural habitats of *Avicennia marina* on the basis of heavy metals concentrations in sediments.

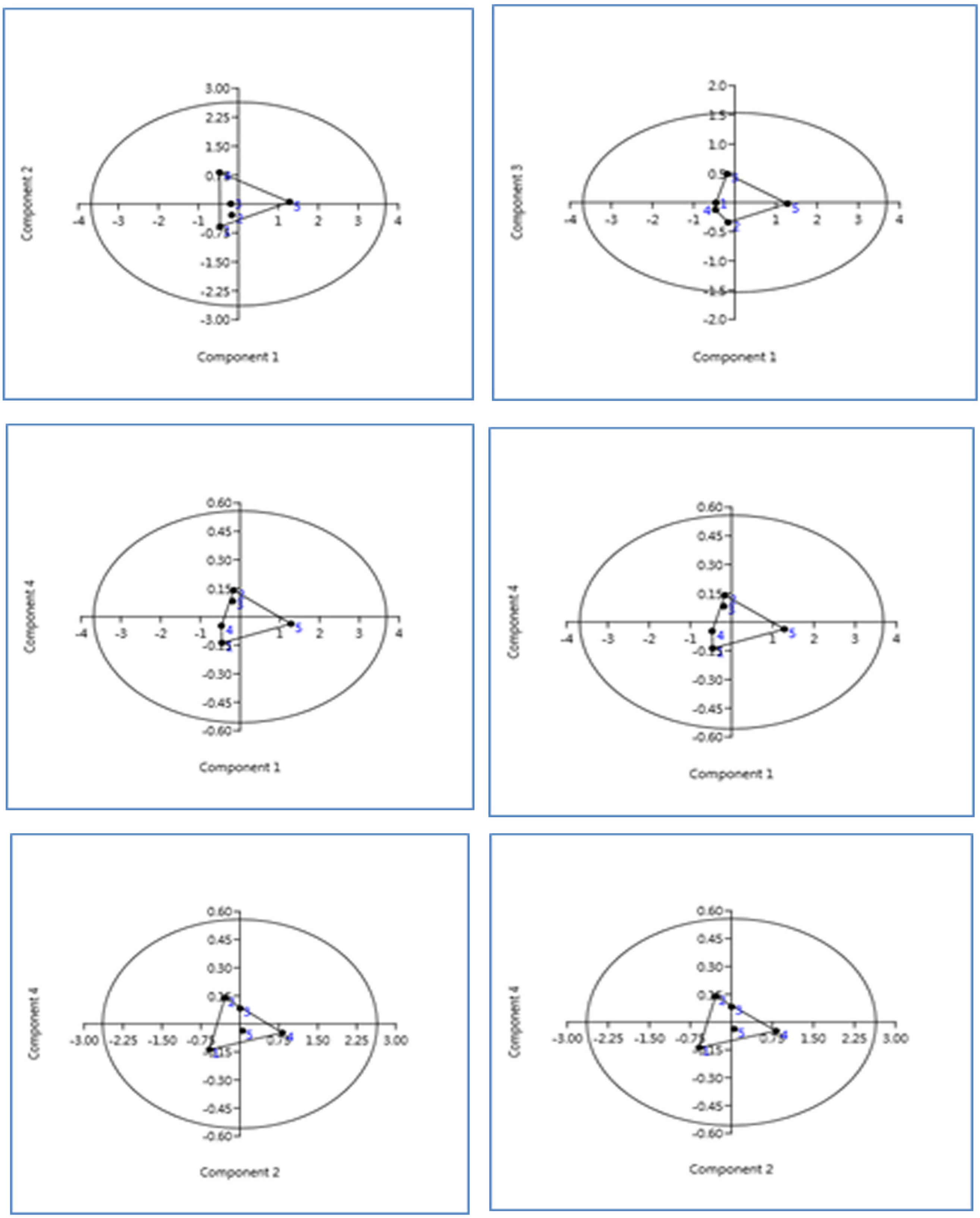


Figure 9 Principal component analysis of heavy metals in sediments of five natural habitats *Avicennia marina* during different seasons (summer, winter and monsoon).

ciple component analysis, it was observed that there was 99.9% total variations were retained on the basis of the eigenvalue. The first and second principle components explained 58.435% and 30.059% of variance in the heavy metals, respectively. However, third and fourth priniciple component explained only 10.167% and 1.338 % of the variance, respectively. This suggests that cluster B which includes Site-4 and Site-5 has more pollution of heavy metals than cluster A which includes Site-1, Site-2 and Site-3 (Figure 9).

4. Discussion

In present studies, there was significant ($p<0.01$) influence of seasonal changes on concentration of heavy metals in sediments of all the five natural habitats of *A. marina* at Diu coast. The concentration of copper was high during summer season as compared to winter and monsoon at all the five habitats. The concentration of cadmium was high during summer and winter season as compared to monsoon. The concentration of chromium was high during monsoon and winter season as compared to summer. The concentration of lead was high during winter and monsoon season as compared to summer. It was assumed that the presence of higher concentration of copper in sediments is due to the presence of many fishing boats that use antifouling paints that contains $CuSO_4$ as a major ingredient which is being also suggested by Usman et al. (2013). The presence of other heavy metals in the natural habitats of mangroves is due to several anthropogenic activities such as fishing, sewage and industrial pollution which are the major causes for the accumulation of heavy metals in the sediments. Major industries present in the Diu region are polyester, paper, cotton yarn, petroleum by products, plasticizers, pharmaceuticals, electrical conductors, marble tiles, and plastics which are the key sources of heavy metal pollution in mangrove sediments. Previous studies have also recorded

Table 3 Comparison of total heavy metals [μg/g] in mangrove sediments at different sites along the coastal region of Diu and standard quality guidelines.

Heavy metals	Cu	Ni	Cd	Cr	Pd	
Threshold effect concentration (TEC) SQGs						
Threshold effect level (TEL)	16	16	0.596	26	31	
Lowest (2000) effect level (LEL)	35.7	18	0.6	37.3	35	MacDonald et al. (2000)
Effect range low (ERL)	70	30	5	80	35	
Canadian sediment quality guidelines (TEL)	18.7	—	0.7	—	30.2	Canadian Council of Ministers of Environment (2002)
Probable effect concentration (PEC) SQGs						
Probable effect level (PEL)	149	36	3.53	90	91.3	
Severe effect level (SEL)	110	75	10	110	250	
Toxic effect threshold (TET)	86	61	3	100	170	
Effective range median (ERM)	390	50	9	145	110	
Canadian sediment quality guidelines (PEL)	108	—	112	—	112	Canadian Council of Ministers of Environment (2002)
Present studies						
Monsoon Season						
Site-1	1.65	0.645	0.061	0.059	0.08	
Site-2	1.34	0.543	0.087	0.088	0.095	
Site-3	1.984	0.784	0.032	0.064	0.043	
Site-4	1.953	0.432	0.051	0.012	0.024	
Site-5	1.569	0.028	0.023	0.099	0.074	
Summer Season						
Site-1	1.785	0.965	0.05	0.018	0.0458	
Site-2	1.976	0.976	0.08	0.018	0.0579	
Site-3	2.432	1.072	0.07	0.016	0.0533	
Site-4	2.355	0.365	0.08	0.015	0.0986	
Site-5	2.643	2.093	0.09	0.015	0.0438	
Winter Season						
Site-1	1.38	0.382	0.043	0.096	0.0723	
Site-2	1.84	0.432	0.063	0.018	0.0843	
Site-3	1.66	0.649	0.057	0.054	0.0753	
Site-4	2.3	0.941	0.075	0.077	0.0432	
Site-5	2.4	0.439	0.051	0.088	0.0379	

high concentrations of heavy metals in mangrove sediments and have suggested that anthropogenic activities are the main source of heavy metal pollution in mangrove habitats (Defew et al., 2005; Tam and Wong, 2000).

The results obtained in the present studies were compared with the standard quality guidelines (SQGs) of heavy metals given by Bakan and Özkoc (2007), Luo et al. (2010) and MacDonald et al. (2000). The threshold effect concentrations of copper, nickel cadmium, chromium and lead were lower than the values of TEL, LEL, ERL, PEL, SEC TET and ERM (Tables 3). This suggest that the season changes has maximum influence on accumulation of heavy metals in sediments of mangroves habitat which can cause infrequent threat to the marine organisms (MacDonald et al., 2000).

In present studies, the highest biological concentration factors among all the five habitats during three seasons were obtained for cadmium and chromium in leaves samples. This clearly suggests the bioavailability of cadmium and chromium in sediments of all the five natural habitats of *A. marina*. The low values of biological concentration factors for copper, nickel, and lead suggests low bioavailability of these metals in the sediments. This reflects the chelation of heavy metals with organic molecules which results in formation of immovable compounds (Li et al., 2016; Nath et al., 2014).

The geo-accumulation index (I_{geo}) was introduced by Müller (1969) to assess metal pollution in sediments. It has been applied in present studies to facilitate the qualitative assessment of heavy metal contamination in sediments of five natural habitats (Shi et al. 2014; Srinivasa et al. 2010). The improved Nemerow index and potential ecological risk index showed the same pattern as that of the Geo-accumulation index. These results suggests that the sediments are influenced by anthropogenic sources such as industrial wastes, untreated sewage effluents and antifouling paints from fishing boats (Usman et al., 2013). The ecological risk index suggested that there is no significant effect of heavy metals on growth of plants in the mangrove ecosystem. Multivariate cluster analysis, non-multidimentinal scaling and Principal component analysis revealed that the samples collected from the natural habitats (Site-4 and Site-5) near the fishing and industrial areas were the main sources of heavy metal contamination.

5. Conclusions

From the present studies, it was concluded that the levels of heavy metals were lower than the toxic effect threshold level and probable effect level which indicates a very limited biological impact on the marine environment. The higher concentrations of heavy metals in the leaves samples of mangroves indicated that *A. marina* accumulates heavy metals. The values of biological concentration factors (BCFs) suggested that the bioavailability of cadmium, chromium and lead were high as compared to copper and nickel. According to I_{geo} index, Site-4 and Site-5 were heavily contaminated with copper and nickel. However, Site-1, Site-2 and Site-3 were moderately affected by heavy metals. The ecological risk index suggested that there is no significant effect of heavy metals on growth of plants in the mangrove ecosystem. Principal component analysis revealed that the samples collected from the natural habitats (Site-4 and Site-5) near the fishing and industrial areas were the main sources of heavy metal contamination.

Conflict of interest declaration

Authors declare that there is no conflict of interest regarding publication of present research work in journal.

Acknowledgements

Authors are highly thankful to Department of Biosciences, Saurashtra University, Rajkot for providing necessary facilities to carry out this research work.

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