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Chapter

Concentrations of Heavy Metals as Proxies of Marine Pollution along Nellore Coast of South District, Andhra Pradesh

Madri Pramod Kumar, Tella Lakshmi Prasad, Kothapalli Nagalakshmi, Nadimikeri Jayaraju and Ballari Lakshmanna

Abstract

Bottom sediment samples from six stations were sampled in pre monsoon 2016, from the Govindampalli – Durgarajupatnam (GP-DP) coast. Heavy metals viz., Fe, Mn, Cr, Cu, Ni, Pb, Zn and Cd analysis was carried out by using ICP-OES, and the average concentrations are as follows Fe > Mn > Zn > Cr > Pb > Ni > Cu > Cd. Various environmental indices like Factor Analysis (FA), Geo-accumulation Index (Igeo), Enrichment Factor (EF) and Pollution Load Index (PLI) were applied to the chemical data in order to know the levels of contamination and factors contributing to the pollution. Correlation coefficient results exhibits significant positive and negative relationships among Fe, Mn, Pb, Zn, Cd. All the environmental indices suggest that heavy metals were present at higher concentrations and the impacts of anthropogenic activities are crucial that serves as source of heavy metals in the zone. Relatively, maximum number of heavy metals viz., Fe, Ni and Pb were accumulated at the brackish environment i.e., at confluence of Swarnamukhi river (GP-S Station).

Keywords: GP-DP coast, heavy metal analysis, factor analysis, geo-accumulation index, enrichment factor, pollution load index

1. Introduction

Across, the world coastal zone provides sea food to more than two billion peoples living in these areas. Apart from this, these also serve as major economic resources either directly or indirectly. From the last couple of decades, marine ecosystems have been experiencing severe and acute environmental stress, due to rapid diversifying human activities like navigation, exploration programmes for economic minerals and ores, exploration of hydrocarbons, fishing, establishment of harbors, ports and oil spills etc., In addition, to the anthropogenic activities on the terrestrial environment, natural processes like volcanism, erosion and weathering etc., also contribute to the enhancement of undesirable and unwanted chemical elements which are called pollutants in the marine environments which ultimately pose major menace to the fragile ecological system.
Among wide range of pollutants, heavy metals are the chemical elements which are generally found in low concentrations in marine and coastal environs. However, anthropogenic activities have inevitably enhanced the heavy metal concentrations which impacts not only marine ecosystem but also humans through consumption of polluted sea food [1, 2]. Heavy metals, because of their persistence and toxic nature in the natural environments, have been receiving utmost attention from the scientific community from the last couple of decades [2–14]. Several scientific studies have obtained that, once these heavy metals introduced in the marine and coastal environments by various sources, they will be redistributed both in water and sediments. Hence, it is important to determine the heavy metal concentrations in the marine ecosystem to evaluate the pollution levels in the sensitive and most fragile marine environs.

The present investigation deals with the evaluating of heavy metal concentrations from the bottom sediments in the study area i.e., Govindampalli – Durgarajapatnam coast (GP-DP coast). Sand, silt, clay and other materials which settle down at the bottom of the water column form the sediment. Various sources like decomposition of animals, plants, erosion, weathering processes on bed rock and soils give rise to form sediment as well. It is observed from the studies of multielemental analysis that, apart from being habitat, sediment provides nutrients for several marine and aquatic biota. Furthermore, these sediments also play a vital role in the absorption of heavy metals in marine environments [9, 14–19]. Thus, it is imperative to determine the levels of heavy metal contaminations, their enrichment levels both in the surface and bottom sediments. Sequentially this will help to acquaint their wide spectrum of implications on the natural ecological systems.

2. Study area

The area under investigation i.e., Govindampalli to Durgarajapatnam coast (GP-DP coast) forms as a part of Nellore district (south to the Krishnapatnam port), Andhra Pradesh, South East Coast of India that lies between 14°01’10” - 14°02’30” N latitudes and 80°08’20” - 80°19’00” E longitudes. It falls in toposheet No.66 B3, 66 B4 & 66 C1& C5 on scale 1:50,000 of Survey of the India (SOI). The GP-DP coast is dissected by two tidal creeks and Swarnamukhi river as well (Figure 1) One creek exits

Figure 1. Location Map of the Study area.
between Tuplipalem north (TP-N) and south (TP-S) beaches and another between Durgarajupatnam north (DP-N) and south (DP-S) beaches. The Swarnamukhi River estuary is situated in between Govindampalli north (GP-N) and south (GP-S) beaches. The total stretch of the GP-DP coast is about 7 km. The Pulicat Lake is located 120 km away towards south from the study area. Buckingham canal is present in western side of the Pulicat Lake. The Swarnamukhi River is an independent river with no tributary and serves as major source for sediment supply in this region.

2.1 Geology and geomorphology

The geology of hinter land consists of Quaternary alluvium, laterites and Triassic sandstones. The Precambrian Basement gneisses, amphibolites and
migmatised pelitic schists are observed. Above these rocks recent alluvial sediments are deposited [14]. The coastal stretch, for some extent is rigorous and laterally compacted by sand. The total area is noticeable with different geomorphological features like asymmetrical sandbars/sand dunes, salt affected lands, mud flats, mangroves, aquaculture ponds etc., (Figure 2). The coastal fringe shows dendritic to subdendritic drainage patterns.

3. Sample collection

For evaluating the heavy metal concentrations, undisturbed bottom sediment samples were collected by using Polymerization of Vinyl Chloride (PVC) pipe from six sampling stations i.e., GP-N, GP-S, TP-N, TP-S, DP-N, and DP-S, in pre monsoon season (2016). The sampling station coordinates were recorded by using hand held Global Positioning System (GPS). The collected sediment samples were placed in polyethylene bags having zip lockers. These were pre-cleaned with double distilled water and index accordingly. Care has been taken that there was no loss or damage to polyethylene bags during transportation from field to the laboratory. Later, each sediment sample was dried at 110⁰C, by using pistle and mortar, the sediment samples were grounded in order to pass through 200 μ sieve. The sieved material comprises of silt and clay which may adsorb heavy metals at higher levels. About 1gm of sieved material was digested with aquaregia (HNO₃: HCl) and filtered through 0.45 μ membrane and the concentrations of heavy metal [Iron (Fe), Chromium (Cr), Manganese (Mn), Nickel (Ni), Copper (Cu), Zinc (Zn), Lead (Pb) and Cadmium (Cd)] were generated by Inductively Coupled Plasma – Optical Emission Spectroscopy (ICP-OES) [20–25]. The data sets thus obtained were subjected to statistical analysis.

4. Statistical analysis (SA)

Various environmental indices like Factor Analysis (FA), Geo-accumulation Index (Igeo), Enrichment Factor (EF) and Pollution Load Index (PLI) were applied to the chemical data in XL-STAT (2013) and SPSS softwares, in order to know the levels of contamination and factors contributing to the pollution. Further it is also used to unearth the levels of their (heavy metals) enrichment with respect to the natural environment.

4.1 Factor analysis (FA)

Factor analysis is very handy if the data generated, constitutes a large amount of variables. Thus, it believes redundancy among some variables, which means certain variables are interconnected with one another because of the same construct [26]. It basically provides information regarding the source of pollution and its (metals) behavior in the form of factors besides giving a glance on the controlling covariance structure among charted variables. In XL-STAT 2013 software, the Carl’s Pearson coefficient matrix is transformed to diagonal matrix to attain Eigen values by using Kaiser standardization. The largest Eigen’s value attributes to be Factor 1 explicates the larger variance among the datasets and the Factor 2 formulates most of the variance in general.

4.2 Geo accumulation index (Igeo)

To evaluate and to quantify the heavy metal pollution in the bottom sediments Geo accumulation Index (Igeo) was used. This was introduced by Muller [27] using the following
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\[ I_{geo} = \log_2 \left( \frac{C_n}{1.5B_n} \right) \]  

(1)

Where, 1.5 is a constant which intends to the potential variation of the lithogenic effects if any [28] and \( B_n \) is the geochemical background value of the respective metal (\( n \)) and \( C_n \) is the (\( n \)) concentration of the element in the collected sample. Nevertheless, the background values for the Indian sediment are not available, so, the average upper continental values were used [29, 30]. To designate the sediment quality, Muller classified the Igeo values into seven categories: extremely contaminated (>5), strong to extremely contaminated (4-5), strongly contaminated (3-4), moderately to strongly contaminated (2-3), moderately contaminated (1-2), uncontaminated to moderately contaminated (0-1) and uncontaminated (<0).

4.3 Enrichment factor (EF)

In general, EF is applied to realize the degree of sediment contamination by heavy metals, other than lithogenic sources. To ascertain EF, earlier workers employed the average upper continental values of Fe and Al to normalize the determined heavy metal concentrations with respect to a background metal value [14, 31–35]. In this study, Fe is used as a conservative element to differentiate the sources of anthropogenic and natural components. Furthermore, the Fe element play a vital role in redox reaction. In the reduction phase Fe acts as sink to the heavy metals and in oxidation phase Fe has significant control on the distribution of heavy metals among the sediments [16, 36]. Owing to these facts, Fe is used as conservation element. For EF calculation the following equation was used

\[ EF = \frac{M_x \times Fe_x}{M_b \times Fe_b} \]  

(2)

Where, \( Fe_x \) is the Fe concentration in the sediments, \( M_b \) and \( F_b \) are their concentration in a suitable baseline reference material and \( M_x \) is the sediment sample concentration of heavy metals. Brich [37], classified the EF values as follow: if the EF values show >40 the sediment or soils falls in extremely high enrichment category, if EF values in range 20-40 signifies very high enrichment category, if the EF values are in range of 5-20 the sediment or soil falls in significant enrichment category, if the values are in between 2 and 5 signifies moderate enrichment and if EF values is 2 then the sediment or soil is considered as deficiency to minimal enrichment category.

4.4 Pollution load index (PLI)

The Pollution Load Index is evaluated for a zone as well as for a particular station and calculated according to Tomlinson [38]. The PLI for a particular station and for zone can be determined by the following formula

\[ PLI \text{ for a Station} = \sqrt{CF_1 \times CF_2 \ldots \times CF_n} \]  

(3)

Where, \( CF = \frac{C_{metal}}{C_{background}} \) (\( C_{metal} \) is the respective metal concentration of the sample and \( C_{background} \) Metal concentration of the background) and \( n \) is the number of metals and contamination factors.

\[ PLI \text{ for zone} = \sqrt{Staion1 \times Station2 \ldots \times Station_n} \]  

(4)
5. Results and discussion

5.1 Heavy metal concentrations

The results of the heavy metal concentrations from the bottom sediments samples of GP – DP coast were obtained by using ICP – OES (Table 1). The range and average concentrations (in parenthesis) in ppm were, Fe 2241.32-3715.15 (2844.79), Cr 9.14 – 11.12 (10.21), Mn 27.19-99.1 (62.02), Ni 5.2-6.67 (6.0), Cu 2.12-7.12 (4.24), Zn 12.65-19.26 (15.0), Pb 5.67-9.14 (7.0), and Cd 0.39-1.15 (0.74). The average concentration of the heavy metals arranged in decreasing as follows: Fe > Mn > zn > Cr > Pb > Ni > Cu > Cd. The concentrations of Fe, Ni, and Pb found to be higher levels at GP-S station (Table 1). The highest concentration of these metals is attributed mainly due to the nearness of GP-S station to the confluence of the Swarnamukhi river (the whole sediment load carried by the river drains into the Bay of Bengal). In addition, intense oblique onshore currents coupled with SW winds aid in the accumulation of heavy metals at the GP-S station [39].

The anomalous concentration of Fe metal in the whole study area is may be due to the anthropogenic activities at the Krishnapatnam port, which is present at the northern side of the investigation area. The Zn and Cd metals were found at higher levels at DP_N station. The presence of Zn at higher concentration in the sediment samples may be attributed to the basement rocks underlain by the Quaternary alluvium, amphibolites, migmatised politic schists and Triassic sandstones [14, 40, 41]. The Cd at DP-N station is contributed by the wide range of anthropogenic activities viz., ignition or burning of urban and domestic wastes at the neighborhood area of the investigation area and combustion of fossil fuels for navigation purposes [42]. Heavy metals like Cu, Cr and Mn were found in higher concentrations at stations GP – N, DP – S and TP – N respectively, contributed by irregular and untreated dissemination of industrial waste in to the drainage system.

To know the degree of linear affiliation among heavy metals, correlation coefficient was carried out (Table 2). Moreover these results help in understanding how closely two variables (metals) move in relation to one another. Positive correlation observed among Fe, Mn, Pb Zn and Cd, which signifies commonality in mutual dependence and identical behavior during transportation. Significant negative correlations were noticed between Fe – Cu, Cr-Cd, Cu-Pb, and Cu-Zn.

The obtained results were compared with the results of Sreenivasulu [14] (Table 3) who studied the geochemistry of bottom sediments in pre-monsoon seasons 2015. By comparison, it is evident in this study that the concentrations of

| Sample ID | Fe   | Cr   | Mn   | Ni   | Cu   | Zn   | Pb   | Cd   |
|-----------|------|------|------|------|------|------|------|------|
| GP-N      | 3462 | 10   | 99   | 5    | 5    | 13   | 7    | 1    |
| GP-S      | 3715 | 11   | 93   | 7    | 2    | 16   | 9    | 1    |
| TP-N      | 2241 | 9    | 49   | 7    | 7    | 13   | 6    | 1    |
| TP-S      | 2312 | 11   | 27   | 6    | 4    | 14   | 6    | 0    |
| DP-N      | 3215 | 9    | 61   | 6    | 2    | 19   | 7    | 1    |
| DP-S      | 2726 | 11   | 41   | 6    | 4    | 13   | 6    | 1    |
| Min       | 2241 | 9    | 27   | 5    | 2    | 13   | 6    | 0    |
| max       | 3715 | 11   | 99   | 7    | 7    | 19   | 9    | 1    |
| avg       | 2845 | 10   | 62   | 6    | 4    | 15   | 7    | 1    |

Table 1. Heavy metal concentrations (ppm) from the study area (Pre-monsoon) 2016.
5.2 Hierarchical cluster analysis (HCA)

To examine the possible sources and associated heavy metals contamination, hierarchical cluster analysis was performed by using XLSTAT software. The obtained result (dendrogram) was shown (Figure 3). The degree of association among heavy metals represents by the distance cluster. Low distance cluster suggests a significant relationship and long distance cluster insignificant relationship among heavy metals. Two clusters were formed i.e., TP- S to DP-S and DP –N to GP –S. Cluster 1 is formed by stations TP –S, TP – N and DP – S and cluster 2 is formed by DP-N, GP–N and GP-S. Cluster1 and 2 showed high and moderate concentration of heavy metals respectively. The high and moderate contaminations are attributed to the sediment load (which was brought down by the Suwarnamukhi River and the creeks) and proximity of these sampling stations to the inlets.

5.3 Factor analysis (FA)

Factor analysis is carried out to the heavy metal data to ascertain potential sources of contamination. A total of three factor loadings emerged with total variance of 81.72% (Table 4), of which factor 1 accounts 42.46%, factor 2, 25.57% and factor 3,
13.68% variance (Figures 4–6). Factor 1 was represented by Fe (0.926), Pb (0.91) and Mn (0.738). Therefore it may be termed as Fe-Pb-Mn assemblage. Cr (0.956) and Mn (0.663) show a significant contribution to the factor loading of 2 and 3 respectively signifies Factors controlling the sediment contamination in the coastal zone.

5.4 Geo accumulation index (Igeo)

The Geo accumulation Index is performed to know the existing environmental conditions. It also aids to determine the amount of heavy metal contamination in the natural environments. The results of the Geo accumulation Index (Igeo) which were calculated by using Eq. (1) were presented (Table 5 and Figure 7).

The range of the Igeo values of the heavy metals in the study area is as follows: Fe (26-27), Mn (14-16), Cr (9), Cu (5-7), Ni (8), Pb (6-7), Zn (10) and Cd (−4 to −2). This reveals that the GP-DP coast has been extremely polluted by Fe, Mn, Cr, Cu, Ni, Pb, Zn metals (Table 5). Applying the Muller’s classification, the results of the

![Dendrogram of grouping of sediment sampling stations.](image)

### Table 4. Factor analysis results of heavy metals.

| Parameters | F1    | F2    | F3    |
|------------|-------|-------|-------|
| Fe         | 0.93  | 0.15  | 0.35  |
| Mn         | 0.74  | 0.06  | 0.66  |
| Cr         | 0.20  | 0.96  | -0.13 |
| Cu         | -0.86 | -0.11 | 0.38  |
| Ni         | 0.18  | -0.20 | -0.33 |
| Pb         | 0.91  | 0.11  | -0.15 |
| Zn         | 0.65  | -0.58 | -0.48 |
| Cd         | 0.38  | -0.88 | 0.30  |
| Total      | 2.94  | -0.48 | 0.60  |
| % of variance | 42.46 | 25.57 | 13.68 |
| cumulative % | 42.46 | 68.04 | 81.72 |

**Bold values are show higher factor score loading/matrix. This indicates the higher pollution levels of respective elements.**
Igeo falls under extremely contaminated environment (>5). The GP-DP coast was unpolluted by the Cd heavy metal (Igeo results of Cd shows negative values).

5.5 Enrichment factor (EF)

In general, Enrichment Factor (EF) is deployed to understand the contribution of metals other than lithogenic origin. In the present study EF was calculated by using Eq. (2) and categorized according to Sutherland's classification [43], which states that the EF value less than or equal to 2 for metals signifies that the elements are from natural weathering processes and for the metals whose EF values are greater than 2. Those elements came from contaminated natural processes [44].
The calculated results of the EF for the sediment samples present study are given (Table 6 and Figure 8).

EF values (ppm) ranges from 0.6 – 1.6 for Mn, 1.5-2.4 for Cr, 1-5.3 for Cu, 1 – 2 for Ni, 4.4 – 6.4 for Pb, 1.8 - 3 for Pb, and 23.5 – 63.2 for Cd. Average values of EF are in the order of Cd > Pb > Cu > Zn > Cr > Ni > Mn > Fe. It is evident that Fe, Mn and Ni values are <=2 which fall under deficiency to minimal enrichment category (Table 6). In 67% stations Cd exists falls under very high enrichment category and 33% of the stations showing extremely high enrichment. The Zn metal shows moderate enrichment in all stations except GP -N which exhibits deficiency to minimal enrichment. About 67% of the stations significant enrichment of Pb and the remaining exhibit moderate enrichment. About 50% of the stations fall under moderate enrichment and 17% of the stations come under significant enrichment category for Cu. Stations viz., TP–N, TP-S and DP–S exhibits moderate enrichment to Cr metal. The anomalous behavior of Cd metal enrichment in the sediment samples reflect anthropogenic activities like, possible burning of fossils fuels, incineration of domestic and urban wastes in the vicinity of the study area.

### 5.6 Pollution Load Index (PLI)

Pollution Load Index (PLI) is an empirical and quick tool proposed by Tomlinson [38] to assess pollution severity and variations along a particulate site as
well as along different sites, besides providing a comparative study based on temporal basis. The Eq. (3 & (3.1)) were used to determine the PLI for a particular station and for specific zone respectively, which in turn aids to decide the sampling sites contains pollution or not. According to Tomlinson classification, if the PLI
value <1 indicates no pollution and if the PLI values show >1 meaning the zone/stations, is/are polluted. The obtained results for zone and for sampling stations were shown (Tables 7 and 8).

The value of the PLI of the whole study area ranges from 0.25 – 1.52 (Table 7). Except Cd, remaining metals i.e., Fe, Mn, Cr, Cu, Ni, Pb, and Zn were recorded below the baseline indicating no pollution when compared to the worldwide sediment. The unusual concentration (1.52) of Cd metal is attributed to direct external sources like, burning of fossil fuels, industrial activities, contaminated agricultural soils, mining waste, municipal sewage effluents and the erosion of hydrothermal mineralized rocks and the black shale deposit [45]. Comparing PLI values of one sampling station with the other ranges from 0.26 to 0.32 (Table 8). According to Tomlinson [38], all the sampling stations were showing less than one, signifying that all the heavy metals at all stations were within threshold values. The variations in the indices are an outcome of the difference in sensitivity of these indices towards the sediment pollutants [14, 46].

6. Conclusion

The present paper deals with the assessment of pollution status by determining the heavy metals concentrations in the bottom sediment samples. By and large heavy metals are the chemical elements which occur in low concentrations in fragile coastal and marine environments. But, subsequent anthropogenic activities have inevitably enhanced concentrations in the marine ecosystem. Thus, in the present study (pre-monsoon 2016) Heavy metals (Fe, Mn, Cr, Cu, Ni, Pb, Zn and Cd) analysis was carried out by using ICP-OES, from the bottom sediments along GP-DP coast. Results show that the average concentrations are as follows Fe > Mn > Zn > Cr > Pb > Ni > Cu > Cd. Fe and Cd were recorded highest and lowest concentrations at all stations respectively in the study area.

Relatively, maximum number of heavy metals viz., Fe, Ni and Pb were accumulated at the brackish environment i.e., at confluence of Swarnamukhi river (GP-S Station) and it is also ascribed to the North-Easterly winds (since the sediment samples were collected in the southwest monsoon season). Multivariate analysis like correlation coefficient and factor analysis were carried out to understand complex dynamics of the pollutants. Correlation coefficient results exhibits significant positive and negative relationships among Fe, Mn, Pb, Zn, Cd and Fe – Cu, Cr – Cd, Cu – Pb and Cu – Zn respectively. A total of three Factors with 81.72% variance explain the controlling elements of sediment contamination. Factor 1 includes Fe- Pb – Mn assemblage, Factor 2 and 3 include Cr and Mn.
To assess the pollution in bottom sediments, Geo-accumulation Index (Igeo), Enrichment Factor, and Pollution Load Index were used. The results of Geo-accumulation Index denote that the GD-DP stretch was extremely polluted with Fe, Cr, Cu, Ni, Mn, Zn and Pb metals. Computation of Enrichment Factor and categorizing the obtained results according to Sutherland’s Classification, Cd metal showed very high enrichment in the 67% of the stations and extremely enriched in 33% of the stations. The Zn-Cr and Pb metals showed moderate and significant enrichment respectively. The Cu exhibits 50% and 17% of the stations falls under moderate and significant enrichment categories respectively. The variations in the indices are an outcome of the difference in sensitivity of these indices towards the sediment pollutants.

Pollution Load Index values (PLI) of Cd metal showed higher value 1.52. This may be due to the influence of direct extraneous sources like burning of fossil fuels for navigation, industrial activities, contaminated agricultural soils etc.,. The obtained results from the present study were compared to the results of Sreenivasulu who has studied the geochemistry of bottom sediments for the two seasons. It is evident that the concentrations of heavy metals viz., Fe, Cr, Cu were increased at all stations in the period of study and the remaining metals showed decreasing trend.

According to the current study, the heavy metals were extremely harmful to marine life. They were present at various higher concentrations and the impact of anthropogenic activities are crucial that serves as source of heavy metals in the zone. Therefore, in order to prevent severe heavy metal contamination in the investigation area, it is mandatory to enforce monitoring, mitigating and remedial strategies to reduce the loadings and cumulative concentrations of heavy metals in the sediments along fragile coastal ecosystem for sustainable development of future generations. Further care has to be taken to educate and inform relevant stakeholders to avoid converting the coastal zones into sink/dustbins in future.

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