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Spatial Distribution of Heavy Metals in Surface Water from Coastal Areas of Ernakulam District, Kerala, South West Coast of India

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ABSTRACT

The concentration of heavy metals and their spatial distribution in surface waters collected from coastal areas of Ernakulam District, Kerala, India were analyzed for this study. The heavy metals concentrations (Zn, Ni, Cd, Cu, and Pb) of samples collected from 8 typical sites were determined by Atomic Absorption Spectroscopy. Multivariate statistical analysis method such as Spearman's correlation coefficient, PCA, and cluster analysis are the methods used for the identification of the heavy metal sources and its relationship between pollutants in surface water. The complete analysis revealed the main source of heavy metal contamination was due Ni and Cd moderate, very low level by Pb, Zn and Cu. The principal component analysis and the correlation analysis showed a positive loading for Ni because of its higher level of pollution. The input for the heavy metal contamination was mainly the anthropogenic inputs such as municipal waste water, sewage discharge, aquaculture waste and fishing activities. The findings of this study revealed the water body is facing the risk of anthropogenic pollution due to heavy metals thereby increasing the toxicity in the coastal environment and affecting the ecosystem.

Key word : Heavy metals, Spatial distribution, Pollution level, Coastal waters

Introduction

Coastal ecosystem is affected by the presence of toxic metal pollutants introduced through various natural and anthropogenic sources. The natural sources including the weathering processes and anthropogenic ecosystems are vulnerable to heavy metal pollution, which are introduced by sources including sewage disposal, agricultural and aquacultural activities and many other human activities which facilitate the disposal of several chemical agents (Balachandran *et al.*, 2002). The heavy metals

cause continuously degradation in the water quality causing serious health hazards to the aquatic organisms (Naqvi *et al.*, 2000) accumulated in the rivers and then deposited in the marine sediments as a sink which forms a major issue related with the persistence of heavy metal which increases toxicity, bioaccumulation and biomagnifications, leading to the adverse effect on the ecosystem, human health and other living organisms (Demirak *et al.*; 2006). Heavy metal contamination in aquatic systems is a matter of serious concern from human health point of view as many of the organisms particularly fish

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which forms an integral part of human diet. Direct hazardous effects or potential risks to human health and ecosystem stability can be caused by heavy metal residues through multi-exposure pathways owing to their transport and transformation among multi-media such as ambient air, soil, surface water, sediments etc. (Cooke *et al.*, 1990). Heavy metals have low solubility and it gets absorbed and accumulated on bottom sediments. Bottom lake sediments are sensitive indicators for monitoring pollutants as they act as a sink and a carrier for contaminations in aquatic environment (Bai *et al.*, 2011). The sediments in the aquatic environment have been widely used as environmental indicators for the assessment of metal pollution in the natural water (Islam *et al.*, 2015). During transportation of heavy metals in the riverine system, it may undergo frequent changes due to dissolution, precipitation and sorption phenomena (Abdel-Ghani *et al.*, 2007). The spatial distribution of the heavy metals in the surface water and assessment of the risks caused by them helps in identification of mechanism of accumulation and transportation of pollutants in aquatic environment. Therefore a better understanding of the status of heavy metal pollution in the water body is inevitable for a sustainable development of the coastal marine ecosystem (Rainbow and Lueoma, 2011)

The coastal area of Kerala which is unique with the presence of large number of perennial and temporary backwaters is endowed with biological and genetic diversity (Thompson, 2001). Some reports have been published on the heavy metal levels in surface water (Saraladevi *et al.*, 1979) trace metal levels in sediment (Venugopal, 1982), chemometric study of numerous ecological studies in the Cochin backwaters, south west coast of India (Gopinathan *et al.*, 1984; Nair *et al.*, 1988). In this study we are reporting the spatial distribution, identification of source and the potential risks of the heavy metals in the surface water from the coastal areas of Ernakulam district, covering the Cochin backwaters.

Cochin backwaters are having a large surface and is shallow in depth (1.5-6m), where the metal speciation and fate in the backwaters is influenced by environmental factors such as inflow of riverine inputs of metals, introduction of industrial effluents and sewage, modification due to anthropogenic activities and hydrographic changes related to complexity of water use (Shibu *et al.*, 1990). Very high levels of Cu, Zn, Pb and Cd in the surface water bodies of

Cochin estuary indicates that in the estuarine environment, suspended particles plays a key role by acting as a source and sink for heavy metals (Rajamani, 1994). It was also reported by Kaladharan *et al.* (2011) that, in Cochin estuary as well as in Cochin inshore waters, cadmium level have reached critical point, while copper and lead is at a level of caution. Temporal trends of heavy metal in Cochin backwater indicated higher concentration of almost all trace metals during pre-monsoon (February to May) and post-monsoon (September to January) (Anju *et al.*, 2011).

The objectives of this study were (1) measure the concentration of heavy metals (Zn, Cu, Ni, Pb, and Cd) (2) to determine the spatial distribution of heavy metals in surface waters from the backwaters of Cochin estuary based on the geo-statistics method of inverse distance weighted interpolation (3) identify the relationship between the contaminants in surface waters and their possible sources in the study area using Spearman's correlation coefficient, principal component analysis (PCA) and cluster analysis.

Materials and Methods

Study Area

The 'Coastal Plains', in Ernakulam district covers an area of 1726 sq. km. has elevation less than 6 m above Mean Sea Level (MSL), parallel to the coast. The width of the coastal plains generally ranges from 10 km – 15 km. Coastal alluvial soils as well as laterites cover the area parallel to the coast dominated by the presence of number of backwater channels. This backwater channels known as Cochin backwaters, forms a part of Vembanad- Kole wetlands which is a Ramsar site. The Cochin backwater (Lat.09°30'-10°10'N and Long.76° 15'-76° 25'E) is topographically divisible into two arms; a southern one extending south of bar mouth (Fig.1) from Cochin to Thanneermukkam and a northern one extending north from Cochin to Azhikode (Ramamirtham *et al.*, 1986). This backwater is characterized by high productivity by acting as a nursery ground for many species of marine and estuarine fin fishes, molluscs and crustaceans. The low lying swamps and tidal creeks dominated by sparse patches of mangroves give shelter to juveniles of many important species. Mangroves provide food and shelter for the benthic organisms and are in-

involved in recycling of nutrients. The area is characterized by fine sediments which is rich in organic matter supporting abundant and benthic fauna (Menon *et al.*, 2000). This ecosystem is under the influence of monsoon, contributing 71% of annual rainfall (Jayaprakash, 2002) with three seasonal conditions prevailing in the area namely monsoon (June–September), post-monsoon (October–January) and pre-monsoon (February– May). The land use and land cover are classified as settlement (75%) cultivable and marsh lands (12%) and water bodies (13%) in the study area. (Fig.1).

Samples collection and preparation

In total eight surface water samples were collected from 8 typical locations (Fig. 1) which were chosen as five from northern side and 3 from the southern side of the Cochin bar mouth around the zone of research area of Cochin backwaters during April 2019. The monitoring sites were selected to represent various land use patterns and anthropogenic activities. The eight typical locations were Edapally (S1), Alangad (S2), Thripunithura (S3), Parur (S4), Puthuvypin (S5), Vypin (S6), Palluruthy (S7) and Mulamthuruthy(S8). Sampling, preservation and analysis protocols strictly followed the standard methods (APHA, 2000). All the samples were collected and preserved in pre-acid washed 1000ml polyethylene terephthalate (PET) bottles. All the PET bottles were rinsed with surface water of each location before sampling and samples were taken from 100 mm below the surface and away from the edge of surface water bodies. The samples filtered through 0.45 μ m cellulose nitrate membrane filter.

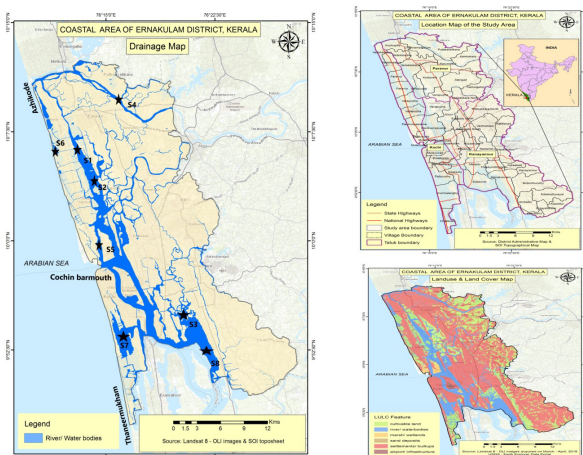


Fig. 1. Location of research area with sampling locations & land use and land cover

Samples were acidified to pH 1-2 with diluted HNO_3 and were stored in -4°C prior to the analysis.

Heavy metal analysis

The concentration of selected heavy metals, i.e. Zn, Ni, Cd, Cu and Ni in all the surface water samples were analyzed using Atomic Absorption Spectrophotometer (AAS).

Statistical analysis

Multivariate analyses of heavy metals in surface water samples were performed using correlation coefficient, Principal component analysis (PCA), and cluster analysis by the software package SPSS version 20.0. The correlativity among the analyzed heavy metals can be measured by Pearson's correlation or Spearman's correlation. The correlation procedure usually computes the pair wise associations for a set of variables and the result is displayed in a matrix. The correlation coefficients computed by correlation procedure lay in the range "1 (for the cases in which a perfect negative relationship exists) to +1 (for a perfect positive relationship) and a value of 0 indicates there is no linear relationship among the variables. For normally distributed variables, the Pearson method can be used to calculate the correlation coefficient. For normally distributed variables, the Pearson correlation was used for bivariate correlation, otherwise non-parametric Spearman method was applied (Zaman *et al.*, 2012). The PCA analyses are done for reducing the complexity of variables to a few latent factors for analyzing the relationship among the variables. In PCA, transformation of the original variables into new, uncorrelated variables (axes), called the principal components is done. This reduces the contribution of less significant variables to simplify the data structure. All significance statements reported in this study are at the $P < 0.05$ level. The cluster analysis classifies a set of observations into two or more mutual groups based on their combination of the variables (Perumal *et al* 2021). Cluster analysis was performed on the data using Ward method and squared Euclidean distance. A dendrogram was produced by cluster analysis. Five studied heavy metals were classified into five groups based on spatial similarities and dissimilarities.

GIS analysis

Geographical information system is used to analyze the spatial distribution of heavy metals in the water samples from Cochin backwaters. The inverse dis-

tance weighted (IDW) approach using Arc.GIS 10.2 software was used for this purpose. This method employs a specific number of nearest points, which are weighted according to their distance from the points being interpolated.

Results and Discussion

Mean heavy metal concentrations in surface water

The basic statistics heavy metals measured during the sampling period at eight typical sites as well as the background values of Cochin backwaters (Anju *et al.*, 2014) are summarized in Table 1. According to the background values of heavy metals in Cochin backwaters (Anju *et al.*, 2014; Robin *et al.*, 2012), it indicated that the heavy metal concentration of surface waters in this study were higher for than their corresponding background values except in concentration of Pb. The concentration of Ni is 17 times, Cd is 4 times and Cu is 7 times higher than their background values. The mean concentration of Ni, Cd, Cu and Zn is higher than their background values, except that of Pb.

Comparing the surface water heavy metal data of coastal area of Ernakulam district with that of the published data of other water bodies at home and abroad, revealed that the heavy metals in coastal water bodies of Ernakulam district were polluted, especially Ni and Cd. The concentration of heavy metals in Muvattupuzha River is much lower than Periyar River which showed higher concentration in Zn, above the USEPA standard (1995). Higher concentration in Zn is also noticed developed countries (China and Georgia), showing higher degree of pollution. Comparing with the heavy metals in surface water samples of coastal areas of Ernakulam district with other rivers in home and abroad, these samples showed higher concentration in Ni and Cd, which can cause the destruction of aquatic life.

Spatial distribution of heavy metals in surface water from coastal water bodies of Ernakulam district

The spatial distribution patterns of Ni, Cd, Cu, Pb and Zn in the surface water bodies from the coastal areas of Ernakulam district are shown in Fig.2. using IDW method. The coastal areas of Ernakulam district was divided into zones as the southern zone extending from south of bar mouth (S1, S2, S4, S5, S6) and a northern zone extending north of Cochin bar mouth (S3, S7, and S8).

According to Fig.2, concentration of Ni from each sampling sites decreased in the order S6>S5>S4>S3>S2> S1>S8>S7. The nickel toxicity is higher in marine invertebrates effecting their survival and reproduction. USEPA (1995) reported the toxicity of nickel for aquatic organisms is 0.074mg/l. All the sampling sites showed above the standard with increase in concentration of Vypin area near the fishing harbor. A lower concentration is noticed in Palluruthy area, which is not much effected by the after effects of fishing and urban settlements.

Even though Cu and Zn are micronutrients for the aquatic life in natural water bodies, they can be toxic at higher concentration thereby causing the destruction of the algae and some fishes by causing damage to gills and other organs. The spatial distribution of Cu decreased in the order S1>S4>S5>S6>S2>S6>S8>S3 and all the sampling sites showed concentration below the USEPA (2007) permissible limit of 0.048 mg/l. By comparing the average concentration of Zn in the sampling sites decreased in the order S1>S7>S8 >S2>S6>S3>S5>S4 and all the 100% of the samples are below the USEPA standard (1995) of 0.090 mg/l. A lower concentration of Pb can cause a threat to the marine life than other heavy metals (Sandip et al 2003). The comparison of spatial distribution of Pb showed a decrease in the order S3>S4>S6>S1>S5>S7>S8>S2

Table 1. Summary statistics of heavy metals in surface water from coastal waters of Ernakulam district in mg/l.

	Ni	Cd	Cu	Pb	Zn
Minimum	0.089	0.021	0.021	0.039	0.020
Maximum	0.633	0.109	0.036	0.053	0.025
Mean	0.449	0.059	0.025	0.047	0.022
S.D*	0.175	0.0342	0.0053	0.0049	0.0019
Background*	0.036	0.024	0.032	0.061	0.017
USEPA standard for surface water for aquatic life	0.074	0.033	0.048	0.210	0.090

S.D.: Standard deviation.

Background: the background value of the heavy metals in Cochin backwater Ernakulam district. (Nair *et al.*, 1990)

and all the sampling sites have the concentration below the USEPA (1984) standard of 0.210 mg/l

Cd has been proved to be carcinogenic to humans and cause damage to the ecological communities in higher concentration. The Cd concentration showed the decrease in spatial distribution in the order S5>S6>S3>S7>S1>S2>S4>S8. The concentration of Cd is above the standards of USEPA (2016) in 50% of samples, i.e. near the Vypin fishing harbor (S5 and S6) which is in the northern zone of Cochin barmouth and in the southern zone (S3 and S7) near the residential and aquaculture area. The concentration of Cd in other sampling sites (S1, S2, S4 and S8) is below USEPA standard of 0.033 mg/l.

Statistical analysis

Spearman’s correlation analysis provides the relationship between the heavy metals and their sources in the environment. In order to deduce the probable common source of metals in surface water samples,

the bivariate correlation procedure was used. In this procedure computing of the pair wise associations for a set of metals is done and is displayed as the results in a matrix. This helps in determining the value of association of the investigated metals. As the obtained data was not normally distributed, Spearman method was applied. The Spearman’s correlation coefficient of heavy metals in surface water from coastal areas of Ernakulam district was done and is shown in Table 5. Table 2 presented the values of Spearman’s correlation coefficient, where the $r > 0.50$, $p < 0.05$ indicated a significant correlations. A significant negative correlation of Zn was noticed with Ni ($r = -0.776$) and a positive correlation of Ni with Cd (0.571) a significance of 0.05 level. The other heavy metals in the study showed no significant correlation with each other.

In order to examine the suitability of data for PCA analysis, Kaiser-Meyer-Olkin (KMO) test were performed. KMO test is the measure of adequacy of

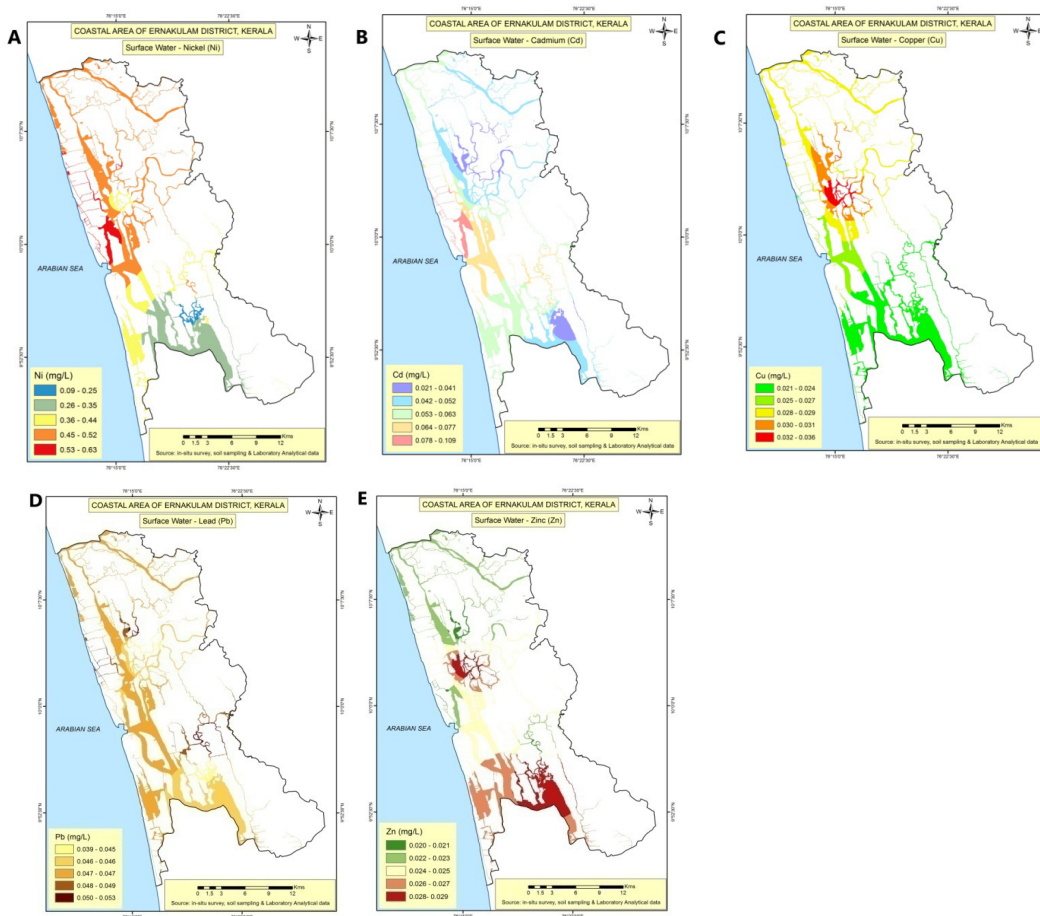


Fig. 2. Spatial distribution of Ni(A), Cd (B), Cu (C), Pb (D), and Zn (E)

sampling, indicating the proportion of variance caused by underlying factors. The KMO test value was 0.528, showing the suitability of the heavy metal concentration data in surface water samples from coastal areas of Ernakulam district to be suitable for factor analysis. (Le *et al.*, 2013). Bartlett's test of sphericity indicated correlation as identity matrix with a significance less than 0.05. The Principal component analysis (PCA) was applied to datasets of 5 variables separately for 8 sampling sites to identify sources of heavy metals in surface water samples from coastal areas of Ernakulam district by Varimax rotation with Kaiser Normalization. The Eigen values >1 and the number of significant factors and the percentage of variance were calculated and shown in Table.6. The result indicated the extraction of three components with total variance of 85.08%. The first component explained 41% of total variance loaded heavily Zn, Ni and Pb. The Second PCA explained the dominance of Cd, accounting a variance of 23.12%. The third PCA was dominated by Cu for total variance of 20.95%.

A hierarchical clustering of the data was done, after standardization and calculation of Euclidean distance for similarities of variables using ward's method. The cluster analysis was used for the grouping of the analyzed parameters i.e. data of the heavy metal concentration. Fig.3 showed 3 statistically significant clusters. (1) Zn, Pb and Cu identified as low contamination level (2) Cd as considerable level (3) Ni as higher level of pollution

The sampling sites were organized and Cluster analysis was done. The dendrogram was obtained which showed polluted sites spatially in clusters. Fig. 4. showed statistically significant clusters as three. (1) S5 and S6 belonged to cluster 1 taken as highly polluted areas. (2) S1, S2, S3, S4 and S8 in moderately polluted (3) S7 as least polluted sites.

Extraction Method: Principal Component Analysis.

Rotation Method: Varimax with Kaiser Normalization.

Rotation converged in 5 iterations.

Bold data are the main contribution element to component

Heavy metal source identification

The main sources of studied heavy metals in surface waters from coastal areas of Ernakulam district identified based on the spatial distribution, Spearman's correlation, Principal component analy-

Table 2. Heavy metal concentration in surface water samples at home and abroad (mg/l)

Study Area	Ni	Cd	Cu	Pb	Zn	References
Muvattupuzha river, Kerala	NA	0.002-0.003	0.001-0.003	0.002-0.004	007-044	Anju <i>et al.</i> , 2011
Cochin Estuary, Kerala	007-0.014	0.002-0.022	0.001-0.032	0.025-0.082	0.002-0.014	Nair <i>et al.</i> , 1990
Periyar river, Kerala	NA	0.003-0.013	0.001-0.002	0.008-0.017	0.41-0.258	Anju <i>et al.</i> , 2011
Agniar estuary, Tamil Nadu	NA	NA	0.0116	NA	0.023	Kamalakkanan <i>et al.</i> , 2016
Mahanadhi, Paradeep, Odisha	0.004.007	0.002-0.006	0.002-0.017	0.0012-0.003	0.016-0.032	Mahandha <i>et al.</i> , 2020
Wen-Rui Tang River, China	NA	0.010	0.020	0.004	0.72	Qu <i>et al.</i> , 2018
Kamaphuli River, Bangladesh	NA	0.011	NA	0.016	NA	Ali <i>et al.</i> , 2016
Mashavera River, Georgia	0.046	0.009	0.047	0.011	0.106	Withanachchi, 2018

NA: not available

sis and cluster analysis are (1) Ni which generally originates from domestic sewage, effluents from the port area and use of paints for fishing boats etc. (2) Cd pollution from effluents from the industries and the concurrent tides entering the cochin backwaters (3)Zn from the river Periyar polluted by effluents mainly from fish processing industries, fertilizer industry, heavy metal processing unit, pesticides, insecticides and associated industries.

Table 3. Spearman correlation coefficient (r) of heavy metals in the sampling stations

Variable	Zinc	Nickel	Cadmium	Copper	Lead
Zinc	1.00				
Nickel	-0.776*	1.00			
Cadmium	-0.158	0.571*	1.00		
Copper	-0.061	0.286	-0.048	1.00	
Lead	-0.494	0.347	0.036	-0.168	1.00

* Correlation is significant at the 0.05 level

Table 4. Rotation component matrix for heavy metals in sampling stations

Elements	PC1	PC2	PC3
Zinc	-0.908	-0.071	0.107
Nickel	0.744	0.478	0.177
Cadmium	0.101	0.948	-0.198
Copper	0.035	-0.153	0.980
Lead	0.813	0.020	0.073
Eigen values	2.050	1.156	1.048
% of Variance	41.00	23.12	20.95
% of Cumulative	41.00	64.12	85.08

One group of elements including Cd and Ni had a similar spatial distribution of higher polluted sites, mainly located in the northern side of the Cochin backwaters (S5 and S6). The petro chemical industries near harbor stations, fish landing zones, market near the area, in northern side of Cochin bar mouth, had led to the distribution, movement, availability of chemical elements in these regions. In the studied region, area with city land use is extensively distributed in northern and central part of the study area. Saraladevi *et al.*, 1979; Kaladharan *et al.*, 2011 and Anju *et al.*, (2011) had reported high concentrations of dissolved Zinc and Cadmium was located towards the northern side of the Cochin backwater which is near the industrial area. In the northern zone of Cochin bar mouth, the Periyar river polluted with industrial effluents and synchronous tides entering through Cochin barmouth and

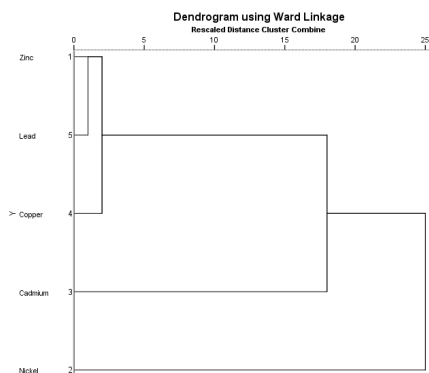


Fig. 3. Dendrogram of the heavy metal concentrations in coastal waters of Ernakulam district

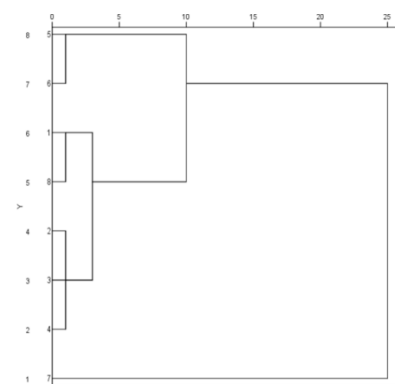


Fig. 4. Dendrogram of the sampling sites (1-8) from the coastal waters of Ernakulam district

Azheekode, had led to high on deposition of Cd and Zn in points (Anu *et al* 2014). In the present study, the sampling stations (S1 and S2) forming the entry point of river Periyar which was polluted by industrial effluents also showed moderate level of Cd and Ni. In the present study the southern region also showed moderate level of pollution Ni and Cd, (S3 and S8) this may be due to the agricultural and domestic waste from the residential regions. Anu *et al.*, 2014 reported that the southern zone of the Cochin estuary showed agricultural and domestic activities influence major portion in the southern part of the estuary makes less pollution compared with the northern arm. It was also reported by Kaladharan *et al.* (2011) that, in Cochin estuary as well as in Cochin inshore waters, cadmium level have reached critical point, while copper and lead have attained levels of caution. Robin *et al.*, (2012) reported the decrease in concentration of dissolved metals in water in the order Pb > Cd > Hg thereby exhibiting a spatial pattern with highest value at

harbor region. The Principal component analysis indicated that the first group of elements Zn, Ni and Pb mainly originated from the point source of pollutants from harbor, fish landing and marketing area with a negative loading for Zn. The concentration of Zn and Pb is below the USEPA standard for surface water for aquatic life of 0.210 and 0.090 mg/l respectively. The second group of element is Cd from, which may be non-point source of agricultural pollution, industries as Cd was clearly separated from other heavy metals in PCA analysis. The third group include Cu, might have a mixed source, from natural erosion, industrial waste and municipal sewage from the urban area.

In this work, five heavy metals from the surface water from the coastal area of Ernakulam district were investigated. This resulted in the assessment that the northern zone of Cochin backwaters is considerably polluted due to Ni and Cd, compared to the southern zone. The main cause of degradation of the estuary is the discharge of industrial, harbor and of municipal sewage from the upper northern urban areas. Movement of ships, barges, fishing boats and shipping activities were also among the major sources responsible for deterioration of the Cochin backwaters. As a whole, the water bodies in the coastal areas of Ernakulam district faces moderate anthropogenic pollution risk. Based on the statistical analysis of the sampling area, the northern zone of the Cochin backwaters is identified as the priority area for environmental monitoring and management. The concentration of Ni and Cd in some regions, exceeded the USEPA standard for surface water for aquatic life. This is to be mitigated and more attention is to be given due to accumulation of heavy metals in organisms which in turn affect the seafood industry and in turn affects the public health.

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Conflict of interest

The authors declares that there is no conflict of interest

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