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Geochemical Characteristics of Heavy Metals Concentration in Sediments of Kayamkulam Estuary

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ABSTRACT
This research work was focused to evaluate the concentration of heavy metals (As , Cd , Cu ,Pb , Hg) in sediment samples collected from the Kayamkulam Estuary. The heavy metals in sediment samples were discover by the atomic absorption spectrophotometer. Results of the analysis manifest that the measured heavy metals have a following tendency to accumulate in the order Hg>Cd>As>Pb>Cu. Remarkable spatial and temporal fluctuations and enriched accumulation was observed for heavy metals in sediments from the study. The pollution status was assessed using Enrichment Factor, Index of Geoaccumulation, Contamination Factor, Degree of Contamination and Pollution Load Index). According to geoaccumulation index, the sediments are generally categorized as unpolluted with account to the detected heavy metals. The enumerated Enrichment Factors make obvious that all heavy metals except Hg have values of up to 1, which clearly indicate that enrichment by the way of lithogenic and anthropogenic sources . Results of the Pollution Load Index finish that sediments from the study areas are generally unpolluted.
***Keywords** : Enrichment Factor; Kayamkulam Estuary; lithogenic; Pollution Load Index.*

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INTRODUCTION
Geochemistry of sediments has paramount importance in evaluation of estuarine health and management (8). Among the various contaminants heavy metals are serious pollutants of aquatic ecosystems, because of their environmental persistence, toxicity and ability to be incorporated into food chain (25). Thus, sediment is an efficient settlement of accumulation and downstream transport of inorganic contaminants like heavy metals (12). Core sediments impart an essential information on the changes in the quality of the Lake from a past period.
The extent of metals and the physico-chemical characteristics of sediment could have a core role in the inter-sites accumulation of the heavy metals (34). Many Researchers have been carried out the study on the heavy mineral content in the coastal sands of Kerala (29). Predominantly, it has been conceding that natural aquatic sediments absorb persistent and toxic chemicals to levels many times higher than the water column concentration (5). Anooja (3) also reports the heavy mineral contents and provenance of late quaternary sediments of southern Kerala including Kayamkulam estuary. Ampilli (2) were found to be observed that the hydrologic sediment parameters affecting the distribution of the Venerid clam *Paphia malabarica* from Ashtamudi estuary and Kayamkulam estuary.

MATERIAL AND METHODS
Kayamkulam Estuary is a shallow brackish water lagoon streaching between Panmana and Karthikapall , it has an outlet to the Arabian sea at Kayamkulam barrage.
Four sites were selected for collection of sediments along the study area during the period of August 2021 - July 2022. Sampling was done by using Van Veen Grab sampler by Johan Van Veen,1933 preserved according to standard methods. The samples were placed in polyethylene bags and transported to the laboratory under frozen condition at 4 ^{0} C. The samples were dried in the laboratory at 104 ^{0} C for 48 hours, ground to a fine

powder and sieved through 106 micrometer stainless steel mesh wire. The samples were then kept in a polyethylene container for acid digestion. Closed vessel microwave assisted acid digestion technique under high temperature and pressure has become routine (CCME, Canadian Water Quality Index 1.0, 1999). 0.5 gm of sediment sample was put into the reference vessel. Then 25 ml of mixture 3: 2: 2 (HCL: H_2SO : HNO_3) were added, which was inserted into the microwave unit. The digested solution was cooled and filtered. The filtered sample was then made up to 50 ml with distilled water and stored in special containers. Atomic Absorption spectrometer was used to detect and measure heavy metal content in sediment sample (Phonix : 986).

GEOCHEMICAL CRITERIA ANALYSIS

The contamination status and environmental impact of metal level in the sediments can be determined with the help of two parameters; the enrichment ratio (ER) and geoaccumulation index (Igeo). The enrichment factor (EF) is a relatively simple and easy tool for assessing the enrichment degree and comparing the contamination of different environmental media (6). The ER is a normalization method proposed by Simex and Helz [31] to assess the concentration of the metals, it normalizes metal concentration as a ratio to another constituent of the sediments. Rubio stated that there is no consensus about the most appropriate sediment constituent to be used for normalization. Al, Fe, total organic carbon, and grain size have been used as the normalization. The constituent chosen for this purpose should also be associated with finer particles (Related to grain size) and its concentration should not be anthropogenically altered (1). Therefore, in the present study, it has chosen to normalize metal concentrations using Fe. The EF is defined as follows:

$$\mathrm{EF} = \frac{(\frac{M}{Fe})_{Sample}}{(\frac{M}{Fe})_{Background}}$$

Where (M/Fe) Sample is the ratio of metal and Fe concentrations in the sample, and (M/Fe) Background is the ratio of metal and Fe concentrations of the background. The world average shale and the world average soil are among the materials obviously used to provide background metal levels. Thus, the background concentrations of As, Cu, Pb, Hg, Cd, and Fe in the average shale obtained from (33) are used in this study (Table 1). According to Zhang and Liu (35) E*F* values between 0.5 and 1.5 indicate the metal is entirely from crustal materials or natural processes, whereas E*F* values greater than 1.5 recommended that the sources are more probable to be anthropogenic.

Contamination Factor (Cf) and the Degree of Contamination (Cd) are widely used to assess the contamination status of sediment. Cf values were calculated using the following expression: $Cf = C_{metal} / C_{background}$

C_{metal} is the concentration of metal in sediment, while $C_{background}$ is the background value for the metal. The average composition of shale from (33) was used as background values for these heavy metals.

The degree of contamination (CD) was enumerated as the sum of the determined contamination factors (CF) for each of the quantified heavy metals in the site. CF values for describing the contamination status were shown in (table2).

Geochemical index (I_{geo}) was originally stated by Muller (1969) in order to determine and define metal contamination in sediments by comparing current concentrations with preindustrial levels, I_{geo} is calculated as follows: $I_{geo} = \log_2 [Cn/1.5B_n]$

Where C_n is the measured concentration in the sediment for the metal n, B_n is the background value for the metal (33), and the factor 1.5 is applied as of feasible variations of the background data cause to lithological variations. The quantity I_{geo} is calculated using the global average shale data from Turekian and Wedepohl (35). Muller determined the descriptive classes for Increasing *I*geo values (24) which are given in (Table3).

According to Tomlinson (33) , the Pollution load index (PLI) was also used in this study for a single site is the nth root of n number multiplying the contamination factors (CF values) together:

$PLI = (CF1 \times CF2 \times CF3 \times .... CFn)\ 1/n$

Where CF value assigned as the contamination factor, n is the number of metals.

Pearson's correlation coefficients (r) were used to determine the correlations between concentrations of heavy metals in the sediment.

Table 1 Enrichment Factor (EF) Categories [23]

Enrichment Factor	Categories
F<2	Deficiency to minimal enrichment
2 ≤ EF <5	Moderate enrichment
5 ≤ EF <20	Significant enrichment
20 ≤ EF < 40	Very high enrichment
EF ≥ 40	Extremely high enrichment

Table 2 Contamination Factor and Level of Contamination (Hakanson 1980)

Contamination Factor (Cf)	Level of Contamination
Cf<1	Low contamination
1≤Cf<3	Moderate contamination
3≤Cf<6	Considerable contamination
Cf>6	Very High contamination

Table 3 Muller's Classification for the Geo-Accumulation Index

I_{geo} Value Range	Class	Sediment Quality
≤0	0	Uncontaminated
1 – 2	2	Moderately
2 – 3	3	From moderately to strongly
3 – 4	4	Strongly
4 – 5	5	From strongly to extremely
≥6	6	Extremely

RESULT

GEOCHEMICAL CRITERIA ANALYSIS

The EF values of (Table 4) the metals show depletion trend As, Cd,Cu, and Pb (<1). Hg value was fairly high (1.4mg/G) at site 4, shows a mild enrichment Hg > 1. Almost uniformly lower values along the entire area negate the absence of local enrichment factors. The contamination factor was very useful to assess the contamination status of sediments of Kayamkulam backwater. The measured CF values for various heavy metals were presented in (Table 5). Cf values and Degree of contamination clearly indicates that all the measured heavy metals exhibiting very low contamination status in sediment sample. On the basis of the mean values of Cf, sediments samples enriched for metals in the same order: Hg>Cd>As>Pb>Cu. According to the Muller classification of Geoaccumulation index, the sediments samples were found to be in class 0, uncontaminated (Table 5). On the basis of the mean values of I geo index, the accumulation of metals in the sediments in following order: Hg>Cd>As>Cu>pb. The Pollution Load Index gives a simple but a comparative means for determining a site quality, where a value of PLI < 1 denotes perfection; PLI = 1 presents that only baseline levels of pollutants are presented and PLI > 1 would indicate declining of site quality (36). The PLI values for heavy metals in the Kayamkulam estuarine sediments are listed in (Table 6).

Table 4 Enrichment Factor of Heavy Metals from Study Sites of Heavy Metals from Study Sites

	Enrichment Factors				
Study Sites	Arsenic	Cadmium	Copper	Lead	Mercury
Ayiramthengu	0.05	0.2	0.005	0.01	0.7
Valiyazheekal	0.05	0.3	0.005	0.009	0.7
Kochiyude jetty	0.08	0.4	0.007	0.01	0.6
Choolatheruvu	0.07	0.3	0.008	0.01	1.4

Table 5 Contamination Factor (CF) and Degree of Contamination (CD) of Heavy Metals from Study Sites

	Contamination Factor					
Sampling Site	As	Cd	Cu	Pb	Hg	Degree of contamination
Ayiramthengu	0.009	0.057	0.0008	0.002	0.113	0.2
Valiyazheekal	0.007	0.05	0.0008	0.001	0.093	0.151
Kochiyude jetty	0.012	0.053	0.001	0.001	0.086	0.153
Choolatheruvu	0.017	0.08	0.002	0.002	0.303	0.404
Mean	0.011	0.06	0.001	0.0015	0.148	0.227

Table 6 Geoaccumulation Index Values Of Heavy Metals from Study Sites

Metal	Site1	Site2	Site3	Site4	Mean I_{geo} Values	I geo Class	Remarks
Arsenic	-0.84	-0.95	-0.73	-0.58	-0.77	0	Unpolluted
Cadmium	-2.47	-2.52	-2.49	-2.33	-2.11	0	Unpolluted
Copper	0.068	0.33	0.13	0.41	-0.234	0	Unpolluted
Lead	0.306	-0.44	-0.35	-0.11	-0.148	0	Unpolluted
Mercury	-2.17	-2.25	-2.28	-1.73	-2.45	0	Unpolluted

Table 7 Showing Pollution Load Index (PLI) values Of Heavy Metals from Study Sites

Site	PLI
Site 1	.0009
Site 2	.0002
Site 3	.0005
Site 4	.0016

DISCUSSION

The accumulation of heavy metals in sediments can be a secondary source of water pollution, once an environmental condition is changed (9). Therefore, an evaluation of heavy metal accumulation in sediments plays an efficient tool to assess the risk of an aquatic environment. In the present study, the accumulation of heavy metals in sediments were found to be observed in their order of abundance as Hg >As>Cu>Pb>Cd. The mean concentration of all heavy metals does not exceed the background values (average shale) as proposed by Turekian and Wedepohl, (35). Arsenic is one of the most dominant heavy metals causing unease from both ecological and individual health standpoints (21). The accumulation of arsenic content was higher(0.08mg/G) at site 4 during the month of April, this result was lower than that of the findings of (14) from Bogdanas river. The values of copper observed are lower during the study than the ones obtained from the reports of (27) from the sediments of Nasarawa River. The accumulation of Cd and Pb content reported are lower in sediment from the work on a water body used for irrigation in Keffi by(20). This finding was also confirmed with the reports of (30) , their results were found to be noticed that the Cd, Pb and Cu reached its maximum value in Lake Manzala sediment samples, this may be attributed to the industrial and agricultural discharge as well as from spill of leaded petrol from fishing boats. (17) reported that the Pb content may be increased as a result of dust which hold a huge amount of lead from the combustion of petrol in automobile cars. Oil tankers at harbor area, commercial boat recreational traffic within the study area released some amounts of Pb, and Cd containing compounds into the water and sediments (26),so this area contains some sources of small quantity of metal contamination at each station. The probable source of Cd in surface water sediments includes leaching from Ni - Cd batteries and untreated discharge of phosphate fertilizers (28). Concerning Hg during the study, its accumulation are higher (0.09 mg/G) in some samples collected from the study site 4, this may be closely encountered with the anthropogenic activities including the disposal of domestic, sewage wastes and industrial effluents that were detected in the study sites. This result was contradicts with the observation of (38), who reported Hg(0.034mg/G) from the sediment samples of Yangtze River system.

Inter-element correlation in sediment provides information on metal sources and pathways in the geoenvironment (10). Pearson's correlation was used for deciphering the relationship between heavy metals in sediment sample. There was a significant positive correlation noticed between metals are Cu and Cd ,similar relationship between metals can be observed by Salah et al., 2012, this strong correlation indicates that two metals have common source. While there was no significant correlation were expressed between As with HG and Cd. (18,19) reported that there were no significant correlations among most of the heavy metals studied in surface sediments of Achankovil river basin and Northern Adriatic lagoon environment respectively, suggesting that these metals are not associated with each other. The significantly positive correlation indicates that the elements were derived from similar sources and also moving together (7), it suggested that all the detected heavy metals were not strongly controlled by natural weathering processes in the study area (35). There were no increasing trends for metal concentrations among the four sites.

During this study, the assessment of metal contamination in sediment samples from the study site were further subjected to descriptive statistical methods including geochemical criteria analysis. The resulting Enrichment Factor values clearly indicate that that As, Cd, Cu and Pb, were not enriched in the surface sediments of the Kayamkulam backwater (23). The E*F* values for Hg were slightly higher (1.4mg/G) at Site 4 comparing among the other metals and it has a minor enrichment. Similar result was reported by (22), The mercury concentration was relatively high in the subsurface sediment samples, indicating the possibility of industrial mercury deposition. The difference in E*F* values for the different metals in the surface sediments may be due to the difference in the magnitude of input for each metal in the sediment (13). E*F* values greater than 1 suggest that the sources are more likely to be anthropogenic (13). The values of EF in the Kayamkulam estuarine sediments were lower than those found by (13) and EF in the Wadi Al-Arab Sediment (13). and the sediments of the Ziqlab Dam are enriched with Pb and Cd by 1.07 and 2.16 respectively. Fuel combustion also increases Pb content in soil (34).

CF values for Pb in Kayamkulam backwater sediments varied from 0.007 to 0.017 with a mean value of 0.011 (Table 5). Four sampling sites has CF less than 0 values. It was observed that all the sampling sites were not contaminated by Pb. The Igeo values for Pb in most of the sampling sites were less than 0 (<0). According to Muller's classification (Table 6**)**, the measured Igeo values for Pb clearly indicate sediment quality be considered as not polluted for all the sampling sites of the study area. Contamination Factor for Cd varied between 0.05 and 0.08 mg/kg with a mean value was 0.06 mg/kg. Sediment samples from four sites of the study area has not more than 0 . The Igeo values for Cd in Kayamkulam back water sediments ranged from -2.33 to-2.52. All sampling sites have Igeo for Cd less than 0. According to Muller's classification (Table 6), the Igeo values for Cd clearly reported that Kayamkulam backwater sediments were not contaminated. Contamination Factor for As, Cu and Hg were not more than 0 value. It was found to be that all the sampling sites were not contaminated by these metals. All the sampling sites have Igeo for As, Cu and Hg were less than 0 value, According to Muller's classification (Table 4), the Igeo values for As,Cu and Hg comes under the class unpolluted, this clearly reported that sediment samples from the study area were not contaminated by these metals. A similar study of Asegire reservoir sediments (15) and Geochemical analysis of sediment samples from Asmara drinking water reservoir (37). Their results were compiled with the findings of the current study during the study period.

The pollution load index (PLI), was used to compare effectively whether the sampling sites become polluted or not. PLI values of the analyzed samples ranged from 0.0002 to 0.0016 with a mean value of 0.0032, the PLI values were less than 1. According to quality (33), four sampling sites of the Study area suggest perfection. Asegire reservoir sediments, Nigeria (15) were also report a similar results supporting with the present study, however (18) Lake Vembanad, India reports the PLI level of >1 was calculated for suggesting the presence of heavy metal contamination in its sediment.(16) reported that the accumulation and release risks of heavy metals As, Cd,Cu,Ni, Pb, and Zn in representative contaminated river-bed and river-side sediments from a groundwater–river water interaction zone currently under restoration.

CONCLUSION

The present study reveals that the sediment of the Kayamkulam Estury was not polluted by heavy metals. Heavy metal indices like Enrichment Factor measurement manifest a minor enrichment of Hg in sediment subjected to future accumulation of metals. The inference after this study is to that these heavy metals constitute menace of contamination of the sediments and overlying surface water.

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CONFLICT OF INTEREST
There is no conflict of Interest

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