

Assessment of heavy metals in the surface sediments of the Emerald Lake using of spatial distribution and multivariate techniques

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Abstract The study aims to assess the heavy metals such as cobalt (Co), nickel (Ni), lead (Pb), chromium (Cr), cupper (Cu), zinc (Zn), manganese (Mn), and iron (Fe) from 25 surface sediment samples at different sites of the Emerald Lake, Tamilnadu, India using spatial distribution and multivariate techniques like Pearson correlation matrix and principal component analysis. From the result, the ranges of Fe, Cu, Cr, Mn, Zn, Ni, Co, and Pb are noticed to be 78,128 to 132,876; 314 to 462; 336 to 523; 520 to 701; 20.1 to 53.21; 128 to 215; 91 to 129.9; and 151

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to 158 μ g g⁻¹, respectively. The order of the average heavy metals concentration is Fe > Mn > Cr > Cu > Pb > Zn > Co > Ni. From the result, Ni, Cu, Cr, Pb, and Cd are found to be considerably correlated as they are usually related to anthropogenic activities, wastewater, and sewage. From the principal corresponding analysis (PCA) results retrieved from PC3 suggest that Fe, Mn, Cr, Cu, Pb, and Ni have common origin and are mainly due to anthropogenic input, inorganic fertilizers in agriculture, human activities, sewage effluents, traffic, and boat activities. The study relatively provides a significant approach for heavy metal pollution origin in the surface sediment in the Emerald Lake.

Keywords Geochemistry · Heavy metals · Principal component analysis · Emerald Lake

Introduction

The sediments in the lake play major role in the contamination of the lake due to the effect of physical, chemical and hydrogeochemical, and biological characteristics of the aquatic system. The reason for the heavy metal contamination in the aquatic system is mainly due to two processes; one is lithogenic process which includes the flow of water, rock weathering, and natural erosion (Singh et al. 2002)

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Fig. 1 Study area map

and the other is anthropogenic process which includes sewage discharge, atmospheric release of pollutants by industry, and fertilizer leaching from agricultural land (Wright and Mason 1999; Dinescu et al. 2004; Deng et al. 2010; Tang et al. 2010; Ahdy and Youssef 2011; Mitra et al. 2012). Change in the mineral composition of sediments and their content of macro and micro elements are the essential parameters which describe the process occurring in the lake. The chemical composition of the sediments is based on the nature of the deposited material, the weathering process, and the diagenesis and geochemistry of the specific elements (Rollinson 1993; Solecki and Chibowski 2000; Mahjoor et al.

Table 1 Summary of the heavy metals abundance in Emerald Lake

Trace element	Min	Max	Mean
Sand %	12.89	72.34	35.66
Mud %	27.66	87.11	64.34
OM %	4.6	9.8	7.35
CaCO ₃ %	2.5	9.8	5.95
C/N ratio	6.1	10.7	8.95
Fe (mg/kg)	7.81	13.88	11.49
Mn (mg/kg)	314	462	370.95
Cr (mg/kg)	336	523	411.48
Cu (mg/kg)	520	701	611.32
Pb (mg/kg)	20.1	53.21	34.04
Zn (mg/kg)	128	215	174.4
Co (mg/kg)	91	129.9	112.64
Ni (mg/kg)	151	158	154.24

Fig. 2 Spatial distribution map of sand



2009). The geochemical composition of the sediments clearly stipulates the present condition of the lake environment.

The specific information about the geochemical properties of the sediments can be gathered from the broad knowledge about the total content of the elements (Boyle 2000; Chabbi 2003; Sanei et al. 2010). Sediments composition, heavy metal concentrations, and their distribution act as a major tool for evaluating the impact of risk and pollution. The organic and inorganic source and their enrichment in the lake sediments are mainly controlled by the deposition of particulate detritus through different natural and anthropogenic sources in the lake (Meyers and Ishiwatari 1993; Holmer and Storkholm 2001).

Consequently, the geochemistry of lake sediments is a combination of different processes such as catchment lithology, weathering type, intensity effect, erosion, and deposition occurring in the lake basins (Minyuk et al. 2007; Khan et al. 2012).

At present, the geochemical investigation of the lake surface sediments is widely used to evaluate the impact on the environment, source of the sediment, intensity of weathering process, etc., in the lake environment (Tarras-Wahlberg et al. 2002; Meyers 2003; Boyle et al. 1998; Yao and Xue 2015). A comprehensive analysis about the physico-chemical parameters of the lake's surface sediments allows us to recognize the control factors responsible for the distribution pattern and the geochemistry of the

Fig. 3 Spatial distribution map of mud



eroded detritus sediments deposited in the lake (Zhang et al. 2014; Chandrajith et al. 2008).

Heavy metal pollution is determined by the sediments in a lake. Heavy metals in the surface sediments are influenced by two aspects like lithogenic and anthropogenic process (Gopal et al. 2017). Lakes have been known for their different ecosystem such as water cycling, climate change regulation which provides habitat for microorganisms in the lake. Lithogenic process includes the flow of water, rock weathering, and natural erosion (Singh et al. 2002), while anthropogenic process includes sewage discharge, atmospheric release of pollutants by industry, and fertilizer leaching from agriculture land. Geochemistry of lake surface sediments is utilized to assess the sediment weathering, environmental change, toxicity, tenacity, and biogeochemical of the lake (Last and Smol 2001; Jin et al. 2001, 2003; Laird et al. 2003; Magesh et al. 2013; Li et al. 2013; Balamurugan et al. 2015; Krishakumar et al. 2016; Gopal et al. 2017).

The primary information are gathered from the study area 58 I/1 (1:50000) by Survey of India (1973). The spatial integration maps are developed by software package like ArcGIS 10.1 and ESRI. Emerald Lake is located in the Yercaud hill station near Salem District in Tamil Nadu, India, and it falls in between the latitudes 11° 47′ 07.6″ N and 11° 46′ 58.5″ N and longitudes 78° 12′ 32.6″ E and 78° 12′ 37.7″ E (Fig. 1) with an average elevation of 1515 m asl. Yercaud has a moderate humid **Fig. 4** Spatial distribution map of OM



subtropical climate. The Emerald Lake has a minimum and maximum depth of 1 to 5.1 m respectively (Venkatachalapathy et al. 2014). It covers an area of about 5.2 ha. In January, the temperature of the Yercaud hill station is moderate. The maximum temperature was 29 °C and minimum was 15 °C and an average of 27 °C in 2017 during the winter season. Average rainfall ranges from 1500 to 2000 mm/a during in the northwest monsoon season. The region is geographically secured by charnockite rock. Laterite is the pre-dominant soil of this region with its average depth of 1.5 m. Yercaud is known for its coffee plantation and oranges in addition to bauxite and granite reserve. Heavy metals such as Fe, Mn, Zn, Cu, Cr, Pb, Ni, and Co from twenty-five surface sediments are tested at various sites of the Emerald Lake with the help of spatial integration and statistical analysis like the correlation matrix and principal component analysis.

Materials and methods

Sampling and analytical procedures

Twenty-five sediment samples of 1 to 5.1 m depth in water were collected from 25 sites of Emerald Lake using a grab sampler during January 2017 as shown in Fig. 1. The sample weighing 1 kg is collected from the lake sites. The samples are put away in fixed Ziploc sacks with lake water and tests Fig. 5 Spatial distribution map of CaCO₃



are passed to the research facility and put away at -20 °C. Prior to the substantial metal examination method, the sample is dried at 60 °C for 48 h. A small amount of dried samples are sieved through a mesh a 63-µm nylon mesh for homogenization and stored in a fixed plastic sacks. CaCO₃ examination was achieved as suggested by Loring and Rantala (1992). OM was studied subsequently after the titration method by Gaudette et al. (1974). In trace element analysis, the sediments were air-dried and disaggregated in an agate mortar. For each sample, 1 g of sediment is absorbed by using concentrated HClO₄ solution (2 ml) near dryness. Then, again a further addition of HClO₄ (1 mg) and HF (10 ml) was prepared, the mixture is then evaporated to near dryness. Finally, HClO₄ was added, then the sample was dried until the white fumes appear. The residues were dissolved in concentrated HCl and diluted to 25 ml (Tessier et al. 1979). By using grade A filters, the acid solution was filtered and the analysis of metals such as Mn, Cr, Cu, Ni, Co, Pb, and Zn were performed by inductively coupled plasma mass spectrometry, Council of Science and Industrial research – National Geophysical research Institute, Hyderabad.

Statistical technique

Usually, statistical techniques are employed to process the scientific information with regard to the conveyance and relationship among the contemplated parameters. The spatial analyst part in ArcGIS software module **Fig. 6** Spatial distribution map of C/N ratio



maps to prepare the spatial distribution maps using inverse distance weighted interpolation technique. Pearson correlation matrix and principal component analysis are determined by IBM SPSS Version 20. Statistical techniques like Pearson correlation and PCA were utilized to discover the relationship between metals and the origin of the metals, in the surface sediments of the investigation sites.

Results and discussion

Organic matter (OM) and calcium carbonate (CaCO₃)

Both the concentration of OM and $CaCO_3$ is shown in Table 1. OM and $CaCO_3$ are mainly terrestrial runoff contributions existing in the water column of the lake. Losses of organic matter are high during transportation of lake sediments (Bernasconi et al. 1997). Concentrations of the studied trace element summaries are given in Table 1. Organic Matter (OM) content varies from 9.8 to 4.6% with an average of 7.35%. Losses of organic matter are high during transportation of lake sediments (Bernasconi et al. 1997). Babeesh et al. (2017) reveals that the range of organic matter and carbonates are 3.8-13.8% and 0.2–1% (very low carbonate content) respectively. The organic matter contents of the marginal sediments and central parts of the lake are attributed to the great turbulences in the former caused by the wind generated waves that move fine organic detritus into the lake. This shows that the

Fig. 7 Spatial distribution map of Fe



lake surface sediments gain a high organic content from the lake turbulences, activities of the tourists and soil erosion. The content of CaCO₃% varies from 2.5 to 9.8% with an average of 5.95%. The amount of CaCO₃% that accumulates on the lake bed and at last buried in the sediments is a function of how much is produced in the epilimnion and how much is destroyed in the hypolimnion. However, if the sediments of these lakes contain a large amount of organic matter, more than about 12% organic carbon, small amount of CaCO₃ is preserved in the sediments. In some lakes, the increase in sedimentary organic matter and dissolution of CaCO₃ happened thousands of years ago in response to natural eutrophication. In other lakes, these processes happened or are happening now, in response to cultural eutrophication. Although increased organic productivity due to eutrophication may increase the pH of the lake water and cause greater precipitation of CaCO₃, the decomposition of the produced organic matter in the hypolimnion and sediments causes much greater dissolution of precipitated CaCO₃. In addition to the effect of organic productivity on CaCO₃ accumulation, it may also affect the biogeochemical cycles of other elements, particularly redoxsensitive elements. Williams Lake precipitates CaCO₃ during summer (McConnaughey et al. 1997) but the surface sediments are virtually carbonatefree. In the present study, the C/N ratio ranges from 6.1 to 10.7 with an average of 8.95% of the Emerald Lake sediments given in Table 1. Overall, the C/N ratio of the aquatic plants namely freshwater Fig. 8 Spatial distribution map of Mn



phytoplankton is greater than 10. A higher C/N ratio (10-20) indicates a combination of aquatic and terrestrial organic materials. In the present study, the C/N ratio that ranges from 6.1 to 10.7 indicates that there is a good aquatic plant phytoplankton (diatom) growth in the lake. It is clear that the source of the proportion of macrophytes to the phytoplankton in the aquatic environment determines the C/N ratio.

Metal concentrations in the surface sediments

The heavy metals like Fe, Zn, Mn, Cu, Cr, Pb, Ni, and Co are analyzed. From the result, the ranges of Fe, Cu, Cr, Mn, Zn, Ni, Co, and Pb are 7.812 to

13.28; 314 to 462; 336 to 523; 520 to 701; 20.1 to 53.21; 128 to 215; 91 to 129.9; and 151 to 158 mg/kg, respectively. Figures 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12, 13, and 14 show the spatial distribution map of these heavy metals. The components are positioned in the following order: Fe > Mn > Cr > Cu > Pb > Zn > Co > Ni. Fe has the most prominent component focus in the middle part of the Emerald Lake in the lake site no: 15. In comparative study, high Fe values were analyzed which ranges from 12.59 to 19.27 mg/kg at Ataturk Dam Lake in Turkey by Karadede and Unlu (2000). According to Kabata-Pendias and Pendias (1992), the higher concentration of Fe as observed in the lake sediments reveals that Fe is the most

Fig. 9 Spatial distribution map of Cr



commonly occurring metal on the earth's crust. A huge amount of Fe source might from the bauxite ore mining activity and other anthropogenic sources (Abd et al. 2011; Pereira et al. 2008). As indicated by Salomons and Forstner (1984), iron hydroxides are able to absorb large quantities of metals through cation exchange processes, and iron oxides also play a significant role in trapping metals in the lake sediments.

In the present investigation, 151 to 158 mg/kg values of Ni and 336 to 523 mg/kg values of Cr are identified to be rich since they are nearly filtered from the abutting rock exposures containing a lot of charnockite rock (mafic mineral like olivine and pyroxenes). In a further report, Ni is found chiefly in the residue fraction, despite the fact that its

substantial concentration is found to be of natural issue from the Goczalkowice reservoir (Kwapulinski and Wiechula 1993). Ni is found mainly in the residual fraction although its high concentration is found in the organic matter from the Goczalkowice reservoir sediments (Kwapulinski and Wiechula 1993). Furthermore, heavy metals, including Cr, Ni, Pb, and Cu, have a similar intonation in Taihu and Dianchi, indicating that the contamination character and detailed study of the heavy metals are comparative in the Taihu and Dianchi lakes (Wei and Wen 2012). From the study, the values of Zn range from 128 to 215 mg/kg in the Emerald Lake. The other possible sources of Zn are from boat oil, grease, sewage sludge, transmission fluids, and concrete. The results observed in this study are **Fig. 10** Spatial distribution map of Cu



consistent with the fact that Pb is generally present in higher concentrations in various sewage discharges, and lake as the sink, thus may received large quantity of external inputs of Pb in Emerald Lake, indicating that they suffer serious Pb contamination. Additional, the main sources of Co, Cr, and Co are due to the municipal wastewater, sewages, landfill leachates, and geogenic materials.

Pearson correlation

The Pearson correlation matrix used among sand, mud, OM, $CaCO_3$, C/N, Fe, Cu, Cr, Mn, Zn, Ni, Co, and Pb components are given in Table 2. The Pearson correlation clarifies the heavy metal relationship and the importance of the main cause in the environment (Karthikeyan et al. 2017; Gopal et al. 2016a, b; Godson et al. 2018). The relationship among the sediments demonstrates that all the trace components are in a strong relationship with sand and CaCO₃. Moreover, elements exhibit high positive relationship with OM and also with mud. Table 2 represents the results of Pearson's correlation analysis and their significance levels. Positiverelated mud versus C/N ($r^2 = 0.435$), Fe ($r^2 = 0.501$), Mn ($r^2 = 0.667$), Cr ($r^2 = 0.415$), Cu ($r^2 = 0.429$), Pb $(r^2 = 0.432)$, Zn $(r^2 = 0.372)$, Co $(r^2 = 0.377)$, and Ni $(r^2 = 0.618)$ is additionally seen in the reservoirs. Also, Fe is effectively correlated with Mn ($r^2 =$ 0.549), Cr ($r^2 = 0.563$), Zn ($r^2 = 0.374$), Co ($r^2 =$ 0.335), and Ni ($r^2 = 0.328$). According to the reports by Lu et al. (2010) and Saeedi et al. (2012), if the **Fig. 11** Spatial distribution map of Pb



correlation coefficient between heavy metal factors is positive, then these factors may have the common source and the identical behavior during transportation. Fe exhibits a high positive correlation with Cr $(r^2 = 0.563)$ and moderate correlation with other trace elements like Mn $(r^2 = 0.549)$, Cu $(r^2 =$ 0.374), and Co $(r^2 = 0.335)$. The elements Cr, Cu, Co, and Ni, Mn are correlated positively. Fe exhibits a very strong correlation with Mn $(r^2 = 0.549)$, Cr $(r^2 = 0.563)$, Cu $(r^2 = 0.295)$, Zn $(r^2 = 0.374)$, Co $(r^2 = 0.335)$, and Ni $(r^2 = 0.328)$. Ni, Cu, Cr, Pb, and Cd are considerably correlated as they are usually related to anthropogenic activities, wastewater, and sewage. Fe shows that the trace elements are obtained from their origin (Bhuiyan et al. 2009) and that these trace elements are redistributed in the sediments by the same hydrogeochemical process (Bai et al. 2011). Finally, significant positive correlations among Mn with Cr, Cu, and Ni component verifies that they fall in the sediments (Fianko et al. 2013). Cu, Ni, Cr, and Pb are significantly identified with anthropogenic activities. For this, Cu, Ni, and Pb are usually derived from anthropogenic wastewater and sewage; Cr is typically related to industrial activities (Li et al. 2009).

Principal component analysis (PCA)

Principal componentaAnalysis (PCA) is a general multivariate analysis which is used in various

Fig. 12 Spatial distribution map of Zn



environmental studies to identify sources of the metals in sediments (natural and anthropogenic activities) and their element characteristics (Bai et al. 2011; Anju and Banerjee 2012; Islam et al. 2015). The rotated component matrixes of the PCA are presented in Table 3. The Kaiser-Meyer-Olkin and Bartlett's Test of Sphericity values are 0.626 and 72.371 (df=28, Sig < 0.01 respectively), suggesting that PCA might be useful in dimensionality reductions. In rotated matrix (Fig. 15) analysis, the PCA plot for various parameters were obtained, then varimax normalization is applied to extract the variables. In the present study, the 13 variables from Emerald Lake surface sediments are summarized by three principal components (PCs), with the percentage of cumulative of 36.579, 49.411, and 71.043 respectively. These three components have 36.579, 12.832, and 9.821% of the variances as shown in Table 3. The loading plot of the first three principal components of the sediment samples is shown in Fig. 15. PC1 explains the 36.579% of total variance and reveals high loading of sand, CaCO₃, and C/N (0.033, 0.057, and 0.530 respectively). PC1 can be regarded as transportation activities, mainly due to the discharge of agricultural wastewater and untreated urban sewage.

PC2 (12.83% of total variance) reveals that strong loadings on Mud, Pb, OM, Zn, and Co (0.185, 0.146, 0.416, and 0.268 respectively) show high positive loading mainly because of the metals which form organic complexes with humic substance in the

Fig. 13 Spatial distribution map of Co



surface sediments (Lepane et al. 2007). These organic matter complexes in surface sediments are often transformed by geochemical composition of natural processes in this area (Loska and Wiechula 2003; Lepane et al. 2007), and PC3 (9.82% of total variance) shows such as Fe, Mn, Cr, Cu, Pb, and Ni (0.381, 0.325, 0.406, 0.179, 0.562, and 0.363 respectively). Results retrieved from PC3 suggest that Fe, Mn, Cr, Cu, Pb, and Ni have common origin and are mainly due to anthropogenic input, inorganic fertilizers in agriculture, human activities, sewage effluents, traffic, and boat activities. Association of Fe and Cr recommended that multisource urban environment prevails in the area due to human activities which is responsible with the findings by Acosta et al. (2011). Cu concentration is controlled by long-term application of inorganic fertilizers in agriculture areas (Acosta et al. 2011). In similar result, Cr and Mn for sediments, which are dominantly contributed by lithogenic sources of Cr in sediments, indicate that it is mostly contributed by anthropogenic activities (Gopal et al. 2017). The cumulative variance of the three PCA is 71.043, which clearly indicates that the lithogenic factor influences the distribution of maximum part of the studied metals. Moreover from the observations, it can be known that the Pb and Zn originate mainly by the concourse of different sewage effluents, traffic, and boat activities. Perhaps, the main source of these elements is from the man-made activities like agriculture, boating, pollution from the traffic and untreated sewage, and domestic wastes.

Fig. 14 Spatial distribution map of Ni



Table 2 Pearson correlations of clay, silt, and sand (%) in surface sediments, OM %, CaCO₃%, and C/N ratio trace element (mg/kg)

	Sand	Mud	ОМ	CCO ₃	C/N	Fe	Mn	Cr	Cu	Pb	Zn	Со	Ni
Sand	1												
Mud	-1	1											
OM	-0.043	0.043	1										
CaCO ₃	0.614**	-0.614**	-0.085	1									
C/N	-0.435	0.435**	-0.254	-0.135	1								
Fe	-0.501**	0.501**	-0.03	-0.379**	0.139	1							
Mn	-0.667**	0.667**	0.179	-0.51	-0.126	0.549**	1						
Cr	-0.415**	0.415**	-0.081	-0.528**	-0.113	0.563**	0.708**	1					
Cu	-0.429**	0.429**	-0.127	-0.108	0.033	0.295	0.416**	0.340*	1				
Pb	-0.432**	0.432**	0.019	-0.178	0.057	0.017	0.075	0.05	0.162	1			
Zn	-0.372**	0.372**	0.138	-0.391**	0.151	0.374*	-0.012	-0.086	0.076	0.082	1		
Co	-0.377**	0.377**	0.105	-0.338**	0.469**	0.335**	0.305	0.132	0.012	0.098	0.26	1	
Ni	-0.618**	0.618**	-0.043	-0.259	-0.139	0.328	0.617**	0.378	0.357*	0.532**	0.146	0.23	1

*Correlation is significant at the 0.01 level (2-tailed)

**Italic Correlation is significant at the 0.05 level (2-tailed)

Elements	Component					
	1	2	3	4	5	
Fe	0.202	-0.127	0.269	0.182	0.040	
Mn	0.436	-0.010	0.004	-0.115	-0.108	
Cr	0.423	-0.113	-0.178	-0.025	-0.078	
Cu	-0.188	-0.051	-0.111	0.035	1.084	
Pb	-0.218	0.661	-0.150	0.138	0.066	
Zn	-0.021	0.010	0.890	-0.184	-0.088	
Со	-0.157	-0.052	- 0.201	1.015	0.008	
Ni	0.267	0.531	0.234	-0.379	-0.219	
% of variance	30.65	18.51	14.73	13.877	12.843	
% of cumulative	30.65	49.15	63.88	77.76	90.604	

 Table 3
 Principal component analysis loading for heavy metals in the Emerald Lake

Extraction method: principal component analysis. Rotation method: varimax with Kaiser normalization. Component: scores

Conclusion

The heavy metals Fe, Mn, Zn, Cu, Cr, Pb, Ni, and Co are analyzed from the 25 surface sediment samples of 25 different sites of the Emerald Lake. The study has focused mainly on spatial integration, statistical analysis like Pearson correlation matrix, and PCA. The order of the heavy metal concentration is Fe > Mn > Cr > Cu > Pb > Zn > Co > Ni.



Fig. 15 Principal component analysis (PCA) of heavy metals in the Emerald Lake

From the result, the bilateral Pearsn correlation matrix of Ni, Cu, Cr, Pb, and Cd is considerably significant as these are usually related to anthropogenic activities, wastewater, and sewage. The highest component loading was established by the PCA were characterized by the highest loading of Cu, Co, and Fe. This result indicates that the Cu, Co, and Fe have different metal concentrations and were mainly sourced from mixed anthropogenic inputs, including homemade wastes and sewages. The study relatively provides a significant approach for spatial map of element pollution source in the surface sediment. Metal concentrations in other research areas are believed to be related to the natural mineralogical structure of the region like wastewater and mineral sources. Considering the individual metal, Fe, Cu, Ni, Cr, Pb, and Cd have high potential biogeological hazard for the greater part of the site of the research area. In the future, research based on geoaccumulation index, enrichment factor, contamination factor, and pollution load index of the sediments in the lake area in India may be considered as well.

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