

Assessment of trace elements in Yercaud Lake sediments, southern India

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Abstract Lake sediments are major sink for carbon and trace elements. Lake water and sediments need to be monitored continuously for environmental and geochemical explorations. In the present study, sediment characteristics, source and distribution of trace metals (Fe, Mn, Cr, Cu, Ni, Co, Pb and Zn) of Yercaud fresh water lake sediments, South India, situated at an elevation of 1515 m asl is presented. Twenty-five surface sediments were collected covering the entire expanse of the lake in a gridded pattern. Detailed chemical analyses reveal the following decreasing order of elements: Fe > Cu > Cr > Mn > Zn > Ni > Co > Pb. Geoaccumulation index, enrichment factor, contamination factor and pollution load index were calculated to evaluate the ecological impacts of these trace metals in the sediments. The index calculation indicates that all the trace elements were derived from natural process, but there are indications of anthropogenic activities. The study area was found to be highly contaminated with Cu, moderately contaminated with Cr and Co, considerably contaminated with Pb, Zn and Ni, and uncontaminated with Mn. The spatial distribution of the trace elements supports the strong association of sediment fine fraction and organic matter. The statistical principal component analysis also

confirms that the concentration of Cr, Cu, Fe and Mn was mainly derived from the natural weathering and non-point agricultural sources. Pb and Zn arise due to the confluence of sewage effluents, traffic and boat activities. These elements need to be further evaluated for pollution control and prohibiting further deterioration in the Yercaud Lake.

Keywords Lake sediments · Geochemistry · Trace metals · Principal component analysis · Yercaud Lake

Introduction

Lakes are major sinks for carbon and trace metals, and lake floor sediments are often enriched in trace metals. Trace metals adhere to the fine fractions of the sediments. Concentrations of trace metals in sediments are affected by both the geogenic and anthropogenic aspects (Lalah et al. 2008). Natural influences include benthic agitation, flow changes, rock weathering and natural erosion, while anthropogenic aspects include sewage discharge, industrial wastewater discharge, atmospheric deposition, agricultural runoff and fertilizer leaching (Romic and Romic 2003; Tang et al. 2010; Choi et al. 2012; Rodriguez-Martin et al. 2013; Su et al. 2013; Islam et al. 2014a, b; Hasrizal et al. 2015; Iqbal et al. 2016). Geochemistry of lake sediments is often influenced by both natural and anthropogenic processes, and trace metal assemblages can be used to investigate the contributions of different forcing mechanisms to the changes in the lake sedimentary environments (Koinig et al. 2003; Routh et al. 2004; Lalah et al. 2008; Li et al. 2013a, b; Ma et al. 2016). It is important to monitor and explore the lake floor sediments for trace metal toxicity, sources and health of the lake water.

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Trace metal pollution in aquatic ecosystems (lakes, rivers, estuaries and Marine) has significant reflexion due to their toxicity, tenacity and biogeochemical buildup (Magesh et al. 2011; Varol 2011; Jiang et al. 2012; Gu et al. 2012, 2014a, b; Li et al. 2013a, b; Magesh et al. 2013; Krishnakumar et al. 2015; Iqbal et al. 2016; Gopal et al. 2016a, b; Krishakumar et al. 2016). Hence, it is important to understand how climatic variations and anthropogenic activities influence the concentrations of geochemical constituents, especially trace metals. High-altitude lakes are commonly considered to be pristine because they are small and sensitive ecosystems that experience rapid flushing rates and also of limited human activity in their catchments (Katrina 1994; Vreča and Muri 2006). However, human activities have faster cycling of geochemical elements that result in elevated metal deliveries to water bodies for the past several decades (Zahra et al. 2014; Zeng et al. 2014). Lacustrine sediments provide archives of natural evolution and anthropogenic influences on lakes and their catchments (Leorri et al. 2014; Guo et al. 2015). An assemblage of sediment organic matter (OM) in these lakes is generally high (Vreča and Muri 2006; Choudhary et al. 2009a, b) because of multiple processes, including high productivity, rapid sedimentation and anoxic conditions on the lake bottom (Meyers 2003). As a result, these organic-rich lacustrine sediments retain their original source signatures (e.g., C/N ratio, specific biomarkers) and thus reflects environmental conditions at the time of deposition. Based on studies on investigative environmental recent changes in lacustrine environments, natural processes affect the transportation of geochemical elements from watersheds to lakes (Braun et al. 2013; Kylander et al. 2013).

A review of recent geochemical work carried out on lake sediments reveal that the fine fractions are positively correlated with organic matter (OM) and total organic carbon (TOC) (Vijayaraj and Achyuthan 2016). Moreover, C/N ratio suggests that OM was largely produced by the lake algae in the Kukkal Lake, while there is a mixed resource of OM in the Berijam Lake (Vijayaraj and Achyuthan 2016). Trace metal analysis of the Kodaikanal Lake sediments indicated that the natural environment surrounding the Kodaikanal Lake is severely contaminated by heavy metals (Cd, Pb, Zn and Cu), and multivariate statistics (discriminant analysis) revealed that the plots fall scattered in the turbidite and eolian environment (Balamurugan et al. 2015). Further, the mercury contamination status of the Vembanad Lake was classified as less contaminated when compared to other mercury-polluted areas around the world (Mohan et al. 2014). However, the surface sediments have the high concentration due to the anthropogenic sources like agricultural runoff, small-scale industries and fuel burning. Further, no systematic

geochemical analyses till date have been carried out on the lake floor sediments that reflect the source area and plausible causes for the contamination.

Yercaud fresh water lake is a popular hill resort in the Shevaroy range of hills in the Eastern Ghat region of southern India. It is also one of the major tourist attractions in the state of Tami Nadu, India, which has been strongly affected due to urban development. The lake receives toxic metals and organic and inorganic pollutants from different sources such as rock weathering, illegal construction activities, automobile exhausts and painting of boat in tourist season every year (Purushothaman et al. 2011). It is serving as important venues for recreational activities. As increasing numbers of tourists visit to escape the summer heat in the plains and enjoy the peaceful mountain setting, it stresses the limited resources in Yercaud town, with respect to housing, transportation, food and drinking water supply. This is evident from increasing levels of environmental pollutants in the sediments and surface water in this region (Chakrapani 2002; Das 2005).

Even though several studies have been carried out to evaluate the quality and chemistry of sediments in some important freshwater lake locations in India, only a few studies are pertinent to the freshwater lake sediments of the entire Indian region. Therefore, the present study assesses the trace metal contamination (Fe, Mn, Cr, Cu, Ni, Co, Pb and Zn) and sediment provenance in the Lake floor sediment samples from Yercaud Lake. The main objectives of this study were to: (1) assess the concentration and spatial distributions of selected trace metals (Fe, Mn, Cr, Cu, Ni, Co, Pb and Zn) in surface sediments from Yercaud Lake, South India, (2) determine the potential environmental risk using multi-pollution indices such as enrichment factor (EF), geoaccumulation index (I_{geo}), contamination factor (CF) and pollution load index (PLI) and (3) identify the nature and/or anthropogenic sources of these metals. It is further predicted that this study would provide geochemical data related to the spatial distribution and contamination of the metals in the freshwater lakes, which would provide essential information to support lacustrine environmental control actions for the anthropogenic pollutants in the natural ecosystem. In the present study, we also compare the trace metal contamination of Yercaud Lake with other high-altitude lakes in the Indian and other regions (Table 5).

Materials and methods

Regional setting

Yercaud is located (11°13'15"N and 77°28'07"E) in the Servarayan range (anglicized as "Shevaroy") of hills in

the Eastern Ghats near Salem, Tamil Nadu. It is located at an altitude of 1515 m from the above mean sea level (Fig. 1) (Sakthivel et al. 2015). The town gets its name from the lake located at its center: in Tamil “Yeri” means “lake” and “Kaadu” means “forest.” The lake is “horse-shoe” shaped and has an effective area of 20.0 acres (8 ha). The lake is also known as emerald lake, and it is the only natural lake among all the hill stations in the southern part of India. It is also called as the Jewel of the South and “Poor man’s Ooty” (Rajkumar et al. 2006). The geological formation in this area comprises both basic and acid types of charnockite bedrock of the Archaean age, weathered into the rugged masses of hills. The Shevaroy range is extensively covered with green grass and has no dense forest cover. The area is under the influence of both the southwest and northeast monsoons, but the northeast monsoon chiefly contributes to the rainfall in the district (1500–2000 mm/a). The climate of Yercaud is the moderate one with the maximum temperature of 34 °C during summer (March to May), and the minimum is 16 °C during the winter; it has a maximum depth of 5.1 m and a minimum depth of 1.5 m (Venkatachalapathy et al. 2014).

Sample collection and storage

Twenty-five surface sediment (0–15 cm, top layer) samples were collected from Yercaud Lake, South India during December 2015. The sediment samples were collected in pre-cleaned zip-locked polythene bags by using a sediments snapper (Ø5 cm). The collected samples were placed in an ice cooler and transported to the laboratory immediately. Then, the samples were dried, ground, homogenized and sieved through ASTM 230-µm nylon mesh for further processing.

Analyses and quality control

Particle size (sand and mud) was determined with a particle size analyzer (Malvern Mastersizer 2000) with a measurement range of 0.02–2000 µm. The Mastersizer 2000 automatically determines size fractions with a measurement precision <1%, and the repeated measurement error is less than 2%. Moment measures were calculated using GRADISTAT version 4 based on Folk (1974) nomenclature. Carbonates in the collected sediments were measured

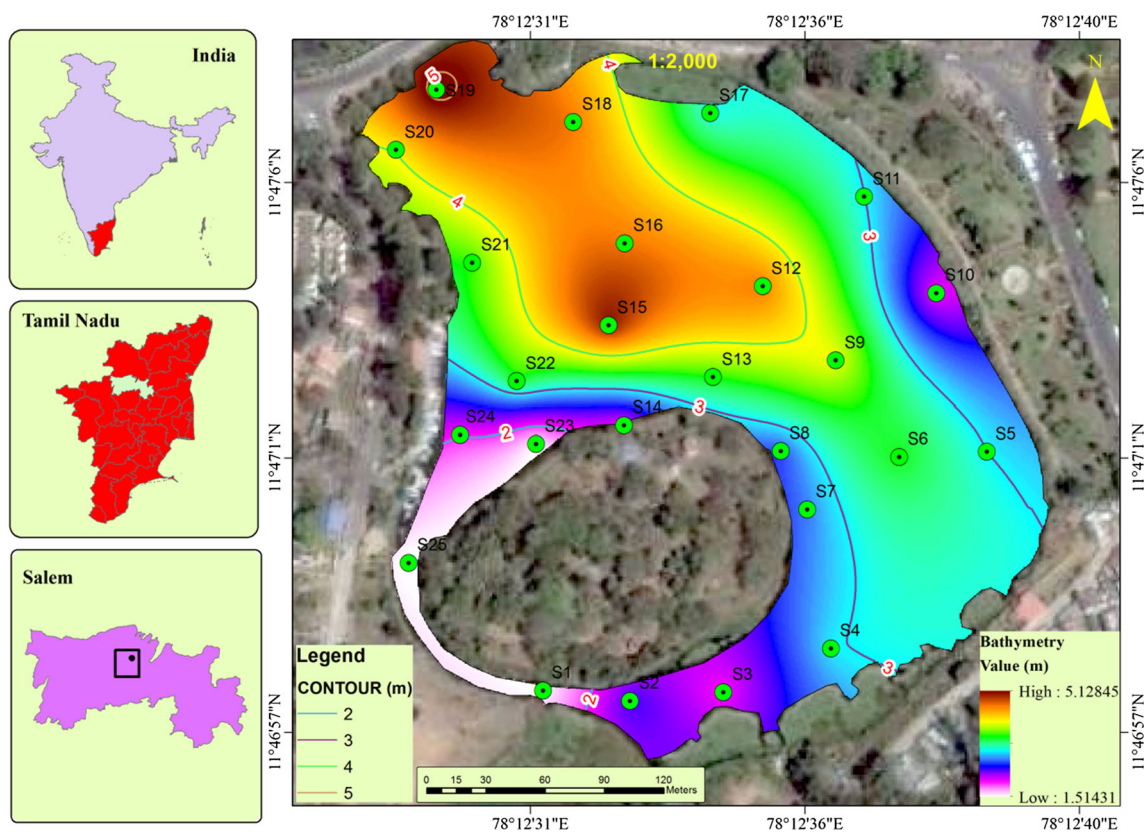


Fig. 1 Location map of the Yercaud Lake

Table 1 Published and obtained analytical results of MESS-1

Elements	MESS-1	Present study	Recovery %
Fe	3.1 ± 0.38	2.95	95.16
Mn	513 ± 25	472	92.00
Cr	71 ± 1.1	69.8	98.30
Cu	25.11 ± 3.88	22.9	91.19
Ni	29.5 ± 2.7	28.3	95.93
Co	10.8 ± 1.9	9.9	91.66
Pb	34 ± 6.1	31.8	93.73
Zn	191 ± 17	173.3	90.73

following the method of Loring and Rantala (1992). Organic matter (OM) was estimated following the titration method by Gaudette et al. (1974). For TTMs analysis, the sediments were air-dried and disaggregated in an agate mortar, before chemical treatment. For each sample, a known quantity (1 g of sediment) was digested with a concentrated solution of HClO₄ (2 ml) and HF (10 ml) to near dryness. Subsequently, a second addition of HClO₄ (1 ml) and HF (10 ml) was prepared, and the mixture was evaporated to near dryness. Finally, HClO₄ alone was added, and the sample was dried until white fumes appeared. The residue was dissolved in concentrated HCl and diluted to 25 ml (Tessier et al. 1979). The acid solution was subsequently filtered using grade A filters, and analyses of metals (Mn, Cr, Cu, Ni, Co, Pb, and Zn) were performed by graphite furnace atomic absorption spectroscopy (GFAAS) PerkinElmer AA-700. The accuracy of the analytical procedures in the present study was assessed using the certified reference material MESS-1 (Table 1) from the National Research Council of Canada (Sundararajan and Srinivasalu 2010; Kalpana et al. 2016; Gopal et al. 2016a, b).

Normalization of trace metal concentration

The trace element enrichment in the lacustrine sediments was normalized to eliminate the influence of grain size and mineral composition. Some types of extraordinarily unaffected metals such as Al, Fe, Mg, Ti, Sc, Li and Cs have been used to normalize the grain size effect for metal concentrations in sediments (Schropp et al. 1990; Loring 1990; Herut et al. 1993; Grousset et al. 1995; Aloupi and Angelidis 2001; Soto Jiménez et al. 2001; Wu et al. 2007; Amin et al. 2009; Mil-Homens et al. 2009; Kim et al. 2010; Bing et al. 2011; Gopal et al. 2016a, b). In the present study, the results are compared with that of mean crust values to identify the present level of enrichment in all stations of Yercaud Lake. Iron has been chosen as normalization element because of its origin being exclusively lithospheric (Bloundi et al. 2009; Kalpana et al. 2016).

Indices of sediment pollution

Detecting the sources of trace metals is of particular importance in environmental investigation. Pollution indices were used for the environmental assessment of Yercaud Lake sediment. The geoaccumulation index (Müller 1981), enrichment factors which were interpreted as suggested by Bloundi et al. (2009), contamination factor and contamination degree (Hakanson 1980) are used to assess heavy metal contamination in sediment. I_{geo} , EF and CF are the top most common tactics to assess the ecological risk by an individual element, whereas PLI appraises the environmental risk posed by mixed elements (Caeiro et al. 2005; Li et al. 2012; Zhao et al. 2012; Cheng et al. 2013; Hou et al. 2013; Wang et al. 2014a, b; Iqbal et al. 2016).

Index of geoaccumulation (I_{geo})

The outstanding reciprocal technique for estimating the enrichment of metal concentration above background is the geoaccumulation index (I_{geo}). This index was introduced by Müller (1981) in order to determine and define metal contamination in sediments by comparing current concentrations with pre-industrial levels. The geoaccumulation index (I_{geo}) was used by most of the researchers for trace element studies in sediments (Jumbe and Nandini 2009; Bing et al. 2011; Magesh et al. 2011; Hasan et al. 2013; Hou et al. 2013; Zahra et al. 2014; Arunachalam et al. 2014; Kalpana et al. 2016; Gopal et al. 2016a, b).

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

where C_n is the measured concentration of examined element in the sediment sample and B_n is the geochemical background for the element which is either directly measured in pre-civilization (pre-industrial) reference sediments of the area or taken from the literature (average shale value described by Taylor (1964). The factor 1.5 is introduced to include possible variation of the background values that are due to lithogenic variations (Chakravarty and Patgiri 2009; Goher et al. 2014), as well as very small anthropogenic influences (Qingjie and Jun 2008).

Six classes of the geoaccumulation index was proposed Müller (1981)

I_{geo} value	I_{geo} class	Designation of sediment quality
>5	6	Extremely contaminated
4–5	5	Strongly to extremely contaminated
3–4	4	Strongly contaminated
2–3	3	Moderately to strongly contaminated
1–2	2	Moderately contaminated
0–1	1	Uncontaminated to moderately contaminated
0<	0	Uncontaminated

Enrichment factor (EF)

Enrichment factors (EFs) were measured to estimate the abundance of trace metals in sediment. The enrichment factor (EF) was based on the standardization of a tested element against a reference elements (Müller 1981). The reference elements may be Al (Chatterjee et al. 2007), Fe and Ti (Zhang et al. 2007), Mn (Liu et al. 2005) or Li, Sc and Zr (Blaser et al. 2000). Iron was chosen as a reference element because of natural sources (1.5% vastly dominate its input (Tippie 1984). Many authors used iron to normalize heavy metal contaminants (Szefer et al. 1998; Schiff and Weisberg 1999; Baptista Neto et al. 2000; Mucha et al. 2003; Goher et al. 2014; Kalpana et al. 2016). It was calculated according to the following formula:

$$EF = \frac{(M/Fe) \text{ sample}}{(M/Fe) \text{ background}}$$

where EF is the enrichment factor, (M/Fe) sample is the ratio of metal to Fe concentration of the sample, and (M/Fe) background is the ratio of metals to Fe concentration of a background. The EF values close to unity indicate crusted origin; those less than 1.0 suggest a possible mobilization or depletion of metals, whereas EF > 1.0 indicates that the element is of anthropogenic origin (Zsefer et al. 1996). Five contamination categories are recognized and interpreted as suggested by Birth (2003). EF < 1 indicates no enrichment, EF < 3 is minor enrichment, EF = 3–5 is moderate enrichment, EF = 5–10 is moderately severe enrichment, EF = 10–25 is severe enrichment, EF = 25–50 is very severe enrichment, and EF > 50 is extremely severe enrichment, as shown in Fig. 5.

Contamination factor (CF)

The level of contamination of lake sediment or a sub-basin by giving toxic substance (metals) suggested by Hakanson (1980) is often expressed in terms of a contamination factor and is calculated as follows.

$$CF = C \text{ metal} / C \text{ background value}$$

where C metal sample is the concentration of a given metal in lake sediment and Cm background is the value of the metal equal to the world surface rock average given by Taylor (1964).

Pollution load index

Pollution load index (PLI) was used to evaluate the extent of pollution by heavy metals in the environment. The range and class are same as I_{geo} . PLI for a particular site has been

calculated following the method planned by Tomlinson et al. (1980) as follows:

$$PLI = (CF_1 \times CF_2 \times CF_3 \times \dots \times CF_n)^{1/n}$$

where n is the number of metals and CF is the contamination factor.

The PLI value of >1 is polluted, whereas <1 indicates no pollution (Harikumar et al. 2009). The world average concentration of Mn (950 µg/g), Cr (100 µg/g), Cu (55 µg/g), Ni (75 µg/g), Co (25 µg/g), Pb (12.5 µg/g) and Zn (70 µg/g) reported for crustal average (Taylor 1964) was considered as the background value.

Statistical analyses

Descriptive statistical analysis was carried out to know the mean, maximum and minimum values. Additionally, principal component analysis (PCA) is the most common multivariate statistical method used in environmental studies and is employed to extract a small number of latent factors for analyzing relationships among the observed variables (Caeiro et al. 2005). Pearson correlation analysis and PCA were used to find the association among metals and the sources of the metals, respectively, in the surface sediments of the study area. The PCA was carried out on the metal data of all surface sediments for source identification using statistical package for social science (SPSS) version 20. The factor analysis was also performed to understand the variability among the different parameters using varimax normalized values.

Results and discussion

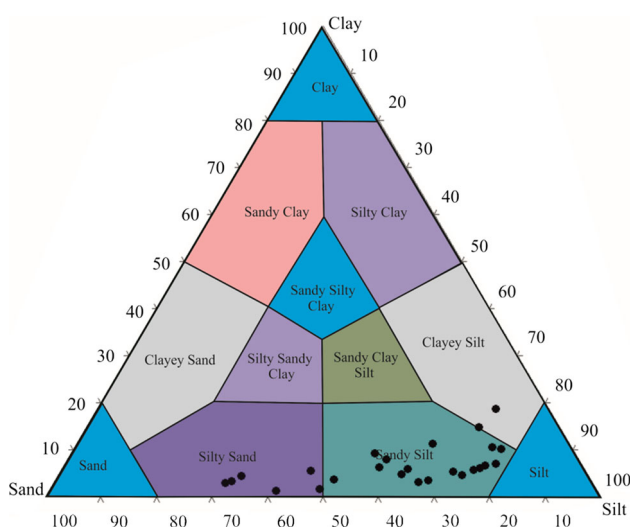
The textural parameter (sand and mud), organic matter and carbonate contents and TTM (Fe, Mn, Cr, Cu, Ni, Co, Pb and Zn) concentrations are given in Table 2.

Sediment characteristics

Sand content varied from 66.14 to 9.58% with an overall average of 32.97%. Likewise, the mud (silt + clay) in the sediments varied from 90.42 to 33.86% with an average of 67.03%. Particle size distribution of lake sediments indicates that Yercaud Lake is rich in (silt + clay) mud. Closely, all sand content is deposited in the shallow regions parallel to the shorelines that are characterized by the high sand content and also showing gradational difference laterally from coarse fractions near the margin to finer fractions more toward the deeper part of the lake. Most of the clay and silt has been transported to the deepest parts of the

Table 2 Sediment characteristics, organic matter, calcium carbonate, C/N ratio and trace element of surface sediments from Yercaud Lake

S. no.	Sand%	Mud%	OM%	CaCO ₃ %	C/N ratio	Fe (μg/g ⁻¹)	Mn (μg/g ⁻¹)	Cr (μg/g ⁻¹)	Cu (μg/g ⁻¹)	Pb (μg/g ⁻¹)	Zn (μg/g ⁻¹)	Co (μg/g ⁻¹)	Ni (μg/g ⁻¹)	PLI
1	33.45	66.55	6.6	2.7	10.5	117,056	390	322.0	575.3	37.4	178.3	113.7	146.8	2.66
2	22.72	77.28	7.7	1.9	9.1	115,273	406	377.5	560.0	35.4	204.5	123.0	146.9	2.79
3	20.02	79.98	8.7	1.8	10.6	109,836	356	386.0	676.0	48.0	258.4	120.6	147.2	3.04
4	23.89	76.11	8.5	2.1	10.3	107,964	379	388.0	552.0	44.9	214.6	119.4	146.8	2.87
5	15.43	84.57	8.6	1.5	8.8	114,400	374	415.0	531.3	36.9	182.3	115.0	146.9	2.72
6	34.68	65.32	5.0	2.8	9.6	109,300	374	351.0	542.0	29.0	128.0	108.8	146.7	2.42
7	12.89	87.11	6.9	1.4	8.8	119,824	396	397.0	640.0	37.9	192.3	114.1	147.0	2.82
8	36.09	63.91	4.7	3.2	10.4	105,500	386	409.0	531.0	20.7	101.4	100.0	146.9	2.26
9	14.58	85.42	6.7	1.1	7.6	111,174	403	393.0	555.0	27.5	183.9	110.9	146.9	2.62
10	14.31	85.69	6.6	0.9	8.7	119,472	413	393.0	658.0	39.3	155.9	111.2	147.2	2.77
11	24.65	75.35	5.9	1.7	8.5	113,200	389	441.2	663.1	31.6	145.7	113.2	147.2	2.69
12	29.35	70.65	7.8	1.9	7.5	105,500	351	397.0	576.6	25.9	177.5	98.7	147.1	2.51
13	57.79	42.21	5.1	5.8	7.4	88,000	350	376.0	520.0	24.2	157.0	102.7	146.9	2.40
14	62.49	37.51	3.7	7.1	8.2	92,700	334	364.0	500.0	15.5	150.5	96.0	146.7	2.18
15	31.28	68.72	6.9	2.9	7.2	106,800	342	379.4	626.0	38.0	175.9	120.0	147.2	2.73
16	9.58	90.42	7.7	0.5	7.1	120,987	423	404.0	614.0	29.4	197.6	102.7	147.1	2.71
17	18.66	81.34	7.6	1.6	7.3	112,500	366	440.0	687.0	39.1	205.3	110.1	147.2	2.89
18	46.12	53.88	5.8	3.9	7.2	96,500	365	367.5	516.0	19.9	169.0	91.1	146.6	2.32
19	36.81	63.19	6.5	2.5	10.4	116,000	398	379.7	480.0	20.5	200.0	130.7	147.1	2.53
20	49.79	50.21	5.5	4.2	10.5	97,100	334	380.0	523.0	27.5	178.0	108.0	146.7	2.49
21	66.14	33.86	3.0	8.1	8.9	97,362	337	352.0	489.0	21.8	109.5	92.7	146.7	2.16
22	17.54	82.46	7.3	1.4	10.2	107,925	369	403.0	619.7	28.0	195.3	116.8	147.2	2.69
23	49.39	50.61	4.0	4.2	10.4	102,897	340	360.0	604.0	25.8	106.8	101.6	146.8	2.31
24	64.83	35.17	3.2	7.3	8.2	84,400	327	336.0	537.0	23.8	101.4	88.0	146.7	2.15
25	31.81	68.19	6.7	2.8	10.2	113,200	389	409.0	594.0	22.5	152.9	100.0	147.2	2.47
Min	9.58	33.86	3.0	0.5	7.1	84,400	327	322.0	480.0	15.5	101.4	88.0	146.6	2.15
Max	66.14	90.42	8.7	8.1	10.6	120,987	423	441.2	687.0	48.0	258.4	130.7	147.2	3.03
Avg	32.97	67.03	6.3	3.0	8.9	107,395	372	384.8	574.8	30.0	168.9	108.4	146.9	2.57

**Fig. 2** Ternary plot showing relative percentage of sand, silt and clay fractions of the Yercaud Lake surface sediments

lake. The textural classification of the Yercaud Lake surface sediments shows a predominant occurrence of sandy silt and silty sand following Shepard's (1954) trilinear diagram (Fig. 2). The spatial diagram shows decreasing sand (feldspar + quartz) content from the margins and is negatively correlated with the increasing clay fraction at the deeper part of the lake (Fig. 3).

The OM content varied from 8.7 to 3.0% with an average of 6.3%. The concentrations of organic matter in the fine-grained fraction of the sediments were often higher than that in the sand-sized fractions (Tam and Wong 2000). This indicates that the lake sediments receive high organic matter from soil erosion and tourist activities (Purushothaman et al. 2011). The C/N ratio is an indicator of changes in the source of organic matter. In general, the C/N ratios from aquatic plants (freshwater phytoplankton) are <10. A higher C/N ratio (10–20) indicates a mixture of aquatic and terrestrial organic material (Mackie et al. 2005;

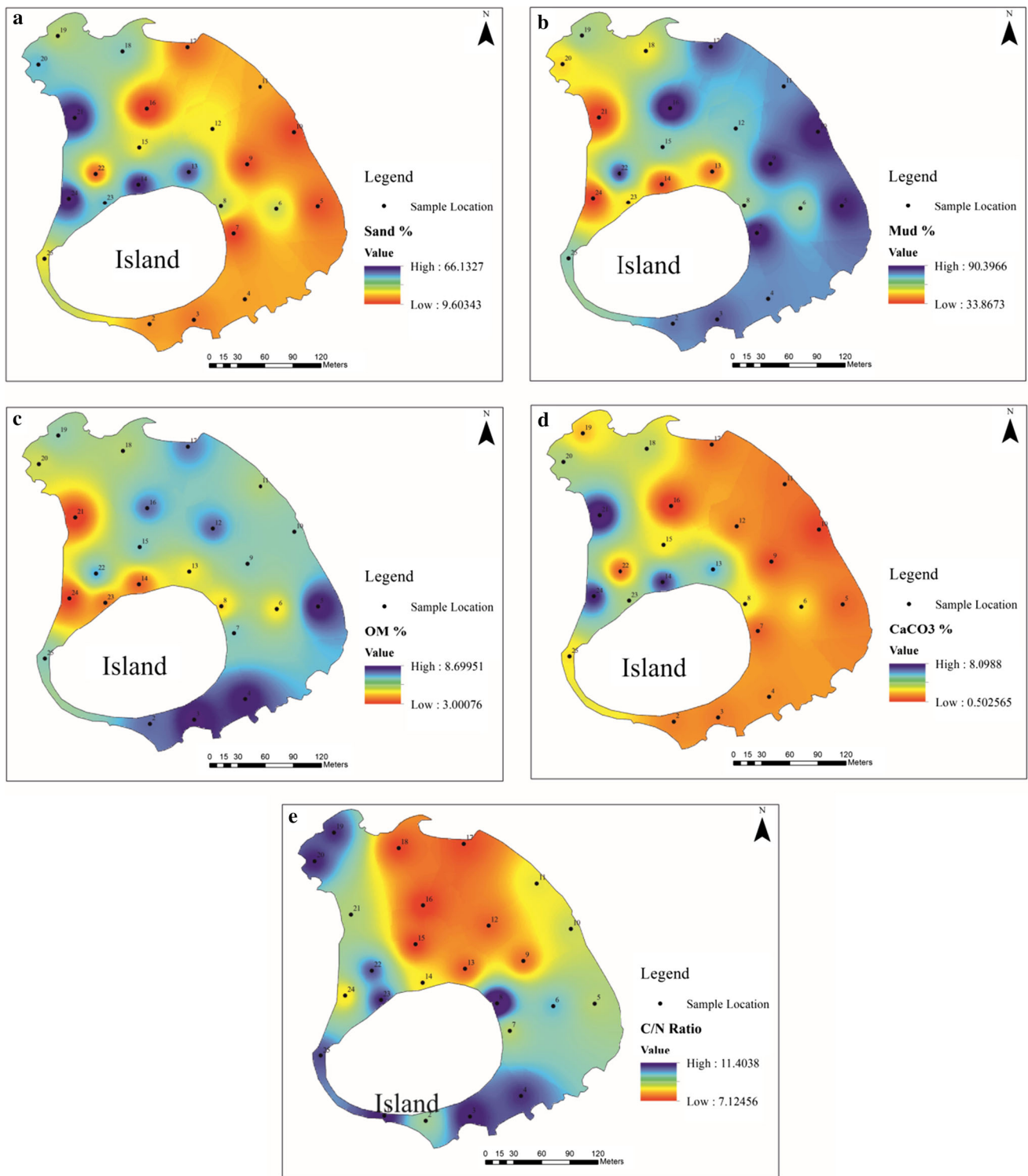


Fig. 3 Spatial distributions of **a** sand, **b** mud (silt + clay), **c** organic matter (OM), **d** calcium carbonate, **e** C/N ratio in the surface sediments from the Yercaud Lake

Zong et al. 2006; Vijayaraj and Achyuthan 2015). C/N ratio of the Yercaud Lake sediments range from 10.6 to 7.1 with an average of 8.9. Most of the C/N values are <10, indicating that the organic matter is from the in-lake algal

production (Meyers and Ishiwatari 1993; Wang et al. 2014a). However, some samples show values higher than 10, suggesting mixed sources of OM (Meyers 1994; Hedges 2002; Sollins et al. 1984). Carbonate content is low

(0.5–8.1%) with an average of 3.0% and is related to the phytoplankton blooms associated with annual lake water mixing and nutrient availability (Lamb et al. 2002a, b).

Metal concentrations in the surface sediments

The overall concentration ranges and mean values of the selected metals in the surface sediments were found as follows: Fe, 84,400–120,987 (107,395); Mn, 327–423 (372); Cr, 322–441.2 (384.8); Cu, 480–687 (574); Pb, 15.5–48.0 (30.0); Zn 101.4–258.4 (168.9); Co, 88–130.7 (108.4); and Ni, 146.6–147.2 (146.9) $\mu\text{g/g}^{-1}$. The metal concentrations in the surface sediments were ranked in decreasing order as follows: Fe > Cu > Cr > Mn > Zn > Ni > Co > Pb (sediments on the surface have significantly high Cu, Cr and Ni concentrations) (Fig. 4a–h). In the present study, nickel and chromium was observed to be abnormally rich, probably leached from the adjoining rock exposures containing large amounts of olivine, pyroxenes, chromite, spinels, etc. All these minerals contain Ni and Cr in appreciable amounts. Also in the case of very strongly and deeply weathered soils of the tropical areas with a humid climate, many elements are lixiviated (e.g., Mg, Ca, Si), but others accumulate in situ (e.g., Cr, Ni, Cu, Mn) and combine with the diverse iron oxide forms (e.g., Nalovic and Quantin 1972; Anand and Gilkes 1987; Becquer et al. 1995; Trolard et al. 1995). Similar observations were made on the basalts and basanites of the French Massif Central (Soubrand-Colin et al. 2007) or of the Réunion Island (Doelsch et al. 2006) and the serpentinites in the Swiss Alps (Gasser et al. 1995).

Ecological risk assessment

The I_{geo} value <0 indicates uncontaminated, while the I_{geo} value >5 represents the upper limit of the contamination. The highest class 6 (very strong contamination) reflects 100-fold enrichment of the metals relative to their background values (Harikumar and Jisha 2010; Goher et al. 2014). The calculated index of geoaccumulation (I_{geo}) of the investigated trace metals in the sediments of the Yercaud Lake and its corresponding contamination intensity is illustrated in Fig. 5. In the present study, I_{geo} values for Mn were found lower than 0, indicating almost unpolluted; Fe, Pb and Zn indicated unpolluted to moderately polluted; Cr and Co indicated moderately polluted; and Cu indicated moderately to strongly polluted sediment quality. Fe and Ni are indicating unpolluted to moderately pollution at all sites; Pb is moderately polluted at site S-1, 3, 4, 7, 10, 15 and 17; and Cu exposed heavy pollution at site S-3, 10, 11 and 17. Overall, the studied sediments were polluted by Cu, Cr and Co,

indicating that these metals might be included by anthropogenic inputs in the lake sediments (Iqbal et al. 2016). The main sources of Cu, Cr and Co in the study area is owing to the discharge of industrial and municipal wastewaters, agrochemicals, landfill leachates and geogenic material. In contrast, mafic rock erosion is the dominant Cr source of lake sediments (DVWK 1998; Wantzen et al. 2008).

The EF values of all studied metals were calculated to evaluate anthropogenic influences on trace metals in the surface sediments of the study area. The EF reflecting metal accumulation for surface sediments is shown in Fig. 3. The enrichment values <1 indicate that the metal is completely derived from geological origin, but EF values >1 propose that the metal is possibly sourced through anthropogenic activities. In the present prediction, the enrichment factor values of the high-altitude Yercaud Lake indicated that Fe indicates <1, showing no enrichment; Cr, Pb, Zn, Co and Ni were between 1 and 3, indicating a minor enrichment, and Cu was between 5 and 10, revealing moderately severe to severe enrichment at all locations. Cr, Pb, Zn, Co and Ni revealed some enrichment at all sites, but they were considered to be contributed by mixed sources. No enrichment of Mn occurred at all sites, in spite of good water circulation and aeration of bottom lake waters during five months of the year. This is the case, for example, no enrichment of Fe and Mn occurs in the Lake Valencia sediments (Bifano and Mogollbn 1995).

Another method used to determine contamination levels was the contamination factor (CF), which is the ratio of metal concentration to the background metal concentration of a given metal, which could be classified into four sets for monitoring the pollution of one single metal over a period of time (Turekian and Wedepohl 1961; Kükroer et al. 2015; Han et al. 2015). It refers to the low degree (CF < 1), moderate degree ($1 \leq \text{CF} < 3$), considerable degree ($3 \leq \text{CF} < 6$) and very high degree (CF ≥ 6). On this basis, the high-altitude Yercaud Lake has very high Cf values for Cu; considerable Cf for Cr, Co; and moderate Cf for Fe, Pb, Zn and Ni except Mn, which is found to be low contaminated. Calculation of the average contamination factor (Cu) indicates humid substances have high affinity with copper, resulting in the formation of stable complexes (Dahrazma and Mulligan 2007). To determine the environmental quality of the sediments, pollution load index (PLI) was used (Suresh et al. 2011; Kükroer et al. 2015). The PLI value 1 represents the baseline for sediment, and values greater than 1 indicate higher contamination. In this respect, as is clear from the PLI values in Table 1, the PLI levels ranged from 2.15 to 3.03 with an average of 2.56. The PLI value was found higher for all sites. It is possible to state that in all stations, the contamination appears to be higher in the surface sediments of the lake.

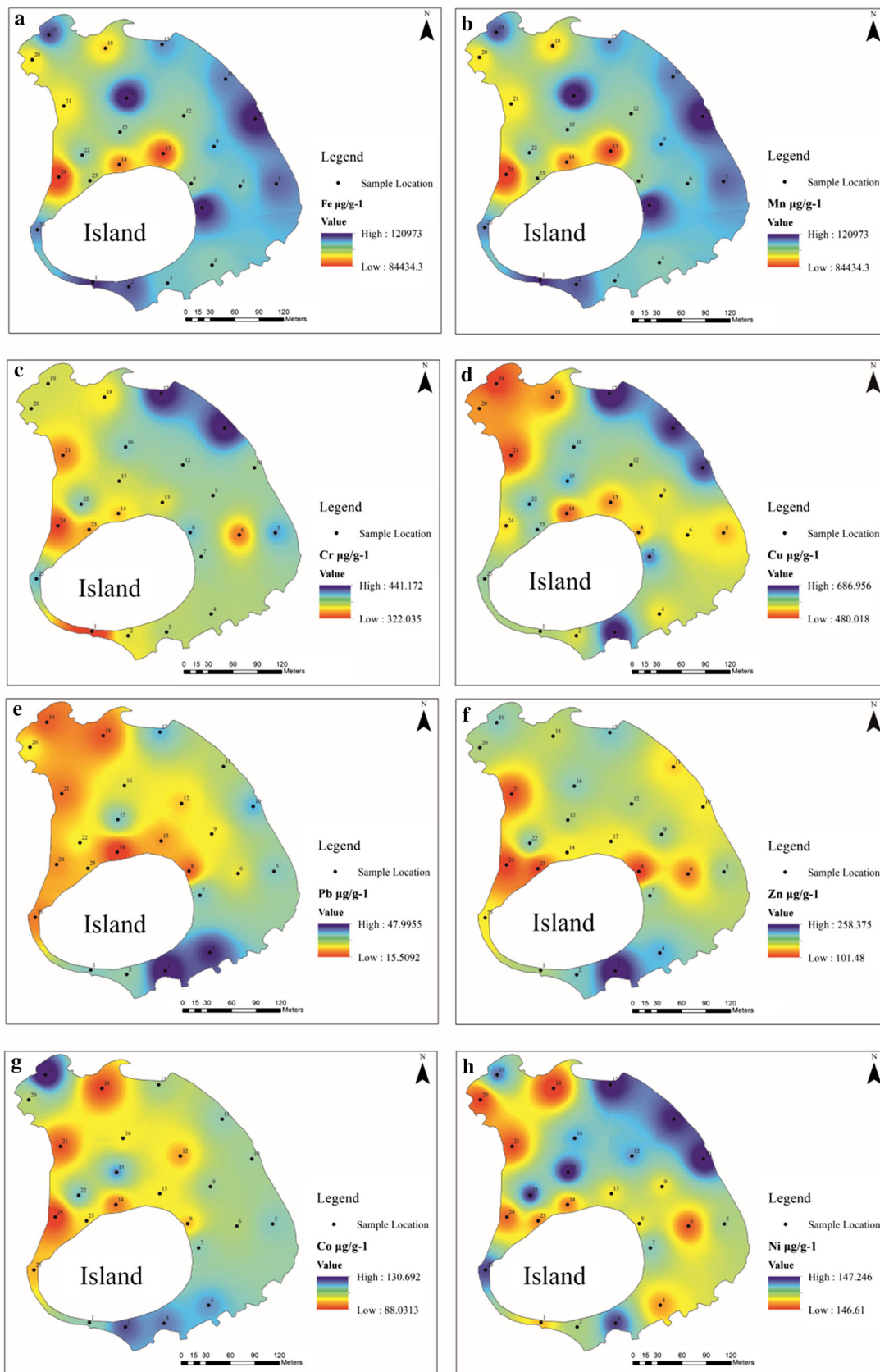


Fig. 4 Spatial distributions of **a** Fe, **b** Mn, **c** Cr, **d** Cu, **e** Pb, **f** Zn, **g** Co and **h** Ni in the floor sediments of the Yercaud Lake

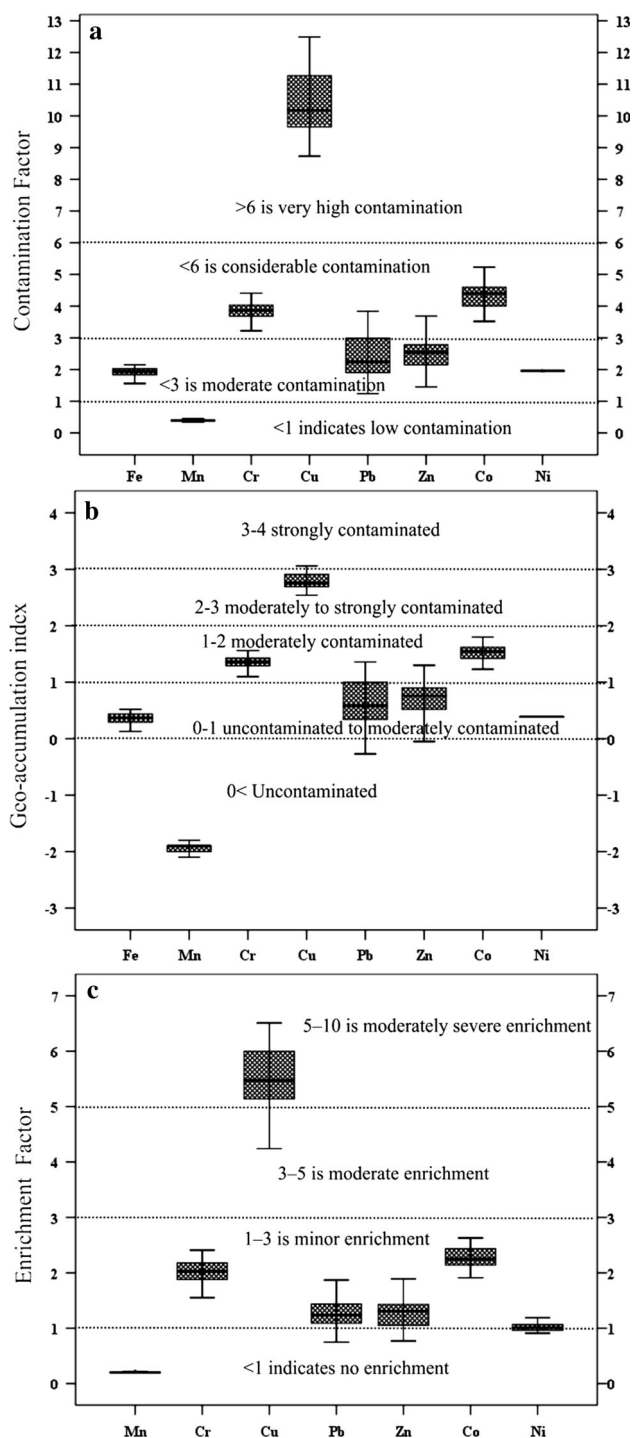


Fig. 5 Box and whisker diagrams of **a** I_{geo} , **b** EF and **c** CF values of the Yercaud Lake floor sediments

Statistical analysis

Correlation matrix

The Pearson correlation coefficients of inter-element relationships provide valuable information on the sources in

the geoenvironment (Usha et al. 2014; Gopal et al. 2016a, b). Correlation matrix was suitable to confirm some new associations between heavy metals. It was generated in the present study using SPSS. In order to evaluate the relationship between textural and chemical parameters (TTMs), correlation matrix analysis was carried out for the entire data set (Table 3). The correlation of the surface sediments shows that all the trace metals are negatively correlated with sand and CaCO_3 . Moreover, trace metals have a high positive correlation with OM and mud fractions. The inter-element relationship between elements specifies a clear comparison with mud vs OM ($r^2 = 0.84$), Fe ($r^2 = 0.86$), Mn ($r^2 = 0.74$), Cr ($r^2 = 0.61$), Cu ($r^2 = 0.64$), Pb ($r^2 = 0.64$), Zn ($r^2 = 0.62$), Co ($r^2 = 0.61$) and Ni ($r^2 = 0.65$). Positive correlation of OM vs Fe ($r^2 = 0.77$), Mn ($r^2 = 0.50$), Cr ($r^2 = 0.51$), Pb ($r^2 = 0.70$), Zn ($r^2 = 0.86$), Co ($r^2 = 0.66$) and Ni ($r^2 = 0.58$) is also observed in the sediments. Fe exhibits high positive correlation with Mn ($r^2 = 0.83$), whereas moderate correlation is found with other trace elements such as Cu, Pb, Co and Ni ($r^2 = 0.51, 0.52, 0.64$ and 0.58). Cr is positively correlated with Cu and Ni and Cu–Pb, Ni and Zn with Co.

Organic matter was positively correlated with trace metals which reveals that they are attached to the organic-rich clay particles in the sediments and has high fluvic/humic acids which have a high metal-adsorbing capacity (Mester et al. 1998; Hlavay et al. 2004; Kargar et al. 2013). Fe has a very strong correlation with other oxides which have a higher affinity with most elements, especially for trace elements (Stumm and Morgan 1996), and that the organic matter contents are important for controlling factors in the abundance of trace metals (Rubio et al. 2000; kalpana et al. 2016). The strong negative values of carbonates with other TTMs suggest that they act as diluents of the metals in the sediments and are also recent in origin (Aloupi and Angelidis 2001). The table presented that high positive correlation happens between Fe and Mn ($r^2 = 0.83$), Co ($r^2 = 0.96$) and Ni ($r^2 = 0.58$). It means that these metals tend to accumulate together. The significant positive correlation with Fe indicates that the metals were derived from similar sources and also moving together (Bhuiyan et al. 2009). Though strong positive correlations among Fe and Mn indicate that these two metals are the constituent of sediments, and they originate mostly from geogenic origin and not from anthropogenic process (Guo et al. 1983), Mn does not show any significant correlation with other trace metals, indicating its independent variables in the sediments (Selvam et al. 2012). In addition, significant correlation between Cu and Ni and the close association of this element confirm their common sink in the sediments (Fianko et al. 2013).

Table 3 Pearson (*r*) correlation coefficients for sediment texture and trace elemental analyses from Yercaud Lake surface sediments

Parameters	Sand	Mud	OM	CaCO ₃	Fe	Mn	Cr	Cu	Pb	Zn	Co	Ni
Sand	1											
Mud	-1.0**	1										
OM	-.84**	.84**	1									
CaCO ₃	.96**	-.96**	-.84**	1								
Fe	-.86**	.86**	.67**	-.85**	1							
Mn	-.74**	.74**	.50**	-.74**	.83**	1						
Cr	-.61**	.61**	.51**	-.59**	.43*	.37	1					
Cu	-.64**	.64**	.47*	-.60**	.51**	.26	.50*	1				
Pb	-.64**	.64**	.70**	-.58**	.52**	.26	.21	.65**	1			
Zn	-.62**	.62**	.86**	-.61**	.47*	.32	.31	.35	.62**	1		
Co	-.61**	.61**	.66**	-.63**	.64**	.44*	.25	.29	.62**	.67**	1	
Ni	-.65**	.65**	.58**	-.63**	.58**	.39	.67**	.73**	.39*	.43*	.48*	1

Bold numbers mark the high positive correlation

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

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

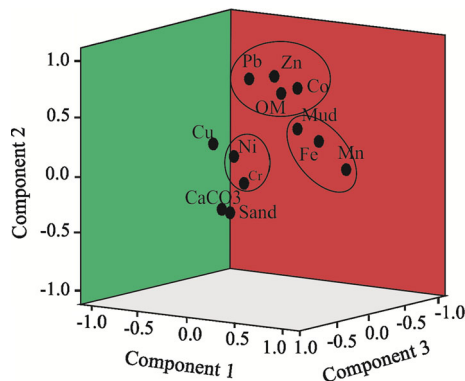


Fig. 6 Rotated loadings showing correlation among trace metals

Source apportionment

PCA was also applied to identify sources of the metals in sediments. The relationships among the metals analyzed based on the three principal components were illustrated in three-dimensional (3D) space (Fig. 6; Table 4). Three principal components (PC) were obtained with eigenvalue greater than 1, explaining more than 84% of total variance. Speciously, the result of PCA resembles well with the correlation coefficients. PC1 explains the 30.15% of the total variance and reveals high loading values for the mud (silt + clay), Fe and Mn (0.69, 0.80 and 0.94, respectively), which are predominantly contributed by the geogenic processes, transportation activities, untreated urban wastes and agricultural runoff. PC2 (29.7% of total variance) is the strong loading of Pb, Zn and Co with organic matter, confirming that metals form organic complexes with humic substances in surface sediments ((Lepane et al. 2007). Those complexes or organic matter in surface

Table 4 Principal component analysis loadings for selected metals

Parameters	PC1	PC2	PC3
Sand	-.69	-.49	-.49
Mud	.69	.49	.49
OM	.44	.74	.34
CaCO ₃	-.72	-.47	-.45
Fe	.80	.37	.30
Mn	.94	.12	.11
Cr	.35	.02	.78
Cu	.08	.35	.83
Pb	.09	.83	.31
Zn	.22	.85	.15
Co	.39	.75	.07
Ni	.27	.26	.80
Eigenvalue %	3.62	3.56	2.97
Variance %	30.15	29.70	24.73
Cumulative %	30.15	59.86	84.58

Bold numbers mark the high correlation coefficients

sediments is often transformed by different biochemical processes (Lepane et al. 2007), and PC3 grouped metals such as Cr, Cu and Ni reveal 24.73% of the total variance. Cr and Cu in PC3 originated from the natural sources such as the catchment bed rock weathering (Lv et al. 2014). Ni and Cr belong to the siderophile elements and are main rock-forming elements, which are derived from terrigenous detritus material transported by surface runoff (Krishna et al. 2011). All three PCA explains 84.58% of the total variance, indicating that the lithogenic factor dominates the distribution of most part of the considered metals in the study. Further, it is observed that Pb and Zn arise due to a

Table 5 Comparison of the investigated trace metals levels ($\mu\text{g/g}^{-1}$) with other international and national studies

S. no.	Lake name	Fe	Mn	Cr	Cu	Pb	Zn	Co	Ni	References
1	Mangla Lake, Pakistan	4959	411	25.0	22.6	30.7	127.3	41.2	70.8	Saleem et al. (2015)
2	Lake Taihu China	–	–	77.4	31.1	33.1	87.3	22.5	29.8	Wei and Wen (2012)
3	Lake Dianchi China	–	–	115.2	90.1	65.8	154.0	33.4	46.0	Wei and Wen (2012)
4	Kodaikanal Lake, India	105,229	48	451.8	54.5	44.7	112.5	1402.3	115.3	Balamurugan et al. (2015)
5	Rewalsar Lake	19,374	387	75.0	42.0	39.4	353.0	–	44.0	Brijraj and Birgit (2003)
6	Lake Vegoritis, Greece	–	–	266.0	36.6	25.8	88.1	25.5	215.8	Skoulikidis et al. (2008)
8	Vembanad Lake in India	54,000	461	107.5	30.9	32.6	184.5	18.6	48.7	Selvam et al. (2012)
9	Lake Nasser, Egypt	12,418	280	30.8	21.8	10.9	35.4	–	27.6	Goher et al. (2014)
10	Maharlu Lake, SW Iran	19,477	554	40.2	38.1	160.3	67.4	145.6	207.0	Moore et al. (2009)
11	Lake Laguna, Philippines	–	–	16.9	103.0	20.0	13.5	–	13.0	Hallare et al. (2005)
12	Geochemical background Value	56,300	950	100.0	55.0	125.0	70.0	25.0	75.0	Taylor (1964)
13	Yercaud high-altitude Lake	107,395	372	384.8	574.8	30.0	168.9	108.4	146.9	This study

confluence of sewage effluents, traffic and boat activities. Probably these elements originated from agricultural activities, sewage sludge, untreated domestic solid wastes, traffic pollution and boating activities (Table 5).

Conclusions

In this study, spatial distribution of eight selected trace metals (Fe, Mn, Cr, Cu, Ni, Co, Pb and Zn) in surface sediments from the higher elevation Yercaud Lake, South India, was examined. Then, potential ecological risk caused by these metals was evaluated by calculating index of geoaccumulation, enrichment factor, contamination factor and pollution load index. Sources were identified with the aid of multivariate statistical analyses such as principal component analysis (PCA). The geogenic and anthropogenic sources could be identified based on multivariate and geostatistical analysis. The measured levels of the studied metals followed the decreasing concentration order: $\text{Fe} > \text{Cu} > \text{Cr} > \text{Mn} > \text{Zn} > \text{Ni} > \text{Co} > \text{Pb}$. High Cu content in the surface sediments, as revealed from the all ecological parameters such as I_{geo} , EF, CF and PLI, could be mainly due to anthropogenic factors along with dispersion or lithogenic influx from the upper catchment. Understanding the ecological risk valuation results, Cu, Cr and Mn were identified as the priority pollutants of concern, though Zn, Ni, Co and Pb could not be ignored as they indicated some enrichment in all the sites. Multivariate statistical techniques (PCA) indicated that Cr and Cu were partially added from anthropogenic sources. However, Cr, Cu, Fe and Mn were mainly derived from the natural erosion and non-point agricultural sources. Pb and Zn were mainly originated from agricultural activities, sewage sludge, untreated domestic solid wastes, traffic pollution and boating activities. Moreover, selected metal

pollution was found relatively higher at all the sites, which receive anthropogenic inputs from the nearby areas such as villages, picnic points and other catchments. These findings provide useful information about sediment quality in the lake. Therefore, the remedial measures are to develop strategies of contamination control and management with the inclusive consideration of the entire area, which is required for aquatic system/human health protection and future restoration of the lake.

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