



# Seasonal and spatial variations in elemental distributions in surface sediments of Chilika Lake in response to change in salinity and grain size distribution

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## Abstract

Chilika lake, the largest brackish water lagoon in Asia, has a unique setting, where the north-eastern part receives freshwater from the terrestrial runoff, and the south-eastern part receives seawater from the Bay of Bengal. The seasonal variability in the quantity of inflowing water and their mixing in the lake controls the mobilization and precipitation of various elements. Seasonal sediment samples were collected from both the river and seawater influenced regions of the lake to understand the spatio-seasonal distributions of various elements along the salinity gradient. The major elements present in the sediments are mostly derived from the parent rock weathering in the source region and subsequently transported into the lake by the rivers. Seasonal variations in trace element concentrations are more prominent in the north-eastern part of the lake (i.e., low salinity region), and their higher concentrations have been observed during the post-rainy period. The affinity of the elements (Al, Fe, Mn, Li, V, Co, Cr, Cu, Th, and Zn) towards fine-grain sediments suggests that the size distribution pattern controls their accumulation, retention, and remobilization. The concentrations of Cr, Cu, and Pb exceeded the effects range low, and effects range median benchmarks indicating the potential biological risk in the low salinity region as compared to the high salinity region. The statistical analysis indicated that the concentrations of elements in the region proximal to the sea mouth are controlled by grain size and physicochemical condition of the lake water. In contrast, the element concentrations in the interior region are associated with anthropogenic activities and weathering processes. Continuous monitoring and assessment of element concentrations of the lake sediments can help to protect the lake ecology from the harmful element contaminations.

**Keywords** Weathering · Anthropogenic disturbances · Element contaminations · Multivariate statistical analysis

## Introduction

Across the globe, the coastal lagoons occupy ~13% of the coastline with their ecosystems increasingly threatened by anthropogenic influences affecting both sediment and water

(Barnes 1980; Kjerfve 1986; McComb et al. 2014; Gokul et al. 2019). The interactions between natural and anthropogenic processes in the estuarine environment make them the most dynamic ecosystems (Barbier et al. 2011; Baustian et al. 2018). The runoff from rivers or drainages from the catchment areas input sediments besides domestic waste, industrial and mining effluent and agricultural runoff (Kjerfve 1986; Cognetti and Maltagliati 2000), which are the major sources of pollutants in addition to the perturbation caused by navigation, fishing, and tourism activities (Halpern et al. 2008; Patel et al. 2018). The nature and rate of rock weathering in the catchment area control the major and trace element concentrations in the influxed water and sediments in the estuaries and lagoons (Angeli et al. 2019). The South Asian rivers supply only 9% of global river water but carry elevated loads of suspended particulate matter (Samanta and Dalai 2018; Singh and Das 2018). The combined effect of weathering and transportation causes remobilization and

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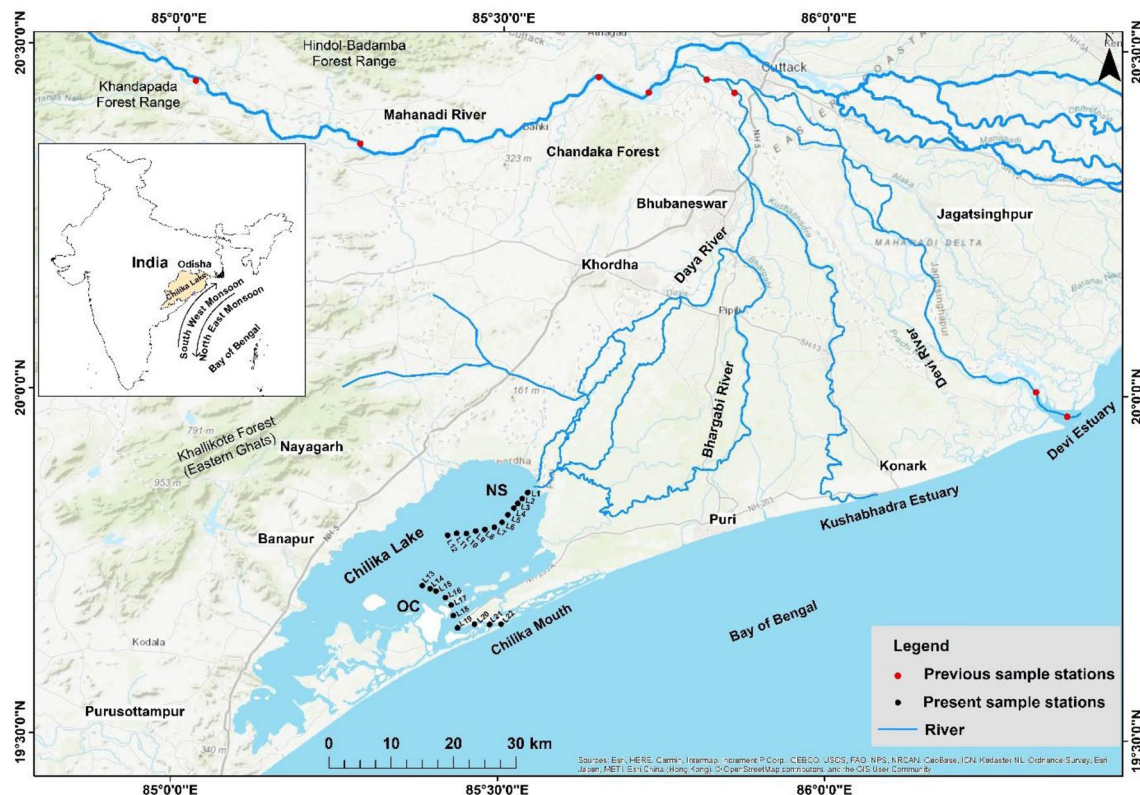
accumulation of trace elements, which become complicated due to various physicochemical processes (Middelburg et al. 1988; Wijsman et al. 2001). Anthropogenic interference exaggerates the element contamination and poses a challenging environment to the biotic community in the brackish/marine ecosystem (Bai et al. 2012; Sarkar 2018), besides the deposition of elements due to natural weathering and transportation into the estuary. Climatic conditions in the catchment area coupled with the weathering intensity, sediment transport processes, and anthropogenic interactions result in significant variations in element concentrations in the estuary (Nath et al. 2000). They may provide environmental indicators to identify the sources of the elements and monitor the contaminants (Bai et al. 2014). Further, the elements have their cycles of removal and reactive element productions due to varying salinity and pH gradients in the estuarine environment (Riba et al. 2004; Karbassi et al. 2014). The monsoon dominated regions witness seasonal variations in freshwater runoff and changes in sediment carrying capacity of rivers leading to significant variations in salinity and pH (Barik et al. 2019) impacting the element concentrations and their retention periods in the estuarine environment (Zwolsman et al. 1993; Sarkar 2018).

This study attempts to assess the seasonal variations in major and trace element distributions in lake floor sediments with response to the change in salinity and pH of bottom water beside their remobilization and retention in the sediments in response to the grain size distribution pattern. The study has also assessed the contamination and ecological risk from elements to the biological community of the lake.

## Materials and methods

### Study region

Chilika is the largest brackish water lagoonal lake of Asia with a mean depth of 1.4 m and a maximum depth of 6 m. Based on the physicochemical parameters and ecological conditions, the lake is divided into four sectors viz. outer channel (OC), central sector (CS), northern sector (NS), and southern sector (SS) (Fig. 1, Kumar et al. 2016). The lake receives around 1.6 million metric tons of sediments with the freshwater from various distributaries of the Mahanadi river in its north-eastern part (NS, Fig. 1, Panigrahi et al. 2009). The lake is connected to the Bay of Bengal through ~ 1.5 km wide channel in the south-eastern side



**Fig. 1** Map of Chilika lake with the catchment of river Mahanadi and its distributaries. The black dots are sampling stations: L1–L12 in northern sector (NS) and L13–L22 in outer channel (OC); red dots

are published sampling locations taken from Sundaray et al. (2011). The base map is the default topographic map of the region in ArcGIS 10.2

(OC, Fig. 1). The rate of sedimentation varies in different sectors of the lake, and the  $^{210}\text{Pb}$  decay rate in sediment cores from NS suggests a sedimentation rate of 0.8 cm/year (Unnikrishnan et al. 2009). The higher sedimentation rate is attributed to the high sediment load in the inflowing water at NS region, which varies significantly from rainy to pre-rainy periods. The deposition of sediments is minimum at OC region due to its dynamic nature and connectivity with the Bay of Bengal.

The lake and its catchment experience dry, sub-humid, and tropical monsoon climate with an average annual minimum temperature of 14 °C during winter (December–January) and maximum temperature of 40 °C during summer (April–May) (Panigrahi et al. 2007). Hydrologically, the season over the lake is divided into three periods viz. rainy (July–September), post-rainy (November–January), and pre-rainy (April–May). During the rainy period, southwest monsoon (SWM) cause more than 75% of precipitation with an average rainfall of 1240 mm in the catchment area (Sahay et al. 2019). This leads to a higher inflow of freshwater and sediments into the lake with water spread area of 1165 sq. km (Das et al. 2016; Kumar et al. 2016; Singh and Das 2018; Sahay et al. 2019). Further, the higher influx of freshwater changes the physicochemical parameters (especially pH and salinity) of the lake, and it behaves as a freshwater ecosystem in NS, which gradually changes to the brackish water ecosystem in OC where it receives seawater (Barik et al. 2019). During post-rainy period, northeast monsoon (NEM) causes minimal precipitation in the catchment area: thus, the river flow becomes sluggish and maintains the base flow. The lake ecosystem during this period changes to brackish water in the river as well as the seawater ends (Barik et al. 2019). During this period, pleasant weather attracts numbers of migratory birds that increases the tourism activities in the lake. During pre-rainy period, the lowest precipitation and high evaporation rate significantly reduce the water inflow in NS and is represented as pre-southwest monsoon (PWSM) (Panigrahi et al. 2007; Muduli et al. 2012). It converts the lake into a brackish water ecosystem near the river end and marine water ecosystem towards the sea end (Barik et al. 2019). The water spread area of the lake is reduced to 906 sq. km during pre-rainy period (Kumar et al. 2016; Sahay et al. 2019). In an aquatic system, the mobility and solubility of elements largely depend upon the chemical characteristics of the water (Du Laing et al. 2008; Acosta et al. 2011; Lim et al. 2012). The significant seasonal changes in water characteristics can modulate the element concentrations in the lake sediments. Besides, the catchment area of the river Mahanadi comprises of various mines (coal and metals), agricultural fields, and domestic farms, which contribute heavy and trace elements into the lake. In the recent past, sediment depositions have considerably narrowed down the connectivity of the lake to the sea, leading to changes in its

salinity and thereby impacting indigenous flora and fauna (Panda et al. 2013; Sahu et al. 2014; Barik et al. 2019). The combined effect of freshwater influx from the north-eastern (NS) and the seawater from the south-eastern (OC) creates spatial and temporal salinity gradients in the lake viz. fresh, brackish, and saline (Barik et al. 2019). The present study is focussed along the salinity gradient from NS to OC, i.e., from low to high salinity region.

## Sample collection

Lake floor sediments (surface) were collected using Ekman box corer from 22 fixed stations, starting near the river mouth and ending towards the sea mouth. The samples were collected during September (2016) representing rainy period denoted as SWM, January (2017) representing post-rainy period denoted as NEM, and from eighteen stations during May (2017) representing pre-rainy period denoted as PSWM (Fig. 1). Four interior stations of NS (L8–L12) were not approachable during PSWM due to very low water depth and high growth of the invasive weed species in the lake. The sampling stations were geo-referenced using a handheld global positioning system (E-trex 20). The top undisturbed surface sediments (< 1 cm) were scooped carefully from the box corer and collected in zip-lock bags and preserved at ~4 °C temperature. Lake bottom water was collected using Niskin water bottle and immediately measured for electrical conductivity (EC) and pH in the field using multiparameter system (Thermo Fisher Scientific, Orion Star A329). Grain size analysis of the sediment samples was carried out in the laboratory using particle size analyzer (Horiba, LA-950V2) after removing carbonate and organic matter by treating the sediments with 10% HCl for 2 h and 30% H<sub>2</sub>O<sub>2</sub> for 24 h, respectively (Barik et al. 2019). The presence of live stained foraminifera in the collected sediment samples suggest that the sediments represent the monitoring period (Barik et al. 2019).

## Geochemical analysis

Finely powdered samples (0.3 g) and Certified Reference Material-HISS 1 (CRM) were digested in Microwave digestion system (Multiwave Pro, Anton Paar) using ultrapure acids in a ratio of HNO<sub>3</sub>:HCl:HF: 1:2:3. A blank was included in each digestion. The filtered (0.45 µm polycarbonate membrane filter) samples were analyzed for Al, Ca, Fe, K, Mg, Mn, Na, Li, Ba, V, Co, Cr, Cu, Pb, Sr, Th, Zn, and Mo concentrations using inductively coupled plasma—optical emission spectroscopy (ICP-OES, Agilent-5110) after neutralizing HF with 10 ml of boric acid (5%) solution. The digested blank was analyzed after every seven samples

and CRM after every 14 samples. The data accuracy was cross-checked with CRM, and the error percentage for the entire analysis was found to be within 10%.

### Clay mineral analysis

Based on the grain size analysis, it was found that clay minerals are concentrated in NS. Six samples from alternate stations of the low salinity region (NS) were processed by adding 5 ml acetic acid and 1 ml hydrogen peroxide for removal of carbonate and organic matter for clay mineral analysis. Based on the Stokes' law, clay content was separated by treating the samples with 10% sodium hexametaphosphate solution, and oriented slides were prepared (Folk 1980). X-ray diffraction (Bruker, D8) patterns of the glycolated oriented slides were obtained with  $2\theta$  ranging from  $4^\circ$  to  $30^\circ$ . The individual peak of clay mineral was identified and quantified from their peak areas by semi-quantitative method (Chiperă and Bish 2001).

### Assessment of sediment quality

The element concentrations were compared with the sediment quality guidelines (SQGs) (McCauley et al. 2000). Effects range low (ERL), effects range median (ERM), threshold effects level (TRL), and probable effects level (PEL) are commonly used to evaluate the sediment quality and its effect on the biological community (Macdonald et al. 1996; Wenning 2005). The elements having concentrations below ERL and TEL values suggest rare or occasional adverse biological effects on the biota, while above ERM and PEL suggest frequent adverse effects on biota (Zahra et al. 2014; Sarkar 2018). Contamination indices for various elements were also calculated to quantify and evaluate the contamination of sediments.

### Contamination factor

Contamination factor (CF), defined by the ratio of the concentration of an element with its background value, was calculated (Savvides et al. 1995; Pekey et al. 2004) for each station to evaluate the sediment contamination levels. In this study, the background concentration values are the average shale values obtained by Turekian and Wedepohl (1961). The CF is expressed as:

$$CF = \frac{C_n}{B_n},$$

where  $C_n$  is the concentration of the element ( $n$ ) in the sediment, and  $B_n$  is the geochemical background of that element

( $n$ ). Based on the CF, contamination levels can be classified as low (CF:  $< 1$ ), moderate (CF: 1–3), considerable (CF: 3–6), and very high (CF:  $> 6$ ) (Hakanson 1980; Savvides et al. 1995; Pekey et al. 2004; Chakraborty et al. 2014).

### Geoaccumulation index

The quality of sediment in terms of element contamination was assessed by calculating geoaccumulation index ( $I_{geo}$ ), which helps in assessing the degree of element contamination with respect to their background values (Sarkar 2018). It is calculated as:

$$I_{geo} = \log_2 C_n / (1.5 \times B_n).$$

Factor 1.5 is used to minimize the background matrix effect due to lithospheric effects. There are 7 classes of element contamination on the basis of  $I_{geo}$  values such as Class 0 (uncontaminated:  $I_{geo} < 0$ ), Class 1 (uncontaminated to moderately contaminated:  $0 \leq I_{geo} < 1$ ), Class 2 (moderately contaminated:  $1 \leq I_{geo} < 2$ ), Class 3 (moderate to highly contaminated:  $2 \leq I_{geo} < 3$ ), Class 4 (highly contaminated:  $3 \leq I_{geo} < 4$ ), Class 5 (high to extremely contaminated:  $4 \leq I_{geo} < 5$ ), and Class 6 (extremely contaminated:  $5 \leq I_{geo}$ ) (Müller 1969).

### Enrichment factor

Enrichment factor (EF) is used as a contamination index to determine the degree of anthropogenic element contamination in sediments based on the element enrichments (Sakan et al. 2009) by standardization of the measured element against a reference element. In the present study, iron (Fe) is taken from Turekian and Wedepohl (1961) and is used as the reference element for standardization because (1) Fe is associated with fine-solid surfaces, (2) its geochemistry is similar to that of many trace elements, and (3) its natural concentration tends to be uniform (Bhuiyan et al. 2010). It is calculated as:

$$EF = \left( \frac{C_n}{C_{Fe}} \right)_{\text{sediments}} / \left( \frac{B_n}{B_{Fe}} \right)_{\text{reference}},$$

where  $C_{Fe}$  is the concentration of Fe in the sediment and  $B_{Fe}$  is the geochemical background of Fe. For different EF values, the element enrichment can be explained as no enrichment ( $< 1$ ), minor enrichment (1–3), moderate enrichment (3–5), moderately severe enrichment (5–10), severe enrichment (10–25), very severe enrichment (25–50), and extremely severe enrichment ( $> 50$ ) (Sakan et al. 2009).



## Pollution load index

Pollution load index (PLI) is calculated to evaluate the element contamination in sediments for each element and station separately (Angulo 1996; Ganugapenta et al. 2018). It is expressed as the  $n$ th root of the product of  $n$  number of contamination factors (Tomlinson et al. 1980).

$$PLI = \sqrt[n]{CF_1 \times CF_2 \times CF_3 \times \dots \times CF_n}$$

where CF is the contamination factor and  $n$  is the number of contamination factors.

As PLI includes all the contamination factors together, it provides an overall level of element contamination in sediments. PLI values higher than 1 indicate the existence of element contamination, while less than 1 indicates no element contamination (Sarkar 2018).

## Statistical analysis

An analysis of variance (one-way ANOVA) was used to observe the seasonal variations in element concentrations with a confidence level of 95% ( $p < 0.05$ ) followed by Tukey's test. For normality assumptions (Shapiro–Wilk and Jarque–Bera tests), Box cox transformation was applied to the data before the ANOVA test (Fletcher et al. 2019; Harguinteguy et al. 2019; Kouassi et al. 2019). Multivariate statistical analysis was executed to observe the relationships of physicochemical parameters of water and grain size distribution with the element concentrations using PAST software (Hammer et al. 2001). For multivariate statistical analysis, log transformation ( $\log(1+x)$ ) was applied to the raw data to manage the scale variability and units of measurement (Sahoo et al. 2015). Values with a significance level of  $\alpha < 0.05$  were considered as significant in Pearson's correlation analysis. Q-mode cluster analysis using paired group method with Euclidean distance as the dissimilarity matrix and R-mode principal component analysis was executed to observe the relationship between the principal components and the element concentrations at various sampling stations.

## Results and discussion

### Physicochemical condition of the lake

The lake water is found to be alkaline in nature (average pH > 7, Table 1). The pH of water plays an important role in element solubility on the lake floor sediments (Jain et al. 2007). The pH is relatively low in the low salinity region of the lake during SWM due to the addition of freshwater from the Mahanadi distributaries and nearby catchment area. This low pH region has high nutrients and bacterial abundances,

which accelerate the element retention in sediments (Muduli et al. 2012; Zahra et al. 2014). The lake experiences huge seasonal and spatial variations in salinity, as observed from the EC values. The average EC value varies between 0.27 ms/cm during SWM and 5.64 ms/cm during NEM in regions proximal to the river mouth (NS), which increases towards the sea mouth (OC) and ranges between 10.97 ms/cm during SWM and 51.83 ms/cm during PSWM (Table 1). Thus, the sediments in these two regions are under the influence of variable water characteristics (from fresh to saline) with the change in season.

The lake sediments are dominated by silt size fraction near the river mouth, sand-size fraction near the sea mouth, and silt to sand-size fraction with small amount of clay in the interior parts of the lake away from the river and sea mouths (Barik et al. 2019). Grain size distribution has clearly explained prevailing of high energy condition towards the sea mouth, low energy condition towards the river mouth, and medium energy condition in the interior part of the lake (Barik et al. 2019). Pearson's correlation analysis suggests that EC (strong negative correlation) plays a dominant role in element distribution in comparison to pH (Appendix A). Fine-grain sediments (silt and clay) also support accumulation and retention of elements on the lake floor sediments.

### Seasonal distribution of elements and their implications

Out of the analyzed 18 elements, seven elements (Al, Ca, Fe, K, Mg, Na, and Mn) have very high concentrations in the lake sediments and are considered as major elements. The remaining 11 elements (Ba, Co, Cr, Cu, Li, Mo, Pb, Sr, Th, V, and Zn) are considered as trace elements. The statistical summary of the major and trace element concentrations with their  $p$  values of ANOVA test are listed in Tables 1 and 2, respectively. The concentrations of major elements show limited seasonal variations when compared with the trace elements in both low (NS) and high (OC) salinity regions since they are present in much higher concentrations (Tables 1 and 2). Lower  $p$  values ( $< 0.05$ ) of ANOVA test suggest significant seasonal variations for Al, Fe, Co, Cu, Cr, Li, Mo, Pb, Th, V, and Zn in NS, and Co, K, Mo, and Pb in OC region. The seasonal variations in NS are attributed to seasonal changes in sediment supply and water influx from the distributaries of the river Mahanadi, which is the second-largest contributor of sediment influx ( $12 \pm 5 \times 10^6$  tons/year) into the Bay of Bengal among the peninsular rivers in India (Bastia and Equeenuddin 2016; Kumar et al. 2016). Bastami et al. (2015) suggested that sediment and energy distribution patterns are the important factors that influence the element concentrations in the estuarine lake. The fine-grain materials have a greater affinity towards the elements due to their higher specific surface

**Table 1** Statistical summary of physicochemical parameters of water (EC, pH), grain size distribution (sand%, silt%, and clay%), and concentrations of major elements in sediments observed during south-west monsoon (SWM), northeast monsoon (NEM), and pre-south-west monsoon (PSWM) from the northern sector and outer channel regions of Chilika lake with ANOVA (*p* values)

Elements	EC	pH	Sand%	Silt%	Clay%	Al	Ca	Fe	K	Mg	Na	Mn
Northern sector												
SWM												
Min	0.15	6.59	33.21	23.55	0.41	33,085	1433	37,275	4279	713	3811	1037
Max	0.46	8.35	73.62	65.65	5.67	213,092	7832	61,884	35,231	10,425	12,907	2999
Mean	0.27	7.52	52.70	44.63	2.67	62,865	2839	55,552	10,000	3065	6319	1850
NEM												
Min	2.66	8.11	26.10	26.99	0.39	29,643	1263	47,038	4627	542	4311	980
Max	10.31	9.24	69.58	72.92	4.29	184,990	8003	57,997	36,477	8506	13,009	2855
Mean	5.64	8.64	53.30	44.60	2.10	70,114	3107	52,373	11,639	3018	7612	1700
PSWM												
Min	2.18	7.26	33.05	45.08	0.44	27,662	1455	42,802	4579	1037	4945	996
Max	12.64	8.91	54.39	64.72	3.57	38,824	4348	53,837	5477	1665	12,595	3607
Mean	4.64	7.89	44.48	53.99	1.53	33,646	2507	48,516	4988	1317	7015	2323
ANOVA ( <i>p</i> )	<b>0.00</b>	<b>0.00</b>	–	–	–	<b>0.05</b>	0.99	<b>0.01</b>	0.15	0.28	0.6	0.3
Outer channel												
SWM												
Min	8.25	6.57	63.57	0.00	0.00	19,326	240	6062	4484	526	3817	119
Max	14.42	7.36	100.00	34.99	1.74	220,695	12,605	64,538	53,965	11,035	13,601	1552
Mean	10.97	7.11	85.23	14.29	0.48	58,043	3947	31,701	12,460	2815	7126	608
NEM												
Min	13.81	8.50	53.83	0.00	0.00	17,367	192	4683	3179	625	4727	95
Max	43.65	9.09	100.01	42.39	4.98	203,901	5377	67,139	36,918	9911	13,875	1309
Mean	29.68	8.74	76.95	21.07	1.99	42,562	2730	33,783	7890	1934	8664	545
PSWM												
Min	50.95	7.90	68.07	0.00	0.00	15,627	60	11,820	3699	372	6137	210
Max	53.39	8.36	100.00	30.75	1.18	34,440	6075	52,762	5975	2102	12,003	816
Mean	51.83	8.25	88.83	10.83	0.35	22,835	2582	29,574	4574	964	9371	400
ANOVA ( <i>p</i> )	<b>0.00</b>	<b>0.00</b>	–	–	–	0.2	0.43	1	<b>0.05</b>	0.3	0.87	0.2

pH is in numerical value, EC in mS/cm, and element concentrations in mg/kg. Values in bold indicate seasonal variation with a significant level of 0.05

area (Singh et al. 1999). From Pearson's correlation matrix (Appendix A), it is observed that most of the elements have a negative correlation with sand% and positive correlations with silt% and clay% in the lake throughout the year. This suggests higher precipitation and retention of elements by fine-grain sediments in medium to low energy condition of the lake.

### Distribution of major elements

Al and Fe are identified as the dominant elements in both low (NS) and high (OC) salinity regions of the lake (Table 1). Both the elements show declining concentrations from low to high salinity regions of the lake (Table 1), which suggests that they are mainly contributed through the river input. Higher Fe and Al concentrations in sediments have been reported due to intense weathering of acidic parent rock

in the catchment region of the river Mahanadi (Zachmann et al. 2009; Govin et al. 2012). Higher concentrations of major elements (Al, Fe, Ca, K, Mg, and Mn) have also been reported in both suspended and river-bed sediments of the river Mahanadi derived from weathering and anthropogenic processes in the catchment (Chakrapani and Subramanian 1993). The river-bed sediments in the low salinity region show more enrichment of Fe and Mn than the Mahanadi river basin (Sundaray et al. 2011). Significant correlations among the major elements (Appendix A) also suggest their common lithogeny origin (parental rock) and similar modes of transportation (Bastami et al. 2015; Ganugapenta et al. 2018).

The major elements are present in very high concentrations in the low salinity region, while limited river influx during PSWM causes a decline in their concentrations (Table 1). A slight deviation in concentrations of some elements from

**Table 2** Statistical summary of trace element concentrations (mg/kg) in sediments observed during southwest monsoon (SWM), northeast monsoon (NEM), and pre-southwest monsoon (PSWM) from the northern sector and outer channel regions of Chilika lake with ANOVA (*p* values)

Elements	Li	Ba	V	Co	Cr	Cu	Pb	Sr	Th	Zn	Mo
Northern sector											
SWM											
Min	41.5	68.8	97.9	18.2	75.6	44.2	29.6	26.8	22.9	71.5	2.2
Max	102.5	1277.2	128.6	42.4	175.0	97.6	107.6	465.3	70.7	179.3	5.4
Mean	72.4	279.0	122.9	26.8	112.7	60.8	52.5	130.3	38.0	110.2	3.4
NEM											
Min	45.8	54.7	117.8	31.2	98.3	62.2	66.4	27.5	23.0	104.8	4.3
Max	142.5	819.6	148.7	41.3	210.4	116.1	102.5	349.8	59.8	225.3	5.5
Mean	86.8	268.8	132.4	36.7	161.8	97.3	84.9	122.6	37.6	177.6	4.8
PSWM											
Min	45.4	72.6	110.0	28.9	119.1	61.0	75.4	30.3	22.3	103.9	4.2
Max	69.0	134.6	122.8	31.8	211.2	81.5	80.3	49.8	26.0	135.0	4.8
Mean	53.6	108.0	116.4	30.2	139.9	70.5	77.5	40.5	23.8	120.4	4.5
ANOVA ( <i>p</i> )	<b>0.01</b>	0.45	<b>0.00</b>	<b>0.01</b>	<b>0.00</b>	<b>0.00</b>	<b>0.00</b>	0.48	<b>0.00</b>	<b>0.00</b>	<b>0.00</b>
Outer channel											
SWM											
Min	11.3	16.8	19.4	12.9	20.7	4.6	22.1	8.0	4.5	24.4	2.2
Max	194.9	945.3	129.9	32.5	232.9	90.7	95.8	462.3	76.1	196.5	6.0
Mean	56.6	341.5	76.3	19.5	77.0	26.9	59.8	109.2	24.7	84.2	4.1
NEM											
Min	9.7	5.0	14.6	16.7	23.0	4.7	60.3	6.9	3.6	24.1	4.5
Max	155.7	569.9	141.9	33.8	219.6	86.7	93.9	239.1	56.2	257.8	5.6
Mean	58.3	244.3	84.4	24.9	105.3	42.0	79.9	67.8	21.1	130.0	5.0
PSWM											
Min	11.4	4.4	28.7	9.1	18.5	3.4	35.5	3.8	5.7	13.7	2.4
Max	77.9	620.3	207.7	21.6	162.2	57.4	80.6	64.8	35.5	121.2	4.9
Mean	36.8	214.7	84.0	16.9	88.2	23.2	56.6	35.7	14.6	66.2	4.0
ANOVA ( <i>p</i> )	0.88	0.47	0.93	<b>0.02</b>	0.64	0.57	<b>0.02</b>	0.13	0.78	0.27	<b>0.03</b>
SQGs											
TEL	–	–	–	–	52.3	18.7	30.2	–	–	124	–
PEL	–	–	–	–	160	108	112	–	–	271	–
ERL	–	–	–	–	81	34	49.7	–	–	150	–
ERM	–	–	–	–	370	270	218	–	–	410	–

Values in bold indicate seasonal variation with a significant level of 0.05

SWM to NEM (Table 1) is attributed to the differential water input from the distributaries of the river Mahanadi (Barik et al. 2017, 2018). Most of the major elements (except for Na) show a clear decreasing trend from SWM to NEM, followed by PSWM in the high salinity region (Table 1). It indicates that the sediment influx during SWM has flushed from low to high salinity region and causes higher concentrations of major elements. During the other two periods, the reduced river inflow allows the ingress of marine water into the high saline region and causes a gradual decrease in major element concentrations (Zahra et al. 2014). The enrichment of Na is controlled by the ingress of seawater (maximum during PSWM), which induces ion exchange between the sediment and water (Rao et al. 2013). Higher concentrations of Mn in PSWM (Table 1) are due to the decomposition of

macrophytes, which leads to a hypoxic condition in the lake with the more flux of Mn in the low salinity region (Rigollet et al. 2004). Remobilization of elements directly depends on the oxic/anoxic condition of the sediment–water interface. The cyclic growth and decomposition of macrophytes affect the oxygen saturation in the Chilika, which plays a significant role in elements remobilization (absorption or precipitation) (Panigrahi et al. 2007; Purushothaman et al. 2012). The bottom-feeding species, especially the benthic organisms, also have a significant role in the elements remobilization (Riedel et al. 1997; Sundaray et al. 2011). The organisms that ingest deposited sediments or suspended particulate matter also ingest the trace elements bounded to the sediments (Gupta et al. 2009; Rezaie-Boroon et al. 2013). The ingested elements are either bio-accumulated or

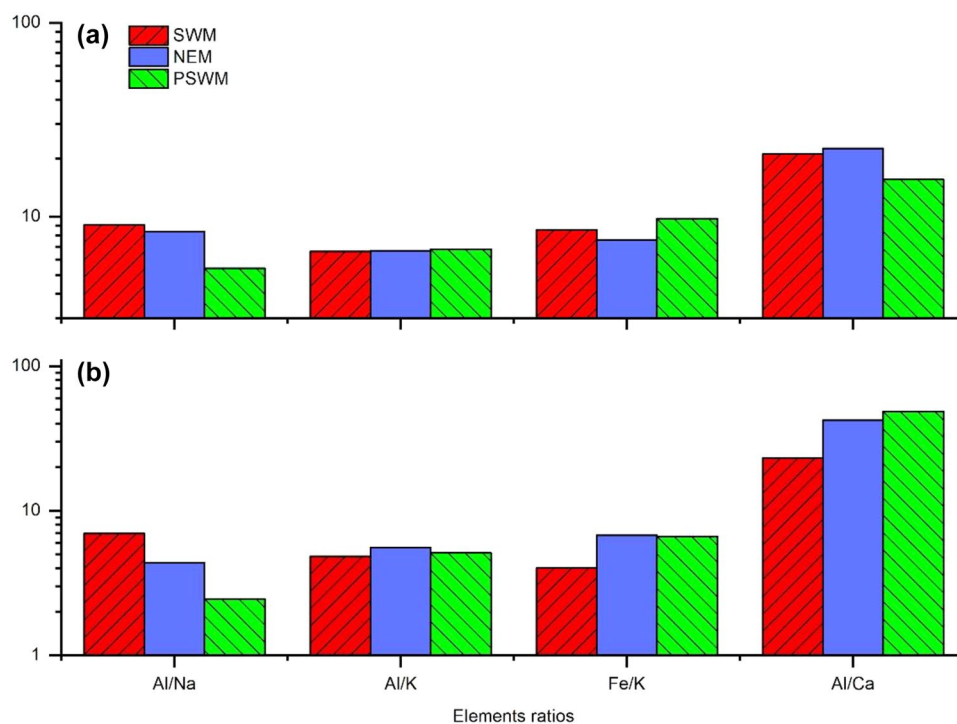
transferred to higher trophic levels via food-chain (Gupta et al. 2009; Sarkar 2018; Harguinteguy et al. 2019). Parida et al. (2017) suggested that the accumulation of trace metals in edible parts of fish was within the prescribed limits in the lake, but the accumulation may increase in higher size fishes. Accumulation of elements viz. K, Ca, Mn, Cu, Zn, Pb was also reported by Mohapatra et al. (2009) in both hard and soft shelled crabs of Chilika lake.

Chemical weathering of bedrock is quite intense in the tropical regions, which affects the concentrations of major elements and causes enrichment of Fe in the weathered products transported by the river water to the sink (Govin et al. 2012; Dutt et al. 2018). The bed sediments of river, lake or any aquatic environment act as an important sink for the elements (Gupta et al. 2009). Al and Fe are conservative towards chemical weathering and become enriched or remain constant in the weathered products in comparison to the parent sediments (Middelburg et al. 1988; Wei et al. 2006). Potassium (K) generally comes from potash-feldspar or illite and is less affected by chemical weathering (Zabel et al. 2001; Govin et al. 2012). On the other hand, Na and Ca get easily removed from the parent sediment and depleted in the weathered product (residue) due to their higher mobility. Thus, their ratios (Al/Na, Al/K, Fe/K, and Al/Ca) are used as proxies to understand the weathering processes. Enrichment of Al or depletion of Na results in higher Al/Na ratio in the estuarine lake sediments, which suggests a higher rate of chemical weathering in the catchment area during SWM (Fig. 2; Wei et al. 2006). The catchment area of the Mahanadi river is mostly dominated by the Eastern Ghats' rocks

comprising of granites, khondalites, gneisses, migmatitic charnockite–leptynite complexes, anorthosites, and ultra-basic rocks along with recent alluvium in the downstream areas (Panigrahy and Raymahashay 2005). Extreme chemical weathering of these rocks results in the enrichment of Al, Fe, and kaolinite in the lake sediments during SWM. The higher Al content in the sediments is indicative of its origin from Al-rich rocks, and higher Fe could be derived from the weathering of ferro-diorite, which is a major rock-type present in the vicinity of Chilika lake (Sarkar et al. 1981; Maji et al. 2010). Hence, Al/Na ratio is the highest during SWM and the lowest during PSWM in both river and marine influence regions, while Al/K and Fe/K ratios do not vary significantly (Fig. 2). Al/Ca ratio has an uneven distribution pattern. Higher values of Al/Na ratio correspond to a higher degree of chemical weathering, which is strongly modulated by climatic factors such as precipitation, humidity, temperature, etc. The abundances of Al and Al/Na ratio in Chilika lake sediments during SWM and their seasonal and spatial variations in the two regions of extreme salinity contrast suggest that they can be used as a potential proxy to reconstruct paleo-monsoon.

Clay mineral characteristics of aquatic sediments are good indicators of climatic conditions, hydrography, geology, and topography of continental source area (Chamley 1989; Thiry 2000), and their variations act as important tools for deciphering the sediment sources and intensity of weathering (Thamban et al. 2002). They also act as carriers of element contaminants by adsorption on sediment surfaces (Ahmed et al. 2010). Kaolinite, an aluminium silicate

**Fig. 2** Seasonal variation in major element concentration ratios in **a** northern sector and **b** outer channel (SWM: southwest monsoon, NEM: northeast monsoon, and PSWM: pre-southwest monsoon)

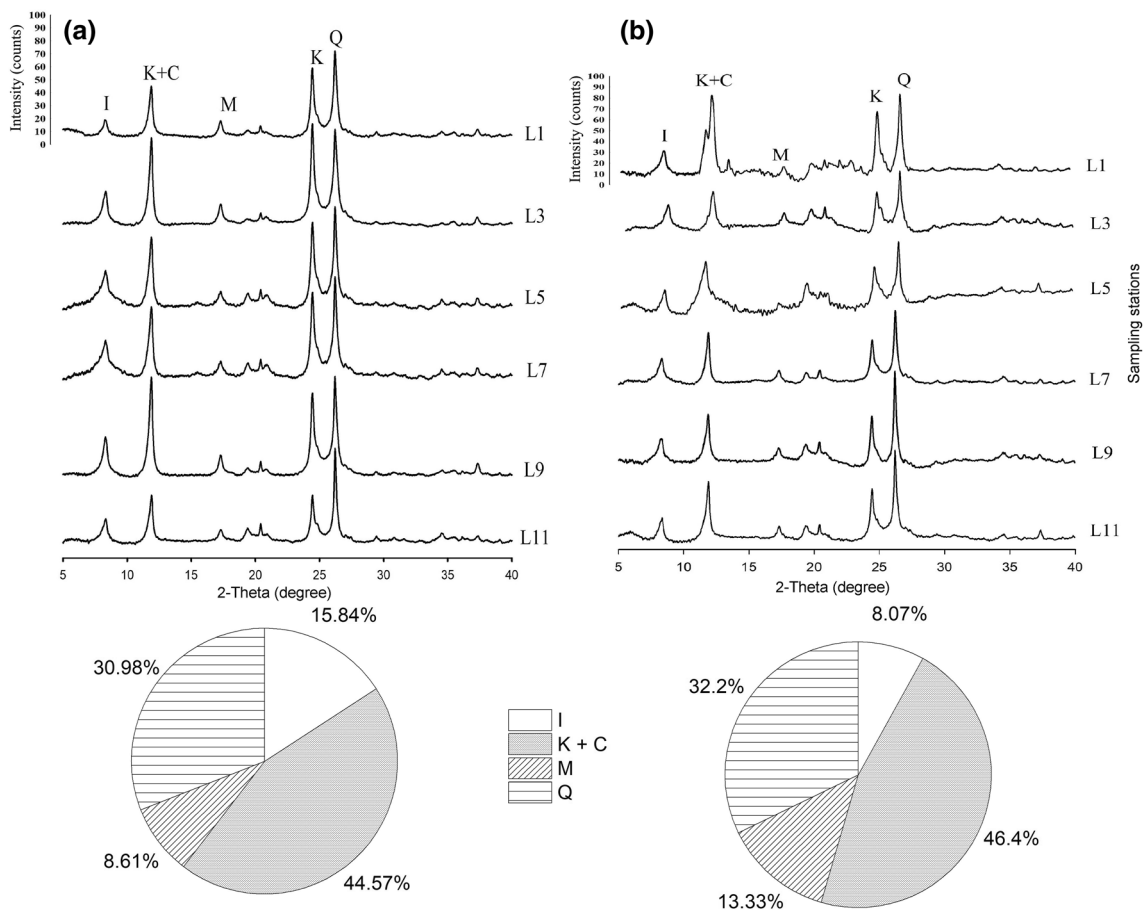




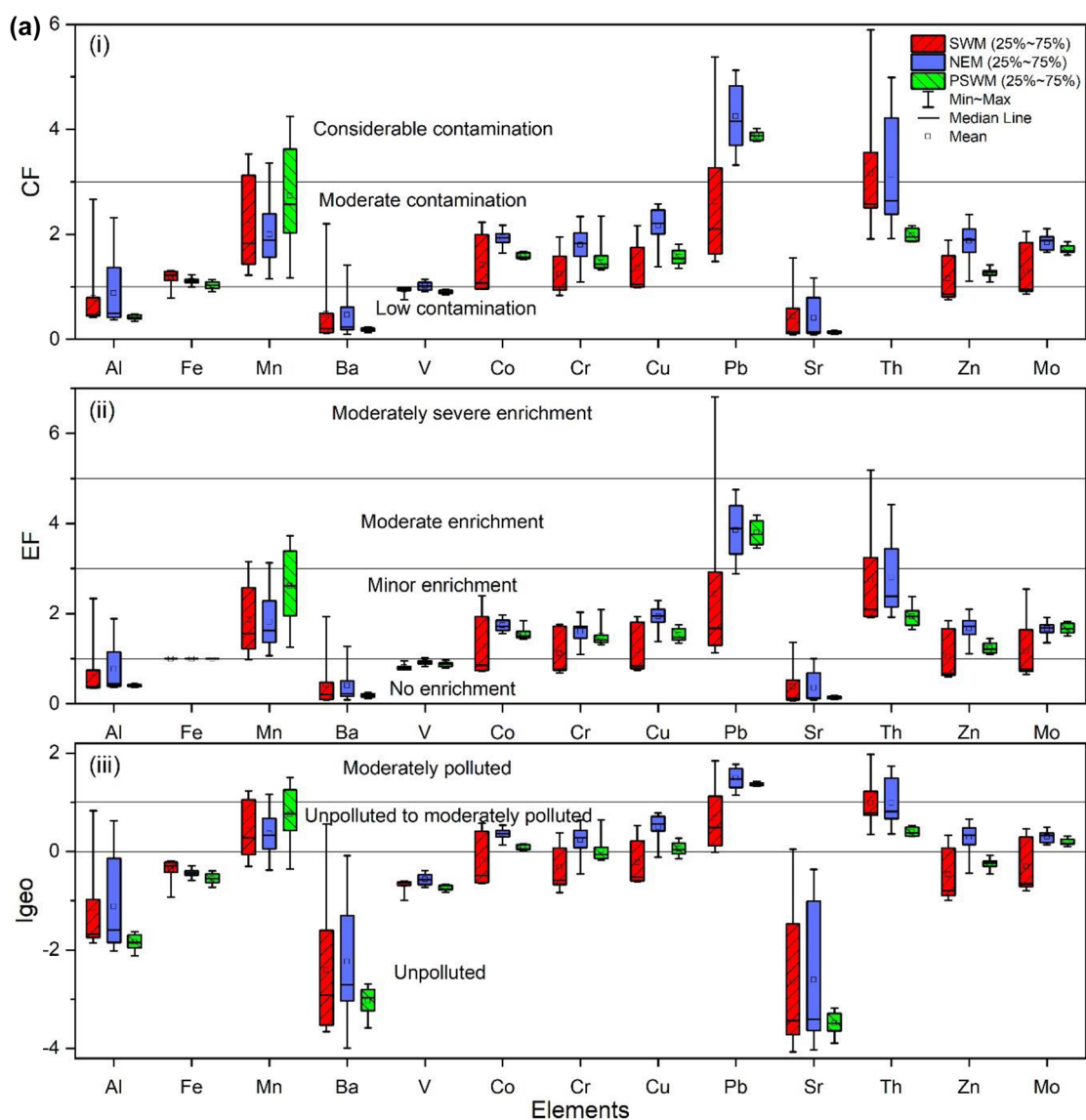
hydroxide, is formed by chemical weathering of aluminium silicate mineral like feldspar in the humid subtropical climate, whereas illite is an altered product of muscovite and feldspar in a temperate climate (Bonatti et al. 1973; Zabel et al. 2001). The weathering of feldspar and plagioclase rich rocks like anorthosites produces clay rich in kaolinite (Jeong and Kim 1993; Papoulis et al. 2004). Clay minerals of sediments from the low salinity region (NS) have higher percentages of kaolinite and chlorite, suggesting the dominance of chemical weathering in the source area (Fig. 3, Zachmann et al. 2009). Kaolinites are almost half of the clay minerals in the lake along with illite and montmorillonite (Fig. 3; Zachmann et al. 2009). It confirms the intense weathering process in the source region during SWM, which provides the major elements into the lake, and the sediments are transported mostly by the distributaries of the river Mahanadi (Sarkar et al. 1981; Bhattacharya et al. 1994).

### Distribution of trace elements

Trace element concentrations in lake sediments depend largely on the hydrological condition of the lake water and sediment influx into the lake, which affects the geochemical processes viz. dissolution, precipitation, redox reactions, adsorption, etc. (Jain et al. 2007; Zahra et al. 2014). The concentrations of Ba, Sr, and Th were high throughout the sampling stations during SWM, which gradually reduced with the decrease in freshwater influx during NEM and PSWM (Table 2). Ba is easily adsorbed on the metal oxides (Fe and Mn) and shows higher concentration during SWM (Charette et al. 2005; Charette and Sholkovitz 2006). Higher concentrations of Sr and Th are also reported in Mahanadi river-bed sediments (Konhauser et al. 1997). However, their relative immobility results in lesser concentrations in the lake sediments in comparison to the major elements (Braun and Pagel 1994; Seyfried et al. 1998). Higher Sr concentration indicates the intensification of chemical weathering as Sr is highly sensitive to the chemical weathering processes (Meyer et al. 2011). The strong significant positive



**Fig. 3** X-ray diffractometer peaks of clay minerals with their quantification during **a** southwest monsoon (SWM) and **b** northeast monsoon (NEM). *I* Illite, *K* kaolinite, *C* chlorite, *M* montmorillonite, *Q* quartz



**Fig. 4 a** Box plots for (i) contamination factor (CF), (ii) enrichment factor (EF) and (iii) geoaccumulation index ( $I_{geo}$ ) of the elements with their seasonal variability in northern sector (SWM southwest monsoon, NEM northeast monsoon, PSWM pre-southwest monsoon).

**b** Box plots for (i) contamination factor (CF), (ii) enrichment factor (EF) and (iii) geoaccumulation index ( $I_{geo}$ ) of the elements with their seasonal variability in outer channel (SWM southwest monsoon, NEM northeast monsoon, PSWM pre-southwest monsoon)

correlations among Sr, Th, and Ba (Appendix A) suggest their similar source and behavior towards chemical reaction and transportation (Zahra et al. 2014; Ganugapenta et al. 2018). The gradual change in Ba, Sr, and Th concentrations from rainy to pre-rainy periods may make them potential paleo-monsoon proxy. Higher concentrations of these elements during SWM are indicative of intensification of chemical weathering, which is related to higher monsoonal precipitation. In the sedimentary solid phase, Ba has already been established as a marine proxy in paleoceanography and may serve as a paleo-productivity tracer (McManus et al. 1998).

The remaining trace elements have higher concentrations during NEM suggesting high accumulation and precipitation rate. Higher anthropogenic activities in the lake could be the reason for the increasing element concentrations due to resuspension. The distributaries of the river Mahanadi carry these elements into the lake from the anthropogenic sources like mining and industrial wastes, domestic sewage, chemical effluents, agricultural pollutants, etc. (Banerjee et al. 2017). The highest enrichment of Cr during NEM (Table 2) is due to anthropogenic impacts as reported by earlier studies in Chilika lake (Panda et al. 2010; Banerjee et al. 2017) and also in other estuaries and estuarine lakes viz. Pulicat lake of India (Kamala-Kannan et al. 2008), Lake

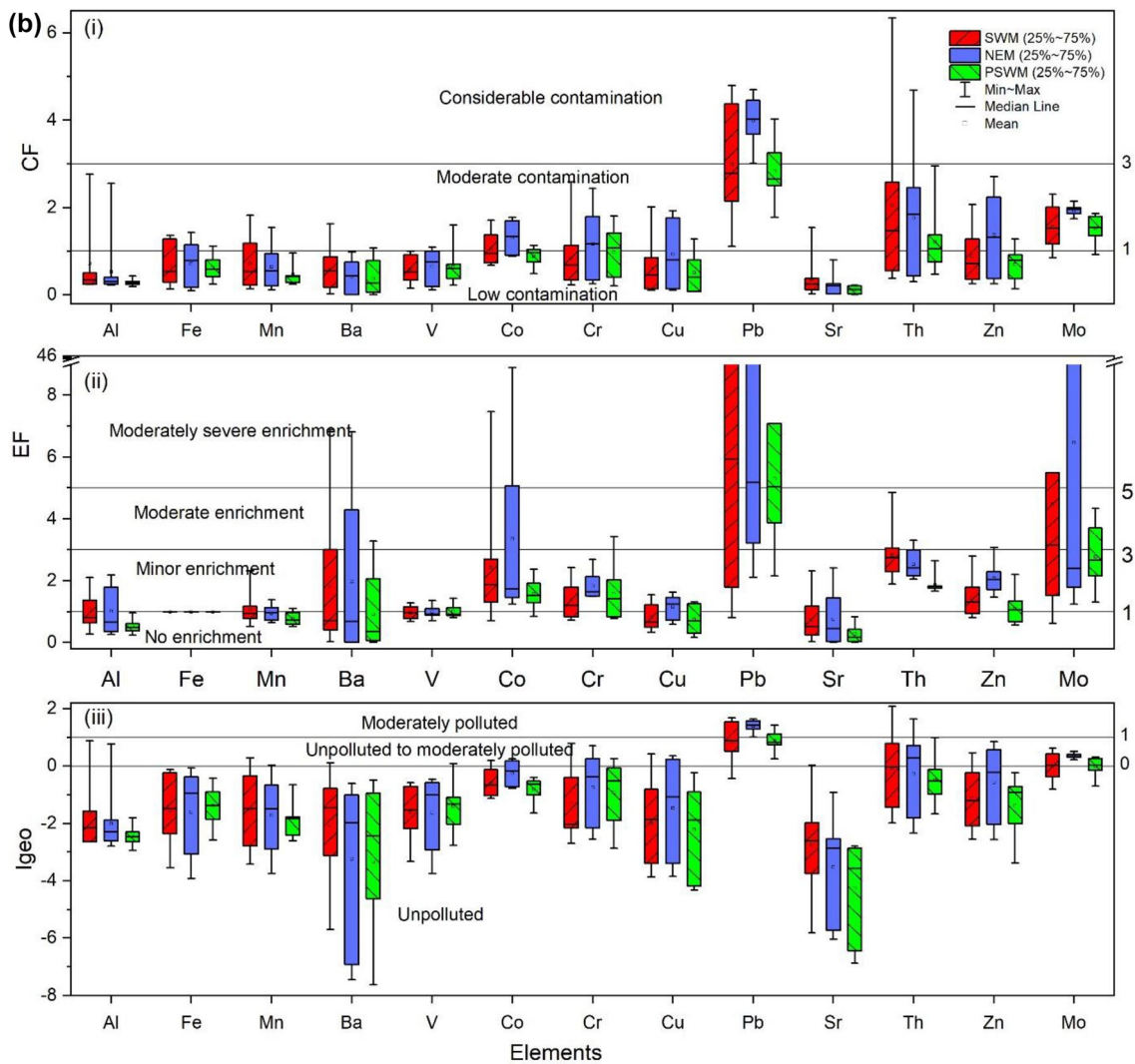


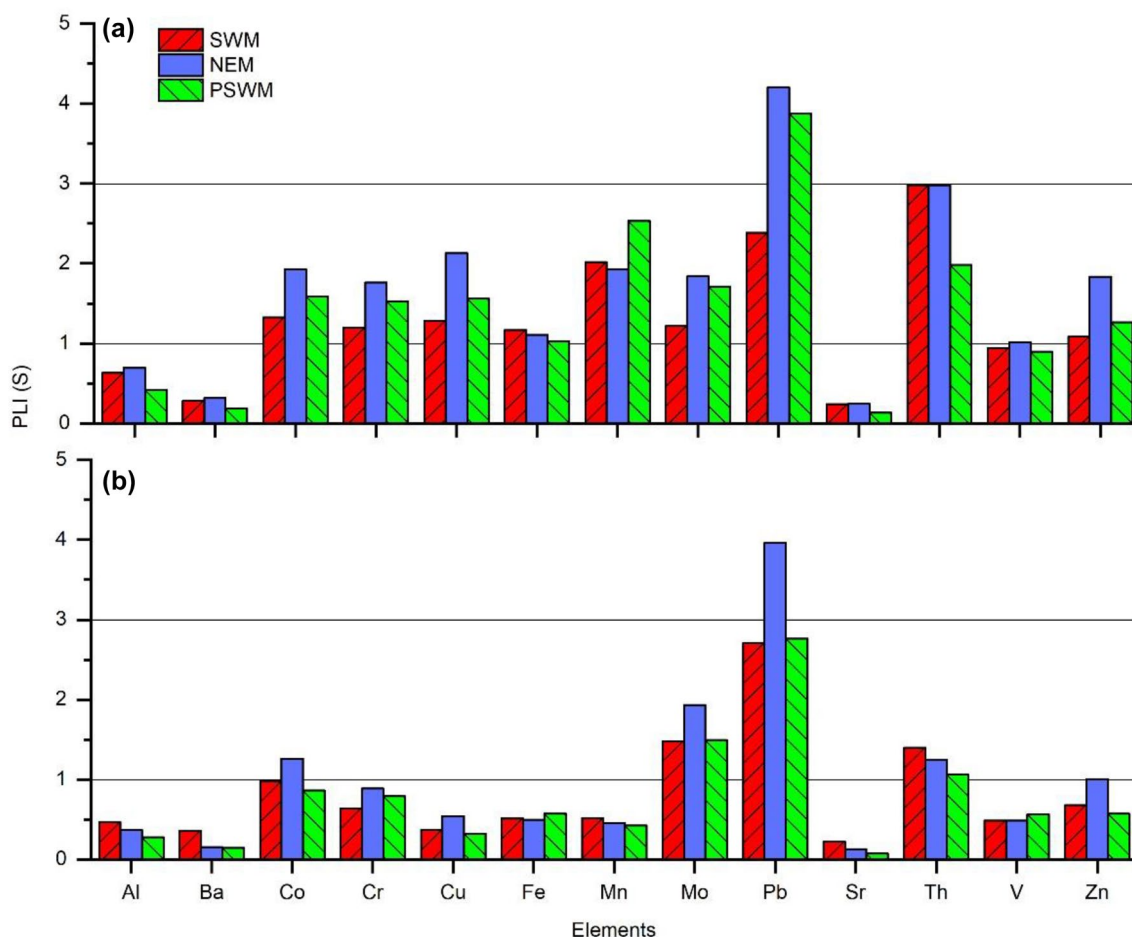
Fig. 4 (continued)

Sapanca of Turkey (Duman et al. 2007), Venice lagoon of Italy (Rigollet et al. 2004), etc. Frequent boating activities during NEM may also enhance the concentrations of some of the trace elements like Cu, Cr, Zn, Pb, etc. in the lake (Table 2, Barik et al. 2018; Panda et al. 2010). The decrease in water level during PSWM causes accumulation or precipitation of elements on the sediments in the land-locked lake (Gupta et al. 2009; Zahra et al. 2014). Interference of river runoff and seawater also provides a unique setting for the accumulation of elements in the Chilika lake sediments. Mo and V have a strong positive correlation (Appendix A), which could be due to their similar chemical and transportation behavior in the lake sediments. Probable anoxic condition of the lake during the non-rainy seasons (NEM & PSWM, Barik et al. 2019) causes their higher concentrations in the sediments through the diffusion process across the water–sediment interface (Table 2, Emerson and Husted,

1991). The concentrations of Cr and Cu are also reported to be higher in Chilika lake when compared with the Mahanadi river-bed sediments, while other trace elements, such as Zn, Co, and Pb do not show significant enrichment in the lake (Sundaray et al. 2011).

**Assessment of sediment quality**

The element concentrations were numerically evaluated with respect to SQGs for understanding the anthropogenic impacts on the biological community of the lake. The trace elements viz. Cr, Cu, Pb, and Zn are found exceeding the TEL and ERL values, which indicate that these elements have adverse effects on the biological community (Table 2; Sarkar 2018). Effect of Cr becomes frequent with the change of seasons as its concentration exceeds PEL and/or ERM values. Higher element retention and accumulation rate



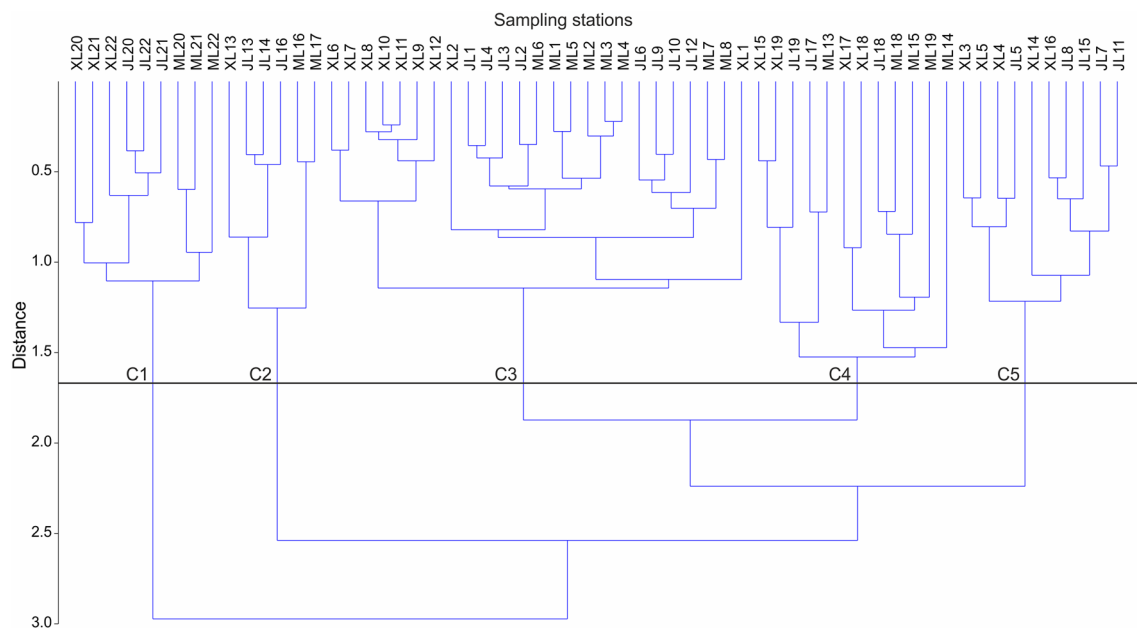
**Fig. 5** Pollution load index (PLI) of elements in **a** northern sector and **b** outer channel (SWM southwest monsoon, NEM northeast monsoon, PSWM pre-southwest monsoon)

imparts a more adverse effect on the biological community in NS than OC (Table 2). Further, various element contamination indices (CF,  $I_{geo}$ , EF, and PLI) have been utilized to assess the sediment quality of the lake. These indices are very useful in evaluating the changes in sediment quality at different environmental conditions as they use the background element concentrations of that region (Sarkar 2018). The distributions of CF, EF, and  $I_{geo}$  for all the elements during SWM, NEM, and PSWM are shown by Box and Whisker plots (Fig. 4).

Based on the CF values, there is moderate to considerable contaminations of Mn, Pb, and Th in NS and Pb in OC. The remaining elements have low to moderate contamination in both the sectors of the lake (Fig. 4a.i, b.i). All the elements have EF values less than 10 in NS, suggesting that they are mostly originated from lithogenic sources (Tuncel et al. 2007; Barbieri 2016). Pb has very high EF values (> 10) in OC, indicating its significant contribution from anthropogenic activities like frequent boating activity, antifouling

paints, leaked oils, etc. However, less enrichment for most of the elements in sediments with respect to crustal average values suggests that manmade inputs are insignificant in the lake (Fig. 4a.ii, b.ii). Based on the  $I_{geo}$  values, NS is found to be more contaminated than OC. The  $I_{geo}$  values for the elements viz. Co, Cr, Cu, Zn, Mn, and Mo range between classes 1 and 2 (uncontaminated to moderately contaminated), while for Pb and Th, it extends up to class 3 (strongly contaminated) in NS of the lake (Fig. 4a.iii). The elements Pb, Th, Zn, and Mo are marked as uncontaminated to moderately contaminated in OC of the lake (Fig. 4b.iii). The remaining elements are falling under class 1 (uncontaminated). The PLI values are found above 1 for all the elements, except for Al, Ba, Sr, and V in the low salinity region, while the elements viz. Pb, Th, and Mo have PLI values more than 1 in the high salinity region (Fig. 5). It also indicates high element contaminations in the low salinity region in comparison to the high salinity region.





**Fig. 6** Clustering of samples using paired group linkage method and Euclidean distance as a measure of dissimilarity. In the clustered samples, the 1st letter represents monitoring season [X for south-

west monsoon (SWM), J for northeast monsoon (NEM), and M for pre-southwest monsoon (PSWM)] and the second letter along with numerical represent the sampling stations as shown in Fig. 1

## Statistical analysis

Cluster analysis grouped the sampling stations of all three periods into five clusters (C1–C5) based on the similarities in element distributions associated with grain size distribution and condition of lake water (Fig. 6). The samples of high salinity region are mostly clustered into three groups (C1, C2, and C4). This suggests that there is a differential/dynamic distribution of elements with variable sediment and water conditions in the high salinity region. C1 comprises samples from stations located nearer to the sea mouth, and they have a gradual decrease in element concentrations with very high salinity and energy condition. The samples from the interior stations of OC away from the sea mouth are grouped in C2 and C4. C3 grouped the maximum number of samples, and all are from the low salinity region stations, which have the highest element concentrations in fine-grain sediments and medium energy condition. C5 grouped the samples collected during all three seasons from both the low and high salinity regions stations having a mixed environment.

Further, to assess the environment of each group of samples, principal component analysis (PCA) was carried out that extracted four principal components (PC1, PC2, PC3, and PC4) explaining 87.9% of cumulative data variance (Appendix B). PC1 having an eigenvalue of 11.1 describes maximum data variance (46.22%) and has strong positive

loadings of silt, clay, Al, Fe, Mn, Li, V, Co, Cr, Cu, Th, Zn and strong negative loading of sand (Fig. 7). This component explains the accumulation and remobilization of the above elements, which are mostly controlled by the grain size distribution pattern and salinity of the lake. The strong affinity of these elements towards fine-grained silty or clayey fractions and organic matter causes their adsorption or precipitation in the alkaline lake water. The PCA biplot also shows that PC1 is closely associated with the samples grouped in C1 and C3 (Fig. 7a, b). C1 samples are explained by the negative loadings of sand and EC, while C3 samples are characterized by high concentrations of the elements associated with PC1. PC2 with the positive factor loadings of Ca, K, Ba, Sr, Al, and Mg describes 19.1% of data variance. These elements are mostly derived from the secondary erosion and deposition of minerals like feldspar and micas, which are easily leachable by chemical weathering. Ba and K are included in this component because of their similar geochemical behavior (Middelburg et al. 1988). This component reaffirms the lithogeny source of the elements coming with the river influx from the Mahanadi distributaries, and they are controlled by the weathering of parent rock from in the catchment area and explains C5 group of samples (Fig. 7a, b). PC3 describes 13.9% of data variance and includes the elements Mo and Pb, which are mostly controlled by anthropogenic activities. C4 samples, located away from the sea mouth, are under the influence of



content, and trace elements viz. Ba, Sr, and Th can be used as a potential proxy to reconstruct paleo-monsoon. Further, this study can serve as baseline data for paleoclimate and paleoenvironment studies. It can be compared globally with similar environments for a better understanding of transitional environment processes of the present and the past.

The trace elements viz. Cr, Cu, Pb, and Zn exceeds the SQGs and have adverse effects on the biological community living the lake. The convergence of the Mahanadi distributaries in NS causes more accumulation of elements than OC of the lake. Most of the trace elements show moderate to considerable contaminations in NS, while Pb is the main contaminant in OC due to its significant contribution by anthropogenic activities. The combined effects of element contaminations are also higher in NS than OC of the lake. The anthropogenic activities need regular monitoring in order to develop a healthy lake environment for the biological community. Further, the development of multiple channels from the lake to the sea could help in reducing or balancing the element concentrations in the lake by discharging them into the sea.

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